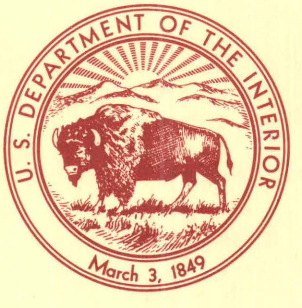
Field Studies of Radon in Rocks, Soils, and Water

U.S. GEOLOGICAL SURVEY BULLETIN 1971



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Field Studies of Radon in Rocks, Soils, and Water

LINDA C.S. GUNDERSEN and RICHARD B. WANTY,
Editors

U.S. Geological Survey research on the geology, geophysics,
and geochemistry of radon in rocks, soils, and water

U.S. GEOLOGICAL SURVEY BULLETIN 1971

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PREFACE

This volume contains 24 papers of geologic and hydrogeologic research on radon-222, a naturally occurring radioactive gas (hereafter referred to as radon). In 1984, R.C. Smith II and co-workers documented the most severe indoor radon problem to date, which was in Colebrookdale Township near Boyertown, Pa. (Pennsylvania Geology, 1987, v. 18, p. 1–7). A residence was found to contain 2,500 pCi/L (picocuries per liter) of radon. The Environmental Protection Agency's current action level for radon mitigation of a home is 4 pCi/L. At 4 pCi/L, the chance of lung cancer from a lifetime exposure, for populations of smokers and nonsmokers, is 1 to 3 persons out of 100. The discovery of high indoor radon levels near Boyertown and in several other parts of the United States prompted nationwide concern and the initiation of federal programs to assess and understand the problem. Despite the obvious geologic controls on concentrations of radon in soil gases, relatively little research has focused on geologic aspects of the radon problem. These factors are important for our understanding of the natural occurrence of radon in soil gases and for the eventual prediction of areas likely to have high radon concentrations. Radon, in addition to being found in soil gas, can also be present in ground water. A ground-water supply enriched in radon is possibly a significant source of radon in indoor air. Inside a home, when water is agitated, such as when a faucet is turned on, radon can be released from the water into the air. Uranium and radium may also be present in ground waters in concentrations that may prove harmful for human consumption. High amounts of radium and radon have been known for several years to exist in ground waters in New England, but their occurrence in domestic well supplies in other parts of the country had not been tested extensively or understood. Research in the past 5 years by the U.S. Geological Survey on these geological and hydrogeological issues has significantly increased our understanding of radon and its occurrence. Compiled in this volume are field studies relating ways to measure, assess, and understand radon in homes and in water. The papers are of a technical nature and are written for the scientific audience.

Radon-222 is generated from the decay of uranium-238, which is present naturally in varying concentrations in all rocks and soils. Radon-222 is the longest lived isotope of radon; it is odorless and colorless in the concentrations observed in homes and so is not an obvious problem in the air in homes. Radium-226 is the immediate parent isotope of radon-222. Figure 73 on page 184 illustrates the uranium-238 decay chain. The formation of radon-222 and its daughter products, polonium-218 and polonium-214, is shown. Radon in air can be inhaled into the lungs, and the polonium isotopes can be deposited in lung tissue as the radon decays. Polonium atoms adhere to particulates in air, which when breathed in, may also deposit them in the lungs. The polonium isotopes emit alpha particles, which cause lung damage and may be responsible for lung-cancer deaths of 5,000 to 20,000 U.S. citizens a year (A Citizen's Guide to Radon, Environmental Protection Agency, 1986, OPA–86–004, 14 p.).

The first half of this volume focuses on rocks and soils, the principle sources of indoor radon. This half is divided into three sections, the first of which addresses ways of measuring radon in the ground and also the problems associated with measurement. Tanner describes "Methods of Characterization of Ground for Assessment of Indoor Radon Potential at a Site." In this report, soil characteristics and their effect on radon mobility and measurement are discussed in detail. Recommendations for avoiding inaccurate measurements and suggestions for the best type of measurement instruments are made. "Simple Techniques for Soil-Gas and Water Sampling for Radon Analyses," by Reimer, illustrates a simple sampling technique for soil air and for radon in ground water that is accessible to most scientists interested in reconnaissance and field techniques of measuring radon. In a paper at the end of this section, Reimer discusses "Derivation of Radon Migration Rates in the Surficial Environment by Use of Helium Injection Experiments." In this study he uses helium injection into the ground to determine radon migration rates and direction within deformed bedrock. A long-term study on the effects of climate on soil radon is presented in "A Preliminary Evaluation of Environmental Factors Influencing Day-to-Day and Seasonal Soil-Gas Radon Concentrations," by Asher-Bolinder, Owen, and Schumann. They found that pedologic and meteorologic conditions interact to cause both day-to-day and seasonal variations in soil radon.

The second section, which concerns geologic field studies of radon, presents specific case histories. "Radon in Sheared Metamorphic and Igneous Rocks," by Gundersen, describes four areas of the Appalachians, including the well known Boyertown site, where severely elevated levels of indoor radon are found associated with sheared bedrock. The paper analyzes the geologic processes that contribute to some of the highest indoor radon levels found in the United States. Agard and Gundersen focus on "The Geology and Geochemistry of Soils in Boyertown and Easton, Pennsylvania." Soil characteristics are examined in two areas having very high indoor radon levels to understand which soil parameters control radon migration and concentration. Henry, Kaeding, and Monteverde present "Radon in Soil Gas and Gamma-Ray Activity of Rocks and Soils at the Mulligan Quarry, Clinton, New Jersey." Soil radon measurements and gamma measurements are compared for an area of carbonate rocks that are sheared and mineralized with uranium. A practical demonstration of radon use in predicting another geologic hazard is given in "Radon in Soil Gas Along Active Faults in Central California," by King, Walkingstick, and Basler. Soil radon was monitored from 1975 to 1985 along a 380-km segment of the San Andreas fault, and correlations with large local earthquakes are shown. "Radon Emanation from Uranium Mill Tailings," by Landa, examines the radon emanation of mine tailings with reference to the original ore. He finds that, although 90 percent of the uranium is generally extracted during the milling processes, most of the daughter-product activity remains with the tailings.

The third section highlights different techniques for assessment of radon potential. Duval describes the "Use of Aerial Gamma-Ray Data to Estimate Relative Amounts of Radon in Soil Gas." He shows how aerial gamma-ray data can be used to assess regions for radon potential and the importance of geology in utilizing the geophysical data. "Regional Radon Characterizations," by Peake and Schumann, demonstrates how indoor radon screening data can be combined predictively with soils, geologic, and radiometric data to indicate areas of high radon potential. Kentucky is used as an example of a State assessment. The final paper of the first half, by Reimer, Gundersen, Szarzi, and Been, describes a "Reconnaissance Approach to Using Geology and Soil-Gas Radon Concentrations for Making Rapid and Preliminary Estimates of Indoor Radon Potential." Simple field techniques for assessment of radon potential are evaluated by use of soil radon collected along a transect perpendicular to strike in the Maryland coastal plain.

The second half of the Bulletin, "Studies of Radon in Water," features geochemical and hydrologic studies of ground waters. The first paper in the introductory section, "A Review of the Chemical Processes Affecting the Mobility of Radionuclides in Natural Waters, with Applications," by Wanty and Schoen, provides a primer on the aqueous geochemistry of radionuclides in the context of geology and the radon problem. This paper also gives a review of the health effects of radionuclides in water. In the second introductory paper, Paulsen presents an extensive overview in "Radionuclides in Ground Water, Rock, Soil, and Indoor Air of the Northeastern United States and Southeastern Canada—A Literature Review and Summary of Data." The report briefly summarizes major investigations of the occurrence of radionuclides in the New England States, New York, and southern Ontario since the 1950's.

The second section addresses techniques for measuring radon in ground waters. Yang describes "Sampling and Analyses of Dissolved Radon-222 in Water by the De-Emanation Method." In this paper he discusses the advantages and disadvantages of the direct de-emanation and alpha-scintillation counting method and finds it to have better precision in the lower concentration ranges than any other method currently available. "A Comparison of Two Techniques for Radon-222 Measurement in Water Samples" is given by Mullin and Wanty. In testing the reproducibility and values of radon concentration measured by liquid scintillation and by a technique developed by Reimer (this volume), the authors found the results to be in moderately good agreement. Although it has the advantage of producing results within a matter of minutes, the Reimer technique shows poor consistence in comparison with the liquid scintillation method because in the Reimer technique radon is lost if samples are stored for a prolonged period.

The third section contains eight papers featuring specific field studies of radon in surface and ground waters throughout the United States. Lee and Hollyday present "Use of Radon Measurements in Carters Creek, Maury County, Tennessee, To Determine Location and Magnitude of Ground-Water Seepage." By measuring the activity of radon-222 in streamflow, the authors found the location and calculated the contribution of ground-water seepage to the total flow in the creek. Szabo and Zapecza examine "Geologic and Geochemical Factors Controlling Uranium, Radium-226, and Radon-222 in Ground Water, Newark Basin, New Jersey." They found that ground-water radionuclide levels in the Newark basin are highest in the Passaic, Lockatong, and Stockton Formations. A number of samples collected exceed the Environmental Protection Agency's maximum contaminant levels and are associated with uranium enrichment in the bedrock. In the report "Radium-226, Radium-228, and Radon-222 in Ground Water in the Chickies Quartzite, Southeastern Pennsylvania," by Cecil, Senior, and Vogel, an anomalously elevated occurrence of radium in domestic well water is investigated. They found that pH may be used to predict elevated radium concentrations and that all samples with pH below 4.7 contained concentrations of total radium that exceeded the Environmental Protection Agency's maximum contaminant level. Their paper also documents the importance of measuring all isotopes of radium as opposed to the more common practice of ignoring radium-228 unless radium-226 is above certain threshold levels. "Radon in Ground Water of the Carson Valley, West-Central Nevada" is the subject of the study by Lico and Rowe. The authors have determined both the sedimentologic and geochemical controls on the highly variable distribution of radon-222 within the valley. However, they believe the principle source of the radon is derived from the Carson Range. High levels of radon in domestic wells in Boyertown and eastern Pennsylvania is the subject of the paper "Geochemistry of Ground Water and Radionuclide Mobility in Two Areas of the Reading Prong, Eastern Pennsylvania," by Wanty, Briggs, and Gundersen. The authors found that aquifer rock type appears to be the dominant factor determining the radon-222 potential of ground water in the two sites examined. Gray and Webb describe "Radionuclides in the Puerco and Lower Little Colorado River Basins, New Mexico and Arizona, Before 1987." Both naturally occurring radionuclides and radionuclides from uranium mine tailings contribute to excessive levels of radioactivity in surface and ground waters in the river basins examined. In "Uranium, Radium, and Radon in Deeply Buried Sediments of the U.S. Gulf Coast," Kraemer describes the geochemistry and secular equilibrium of uranium, radium, and radon in water, rock, and gases of buried coastal plain sediments. The final paper of the Bulletin is "Radium, Helium, and Other Gases in Shallow Ground Waters of Uraniferous Holocene Alluvium, Flodelle Creek, Stevens

County, Northeast Washington," by Otton and Reimer. The authors use various gas species including radon, helium, oxygen, carbon dioxide, and methane to examine radionuclide source, ground-water flow, and geochemistry in a drainage basin.

The editors would like to especially thank John Watson for his diligence, humor, patience, and excellent technical editing during the compilation of this Bulletin.

Linda C.S. Gundersen
Richard B. Wanty

CONTENTS

Part 1: Studies of Radon in Rocks and Soil
Section 1: Techniques for Measuring Radon in Soil

Section 2: Geologic Field Studies

Section 3: Estimating Radon Potential

Part 2: Studies of Radon in Water
Section 1: Introduction

Section 2: Techniques for Measuring Radon in Water

Section 3: Field Studies of Radon in Surface and Ground Waters

Radon, Helium, and Other Gases in Shallow Ground Waters of Uraniferous Holocene Alluvium, Flodelle Creek, Stevens County, Northeast Washington, *by* James K. Otton *and* G.M. Reimer **319**

FIGURES

TABLES

METRIC CONVERSION FACTORS

For readers who wish to convert measurements from the inch-pound system of units to the metric system of units, the conversion factors are listed below:

Multiply	By	To obtain
	Length	
micrometer (µm)	0.000 039 37	inch (in)
millimeter (mm)	0.039 37	inch (in)
centimeter (cm)	0.393 7	inch (in)
meter (m)	3.281	foot (ft)
	1.094	yard (yd)
kilometer (km)	0.621 4	mile (mi)
	Area	
centimeter^{2} (cm^{2})	0.155 0	inch^{2} (in^{2})
meter^{2} (m^{2})	10.76	foot^{2} (ft^{2})
	1.196	yard^{2} (yd^{2})
	0.000 247 1	acre
hectometer^{2} (hm^{2})	2.471	acre
kilometer^{2} (km^{2})	0.386 1	mile^{2} (mi^{2})
	Volume	
centimeter^{3} (cm^{3})	0.061 02	inch^{3} (in^{3})
milliliter (mL)	0.061 02	inch^{3} (in^{3})
liter (L)	61.02	inch^{3} (in^{3})
	0.035 31	foot^{3} (ft^{3})
	33.82	ounce, fluid (oz)
	2.113	pint (pt)
	1.057	quart (qt)
	0.264 2	gallon (gal)
meter^{3} (m^{3})	35.31	foot^{3} (ft^{3})
	1.308	yard^{3} (yd^{3})
	264.2	gallon (gal)
	0.000 810 7	acre-foot (acre-ft)
kilometer^{3} (km^{3})	0.239 9	mile^{3} (mi^{3})
	Volume per unit time (includes flow)	
gram per minute (g/min)	0.035 27	ounce (avoirdupois) per minute (oz/min)
milliliter per minute (mL/min)	0.033 82	ounce (fluid) per minute (oz/min)
	0.035 31	foot^{3} per second (ft^{3}/s)
liter per second (L/s)	15.85	gallon per minute (gal/min)
meter per second (m/s)	3.281	foot per second (ft/s)
meter per day (m/d)	3.281	foot per day (ft/d)
meter^{2} per day (m^{2}/d)	10.76	foot^{2} per day (ft^{2}/d)

Multiply	By	To obtain
	Volume per unit time (includes flow)—Continued	
meter^{3} per second (m^{3}/s)	35.31	foot^{3} per second (ft^{3}/s)
	15 850	gallon per minute (gal/min)
	Mass	
microgram (µg)	0.000 015 43	grain (gr)
gram (g)	0.035 27	ounce, avoirdupois (oz avdp)
kilogram (kg)	0.002 205	pound, avoirdupois (lb avdp)
	Mass per unit volume	
microgram per liter (µg/L)	0.000 058 41	grain per gallon (gr/gal)
milligram per liter (mg/L)	0.058 41	grain per gallon (gr/gal)
	Temperature	
	degree Kelvin=degree Celcius+273.15	
degree Celsius (°C)	Temp °F=1.8 temp °C+32	degree Fahrenheit (°F)
	Specific conductance	
microsiemens per centimeter at 25 degrees Celsius (µS/cm at 25 °C)	1.000	micromho per centimeter at 25 degrees Celsius (µmho/cm at 25 °C)
millisiemens per meter at 25 degrees Celsius (mS/m at 25 °C)	1.000	millimho per meter at 25 degrees Celsius (mmho/m at 25 °C)

Activity

1 becquerel (Bq)=1 disintegration per second
1 Curie (Ci)=3.7×10^{10} Bq
1 picocurie (pCi)=0.037 Bq

Radon concentration

1 Bq/m^{3}=2.7×10^{-2} pCi/L
1 pCi/L=37 Bq/m^{3}

Radon progeny concentration

Working level (WL)=7.4×10^{3} Bq/m^{3}
=2×10^{2} pCi/L

ALTITUDE DATUM

National Geodetic Vertical Datum of 1929 (NGVD of 1929): A geodetic datum derived from a general adjustment of the first-order level nets of both the United States and Canada, formerly called Sea Level Datum of 1929.

PART 1: STUDIES OF RADON IN ROCKS AND SOIL

SECTION 1: TECHNIQUES FOR MEASURING RADON IN SOIL

METHODS OF CHARACTERIZATION OF GROUND FOR ASSESSMENT OF INDOOR RADON POTENTIAL AT A SITE

Allan B. Tanner

Abstract

The concentration and mobility of radon-222 in soil interstices control the potential of ground to supply radon to a structure. Radon mobility depends on its diffusivity and on the permeability of the ground to flow of soil gas. Several soil-probe methods currently under development can be used to measure the radon concentration in soil gas and to obtain gas-flow characteristics from which the gas permeability of the ground can be inferred. The radon diffusion coefficient can be inferred from measurements of the porosity and pore water fraction of samples of soil removed for emplacement of the soil probe. General principles of soil development and tests made with a special soil probe indicate that layered soils are more permeable to gas flow in the horizontal than in the vertical direction, although cracking in soils can cause high vertical permeability along the cracks. Soil cracks and rock fracture systems pose problems of test-probe spacing and modeling that have not been resolved. Changes in the degree of pore water saturation can affect the radon concentration in the gas phase by as much as a factor of 4 and radon mobility by as much as 3 orders of magnitude. In order to lessen the effects of short-term meteorological and seasonal changes on characterization of the ground, soil-probe measurements should be made as deeply as is feasible, ideally to the depth of the slab of a building to be constructed at the site. For slab-on-grade and crawl-space house sites, the soil may be best characterized by means of soil-gas accumulators operated with a degree of suction comparable with that exerted by a house during the season in which the house is heated.

INTRODUCTION

Testing of the ground in order to determine its indoor radon potential for an existing structure is more expensive than a straightforward test of the radon concentration in the structure. Ground tests may not be economical in the preconstruction phase for single structures of typical design if the ground has normal or moderately elevated indoor radon potential, because the cost of testing may approach that of incorporating aids to radon mitigation into the structure. The cost of constructing a new house so as to facilitate mitigation may range from \$231 to \$1,381 (John Spears, National Association of Home Builders, written commun., 1988). For structures of designs incompatible with the usual mitigation techniques, for large or extended structures, and for developments containing large numbers of homes, preconstruction testing may allow the builder to avoid unnecessary provision for radon mitigation or may indicate that extraordinary measures must be taken to avoid indoor radon problems. Although developers and home builders frequently ask consultants and site engineers to determine the indoor radon potential of sites, no such test has yet been proven reliable. Problems in developing a standard test include accounting for spatial variations of radon's soil-gas concentration and mobility on a meter-to-meter scale and temporal variations of the same factors caused by freezing or variable water content of the soil. Also, no agreement has been reached on the suitability of measurement techniques.

The emanation of radon from rock and soil minerals and migration of radon in the ground have been reviewed by Tanner (1964, 1980); references more specific to the ground as a source of indoor radon were given by Åkerblom and others (1984), Nero and Nazaroff (1984), Eaton and Scott (1984), Nero (1985), Tanner (1986), and Nazaroff and others (1988). Research on site-specific measurements to assess indoor radon potential is active in Scandinavia, in the United States, and probably in several other countries, but little has appeared in the literature about quantitative approaches. Åkerblom (1986) gave Swedish criteria for classifying radon risk based on radon concentrations in soil gas: less than 10 kBq/m^3 (kilobecquerels per cubic meter) (270 pCi/L, picocuries per liter) is classed as "low risk"; greater than 50 kBq/m^3 (1,350 pCi/L) is classed as "high risk"; and between 10 and 50 kBq/m^3 is classed as "normal risk." He noted, however, that low permeability of the soil (high resistance to flow of the soil gas) works to shift the risk classification to the next lower level; conversely, high permeability works to shift the risk classification to the next higher level. Measurements of radon in soil gas are used in many countries; see, for example, Hesselbom (1985), Lindmark and Rosen (1985), Reimer (1988), and other papers in this Bulletin. In Finland, a procedure involving laboratory measurements of radon production and diffusion

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and permeability to air flow in soil samples is under development for ground classification (Slunga and Holkko, 1987). The flux of radon obtained by suction at the ground surface is the basis of an investigatory method in Denmark (Kaare Ulbak, Statens Institut for Strålehygiene, Brønshøj, oral commun., 1986). In New York State, Kothari and others (1985) have made extensive use of a soil probe to extract soil gas for radon analysis and to determine simultaneously the permeability of the ground at the sampling depth, following a scheme devised by Scott (DSMA Atcon, Ltd., 1983). Similar apparatus are used by the Lawrence Berkeley Laboratory (R.G. Sextro, oral communication, 1987), by the Environmental Protection Agency (R.T. Peake, written communication, 1988), and by the U.S. Geological Survey (Tanner, 1988), but the data are used differently. Kunz (1988) used an equation relating a "radon index number" to radon concentration in the soil gas, depth to the water table or bedrock, and soil permeability. The Environmental Protection Agency approach (Peake, 1988) is similar to that of Åkerblom (1986). Nazaroff (1988) modeled radon entry into a basement structure on the basis of the permeability of the ground outside the foundation and radon entry at the level of the basement slab. Tanner (1988) proposed a protocol, described below, that applies values of molecular diffusivity and permeability obtained from in situ sampling to a model of one-dimensional radon migration. All these approaches assume that the ground is homogeneous and isotropic (that is, that radon diffusion or soil-gas flow proceeds with equal ease in all directions), although inhomogeneity and anisotropy of permeability are known to be common, as discussed below. It is also assumed that measurements made at a particular time are either representative of the site or may be corrected to be representative, although significant variation is to be expected because of precipitation, seasonal temperature changes, and barometric pressure fluctuations. This report is intended to examine these variables as they affect in situ measurements in general and the soil-probe methods in particular.

THE EMANATION PROCESS

Only a fraction of the radon that is generated by decay of radium-226 (^{226}Ra) in rock and soil escapes from solid material and into the rock or soil pores. This fraction was termed "emanating power" about 1900 (Rutherford, 1901), but in the past two decades different names have proliferated, the most common name being "emanation coefficient." The emanation process was discussed by Tanner (1964, 1980) from concepts and evidence of many investigators. In packed aggregates of mineral grains, emanating power is usually of the order of 1 percent unless water or other liquid is present in the smaller interstices, fractures, and capillaries to absorb the kinetic energy of radon ions recoiling from the radium alpha-particle decay. The water content of most soils is sufficient to make the emanating power much greater than 1 percent; Rogers and Nielson (1988) reported a mean value of 22 percent for 56 western soils. The siting of the radium atoms in rock and soil is also very important in determining how readily the radon atoms can escape upon radium decay. The emanating powers typically reported for soils are consistent with the siting of significant fractions of the radium on grain surfaces and within secondary coatings on the grains. Both the development of external and internal surface areas of the mineral grains and the deposition of radium on surfaces and within surface coatings proceed with biological and chemical activity that attacks the grains; the fraction of radium exposed to the pores may correlate to some degree with the age of a soil.

Most of the site-specific methods of assessing indoor radon potential measure the radon concentration of soil gas rather than the total radium content of the rock or soil; emanating power determinations are then unnecessary. However, for assessments that use gamma radiation or chemical uranium measurements to gauge the inherent radioactivity of the ground, emanating power needs to be evaluated.

PERMEABILITY

Soil particles tend to be oriented with their longest dimensions in the horizontal plane. Nazaroff and others (1988) cited works by Rice and others (1970) and by Bowles (1979) that report markedly greater permeability in the horizontal than in the vertical direction, particularly for fine-grained soils having significant fractions of clay minerals. Bowles (1979, p. 212–213) stated that the horizontal permeability is commonly on the order of 10 to 1,000 times greater than the vertical permeability. This anisotropic permeability can be evaluated to some extent from measurements made with the soil probe shown in figure 1. The three inflated rubber packers create three isolated annular spaces in the borehole. If suction is applied to the middle space only, soil gas in an uncracked soil flows into the middle space along the radii of a sphere. If suction is applied to the upper and middle spaces, flow is along radii of a hemisphere that has its convex surface extending downward. If suction is applied to the middle and lower spaces, flow is along radii of a hemisphere that has its convex surface extending upward. If suction is applied to all three spaces, the flow into the middle space is along radii of a cylinder of height equal to the spacing of the packers. These flow patterns are, of course, only approximate because of the interference of the probe itself and because the soil-gas pressures ultimately are asymmetrically referred to the ground surface rather than to a spherical surface at distance from the sampling probe. However, the flow paths near the

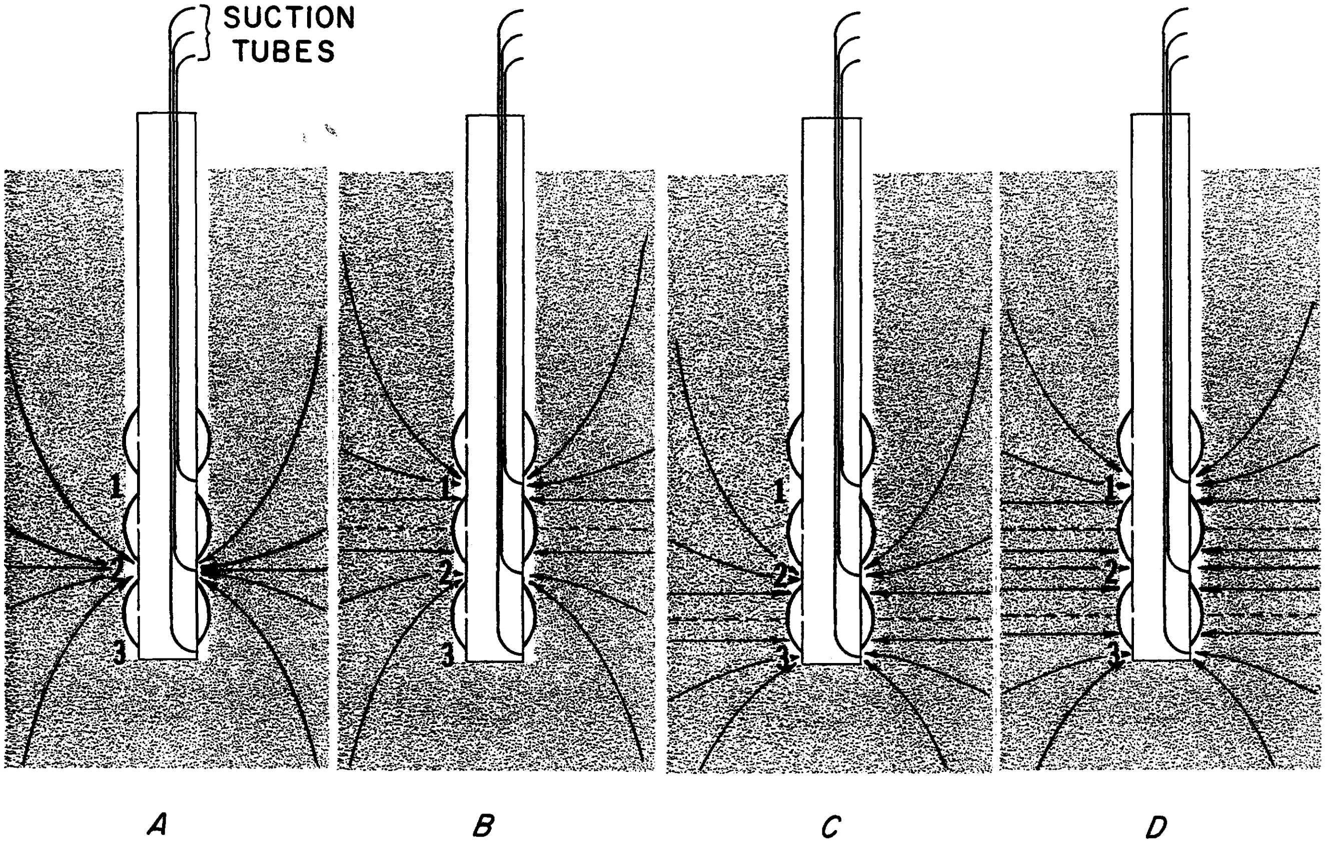

Figure 1. Modes of operation of three-packer soil probe. (Tubes for inflation of the packers and for pressure monitoring are not shown.) Atmospheric absolute pressure p_a of approximately 101 kPa (kilopascals) is assumed to prevail initially in the soil. The flow patterns can be controlled by reducing the absolute pressures by small amounts (less than 5 kPa) in the annular spaces created by the inflated rubber packers. *A*, No suction is exerted on spaces 1 and 3. The flow into space 2 is approximately spherical (along radii of a sphere centered at space 2). *B*, Suction is exerted on spaces 1 and 2. The flow into space 2 is from the hemisphere whose convex surface is downward. *C*, Suction is exerted on spaces 2 and 3. The flow into space 2 is from the hemisphere whose convex surface is upward. *D*, Suction is exerted on spaces 1, 2, and 3. The flow into space 2 is along radii of an annular cylinder coaxial with the borehole. Near the sampling probe, the flow lines deviate from radial because of the interference width of the probe. After prolonged suction, the flow lines must be distorted toward the ground surface, because the ultimate source of air is the atmosphere; however, the indicated flow paths should obtain for a typical measurement period of several minutes. Anisotropic permeability affects both the flow lines and the proportions of soil gas entering space 2 from different horizons.

probe have been calculated by Nazaroff (1988, fig. 2) to satisfy the spherical flow pattern approximately. In many soils of high clay content, I found changing from the downward-hemispherical mode of operation to the spherical mode resulted in a small to unmeasurable increase of flow into the middle space. The implication is that vertical flow from above was negligible and that the flow was predominantly horizontal or from the downward hemisphere. It has not been determined whether the apparent permeability derived from measurements in spherical or hemispherical geometry or the permeability derived from measurements with a controlled cylindrical geometry gives a more appropriate value of permeability to be used in a test to evaluate the total soil-gas movement to a structure's foundation (see appendixes B and C).

Constructing a building that has a full or partial basement usually involves excavating, laying down coarse aggregate to break capillary action between the ground and the basement slab and to promote drainage under the slab, and backfilling the gap, typically about 0.5 m (meter) wide, between the sides of the excavation and the basement walls. The coarse aggregate is much more permeable, and the backfill material may be significantly more permeable than the undisturbed ground. The voids in this backfill-subslab aggregate zone may constitute a reservoir, or plenum, for radon that emanates from the material in the zone and for radon that can migrate into the zone from the undisturbed ground beyond it. Once in the zone, radon-bearing soil gas may move through available entry points in response to reduced pressure in the building. Various investigators have noted increased radon entry with a decrease in indoor atmospheric pressure. The decrease may be that accompanying a decrease of outdoor barometric pressure and may cause the greater soil-gas pressure under a house to be relieved by flow into the house (Schmid, 1932), or indoor pressure may by various means be reduced relative to the outdoors, with the same result (Åkerblom and others, 1984; Eaton and Scott, 1984; Nero and Nazaroff, 1984; Nazaroff and Doyle, 1985; Nazaroff and others, 1985; Nero, 1985; and Sextro and others, 1987). High permeability in the backfill-subslab aggregate zone may account for radon entry that is due to pressure-driven flow. Whether the radon entering a house originates mainly within this zone or in the undisturbed ground beyond is considered next.

DIFFUSION

A concentration of radon in soil gas, resulting from the continual decay of radium in the soil, causes movement of radon in the direction of lessening concentration, without any necessary flow of the soil gas, by the process of diffusion. Radon migration in soil proceeds both by diffusion and by transport with moving soil gas. The ease of migration by diffusion is measured by the effective bulk diffusion coefficient, D_e, of the soil, and the ease of soil-gas flow is measured by its permeability. Nazaroff and others (1988, p. 91) have shown that diffusive migration is dominant if D_e/ϵ is larger than $k\ \Delta P_0/\mu$, where ϵ is the porosity of the soil, k is its permeability, ΔP_0 is a pressure difference driving soil-gas flow, and μ is the absolute viscosity of the soil gas. Pressure-driven flow is dominant if the reverse is true. In general, radon migration by advection (horizontal flow) or convection (vertical flow) predominates in soils composed of well-sorted sand or gravel, and diffusion predominates in poorly sorted and fine-grained uncracked soils. Because the average upward and downward pressure gradients in the soil tend to balance over several days' time, the depletion of radon from the soil to the atmosphere tends to be little more than that which would result from diffusion alone (Schery and others, 1984). The distance that radon moves by diffusion through the soil interstices during one mean life is called its diffusion length, equal to $\sqrt{\tau D_e/\epsilon}$, where τ is the mean life of radon atoms (5.52 days). Søgaard-Hansen and Damkjaer (1987) measured radon diffusion lengths from 1.6 to 1.9 m for dry silt, sand, and gravel samples, and 0.01 m for wet clay. Water in soil pores has long been known to reduce the diffusion coefficient markedly, the inferred mechanism being the blocking of interpore openings by the water. Diffusion is little affected by pore and grain size in most sediments, but permeability is a function of pore and grain size. Both radon diffusion and gas permeability are strongly decreased by water in the pores. No substantial set of data is known to indicate whether an increasing pore water fraction can reduce radon movement by diffusion to a greater extent than by soil-gas flow. Both diffusion and soil-gas flow are retarded by solid soil grains, which increase the path length actually followed by gas molecules to achieve movement in a given direction. The ratio of the actual path length to the distance traversed in the given direction is called tortuosity (Levorsen, 1958, p. 94); Bear (1969, p. 164) treated tortuosity as a dimensionless second-rank tensor.

MODELS OF THE SOIL REGIME NEAR A HOUSE

Slab-on-grade buildings and buildings that have crawl spaces usually are constructed near ground level. For such buildings, the upward flux density of radon through the ground surface is the logical basis for evaluation of the indoor radon potential of the ground. Israël (1962) showed that, under steady diffusion from homogeneous ground to the atmosphere, the amount of radon that can be sustained in a column of the atmosphere of unit ground area is the amount generated in the ground per unit area to a depth equal to the diffusion length discussed previously. An analogous "interstitial mean migration distance" can be obtained by including the effect of steady-state convection due to the pressure difference between the air in the soil and the air in a hypothetical building resting on the soil. Clements (1974) derived an equation (see appendix A) incorporating both diffusion and convection for the steady-state radon flux density from the soil. Multiplication of this flux density by the mean life of radon yields the maximum amount of radon per unit area that can be sustained externally to the soil. Methods of measuring the flux density of radon are called "accumulator methods" and have been reviewed by Collé and others (1981). An accumulator method that includes a provision for suction to measure the convective response of the ground seems to be the most logical approach to evaluate indoor radon potential for slab-on-grade and crawl-space buildings.

For buildings that are to be extended well below grade, where the soil characteristics may differ significantly from those at the surface, flux-density measurements at the surface are less appropriate than measurements of soil characteristics at depth. Several investigators, mentioned in the introduction, are currently making such measurements. Nazaroff (1988) and Tanner (1988) have developed models for using the soil characteristics to predict radon entry into or availability to houses that have basements.

Nazaroff's (1988) model considered a homogeneous ground outside the foundation of a house that has a basement and air flow from the atmosphere down through the soil to an entry zone at the level of the basement slab. The model was applied to a house in ground sufficiently permeable that diffusion could be neglected.

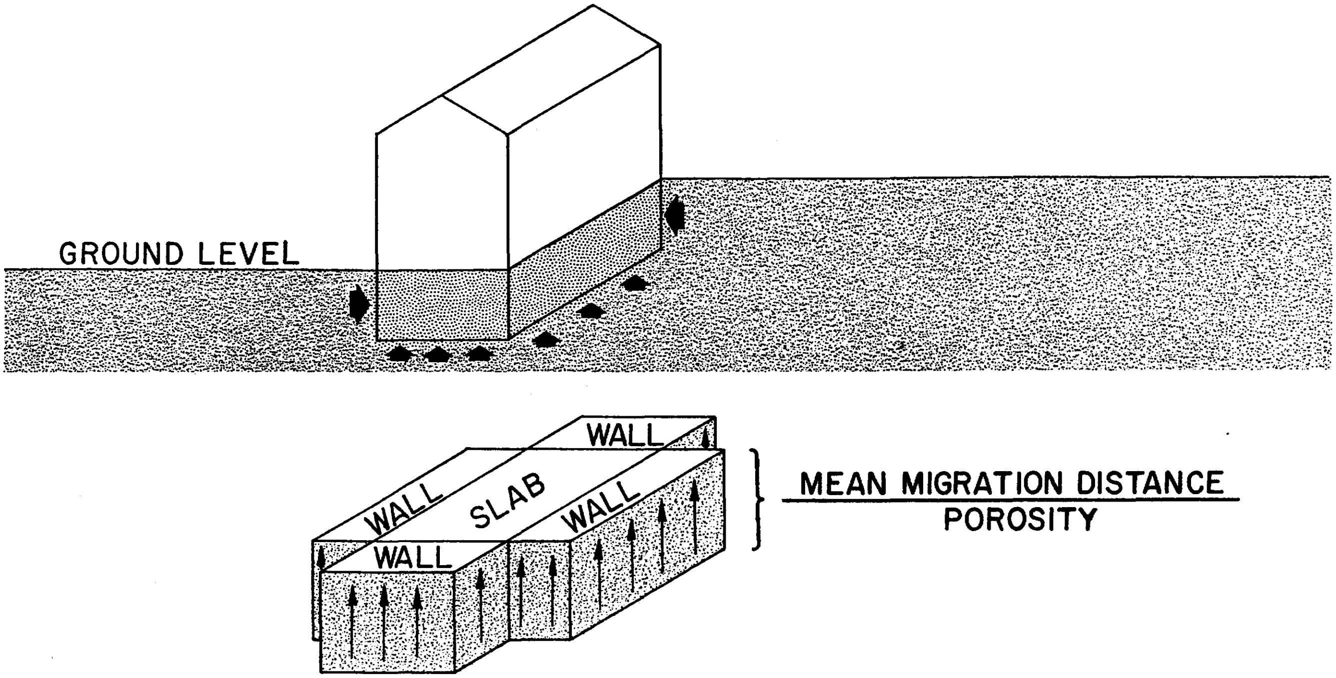

Figure 2. The model used to define radon availability (Tanner, 1988). The upper part of the figure shows a house with a basement and the assumed paths of radon movement to the backfill zones next to the foundation walls and to the subslab aggregate. The lower part is a crude transformation of the upper part to a one-dimensional flow regime and is equal in height to the mean migration distance divided by the porosity.

As part of a proposed protocol, Tanner (1988) also modeled a house having a basement in homogeneous ground but considered both diffusive and convective-advective migration of radon to the foundation and recognized the backfill-subslab aggregate zone only as a conduit for easy transport of radon to entry points. In order to facilitate computation in the protocol, a crude transformation is made by unfolding the basement walls to the plane of projection of the slab, and the migration is modeled as one dimensional (fig. 2) and comparable with the situation for slab-on-grade structures. The protocol obtains a "mean migration distance," which is the interstitial mean migration distance discussed previously, multiplied by the soil porosity. The mean migration distance is the net interstitial volume of radon-bearing soil gas available per unit area, instead of a soil volume per unit area. The apparatus used in the protocol is shown in figure 3. A hole 3.5 cm (centimeters) in diameter is drilled or augered into the ground to a depth of at least 1 m, if possible, and preferably to the depth of the basement slab of the structure proposed for the site. A sample of the soil from the bottom of the hole, packed as nearly as possible to its original density into a canister of known volume, is taken to the laboratory for weighing and determination of its porosity and pore water fraction. The coefficient of molecular diffusion for radon in the porous ground is calculated from the porosity and pore water fraction by means of an empirical relation found by Rogers and others (1984). The sampling probe is inserted into the hole, the rubber packers are inflated by means of a bicycle pump, and soil air is continuously pumped from the sampling space through a flow chamber that responds to the alpha-particle radioactivity of radon and its first decay product by scintillations in a phosphor coating inside the chamber. The scintillations are detected and counted by a photomultiplier tube and its associated electronics to obtain the radon concentration in the soil gas. The rate of flow of the soil gas is measured by a rotameter, and the pressure difference between the sample space and the atmosphere is measured by the sample pressure gauge; these two quantities enable calculation of the permeability of the ground at the sampling point (see appendix B). A "radon availability number" (RAN) is then calculated by inserting the values of radon concentration, diffusion coefficient, and permeability into an equation that is based on a model of one-dimensional upward migration under the influence of a standard pressure difference (see appendix A). The small number of radon availability determinations made so far have correlated roughly with indoor radon values at the respective sites (fig. 4). Because of inhomogeneity of the ground, a yet-undetermined number of measurements must be taken at a site to assure that representative radon availability numbers are obtained. Short- and long-term variability of the soil characteristics also causes significant uncertainties in the method.

Certain soils of moderate to high clay content tend to crack when dry, and as a result permeability is greater along

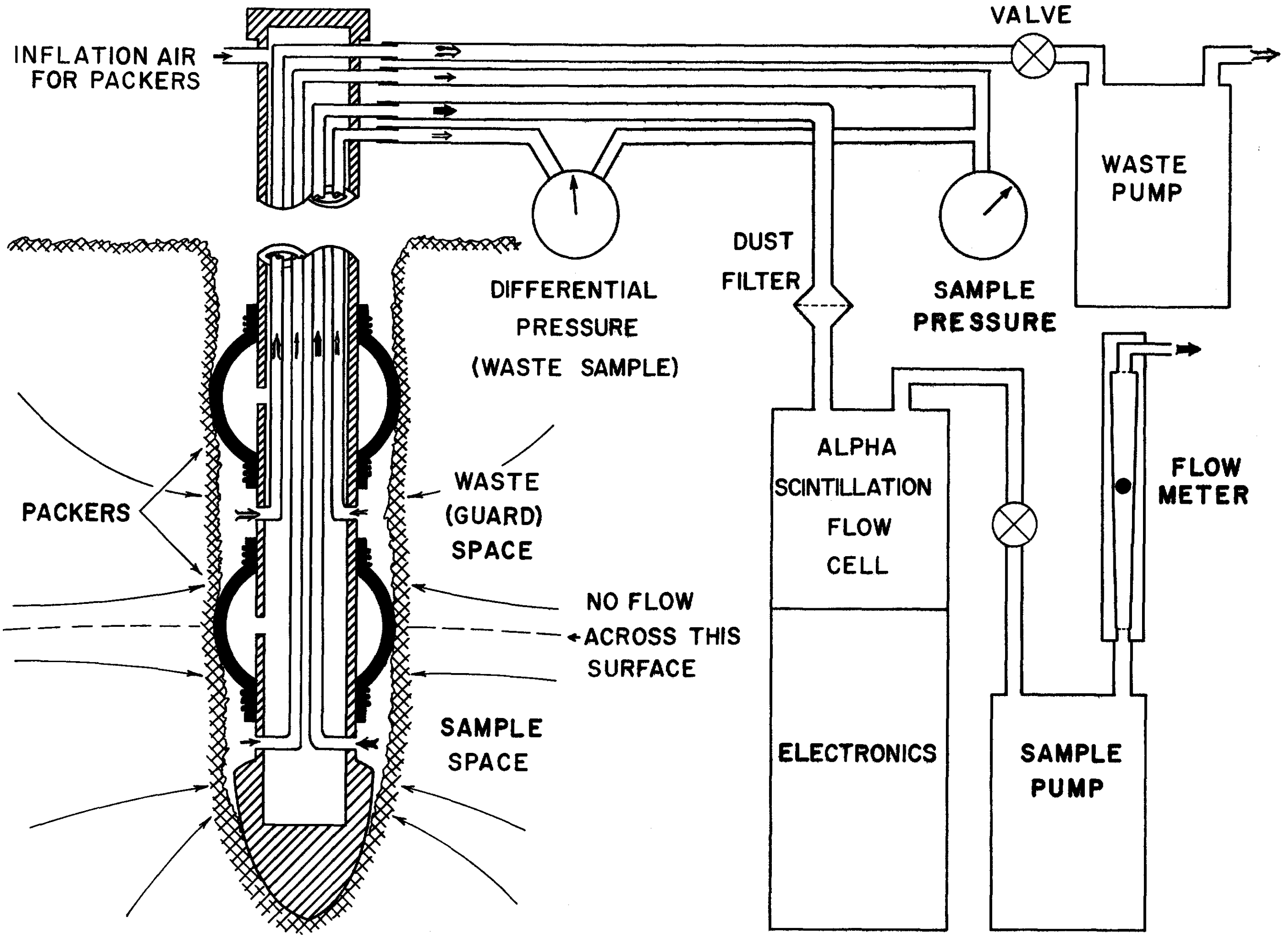

Figure 3. Apparatus for extracting and measuring radon concentration of soil gas and measuring flow rate and driving pressures for permeability calculation. The hole diameter is about 3.5 cm but is not critical. The absolute pressure maintained in the waste space is slightly less than that in the sample space, in order to ensure that atmospheric air does not dilute the soil gas sample taken from the sample space.

the cracks than in the uncracked parts of the soils. Because of their discontinuous nature, cracked soils and rock fractures are difficult to incorporate into models of radon migration into structures. Unless the cracks are closely spaced, test probes are likely to underestimate permeability to an indeterminate degree. Neither the sampling nor the modeling problems associated with cracks and fractures have apparently been resolved at present. However, Schery and Siegel (1986) stated that radon moving through the cracks in soils tends to diffuse transversely into the uncracked parts of the soil and thereby reduce the increase of radon availability due to the cracks.

The maximum amount of interstitial radon originating within the backfill and subslab aggregate zones per unit area can be calculated by multiplying the zones' measured or estimated interstitial radon concentrations by their porosities and thicknesses. Because the stone used under the slab is usually of types that emanate little radon, and because the subslab aggregate layer is normally much thinner than the backfilled gap, the backfill provides most of the radon. If one assumes that the backfilling is done with the native soil, an estimate of the relative importance of the backfill-subslab aggregate zone and of the undisturbed ground as sources of radon may be made by comparing the width of the backfill with the interstitial mean migration distance. Table 1 lists the interstitial mean migration distances of radon calculated for different soil types from representative values of permeability and the interstitial effective diffusion coefficient.

If the undisturbed ground typically is moist or wet clay, the interstitial mean migration distance is much less than the width of the backfilled zone, and the radon available to a structure should be determined by the backfill. It follows that use of backfill of low radium content should minimize radon availability to a structure in fine-grained or typically very moist ground. If the undisturbed ground is fine grained and not normally very moist, both the undisturbed ground and the backfill should contribute sig-

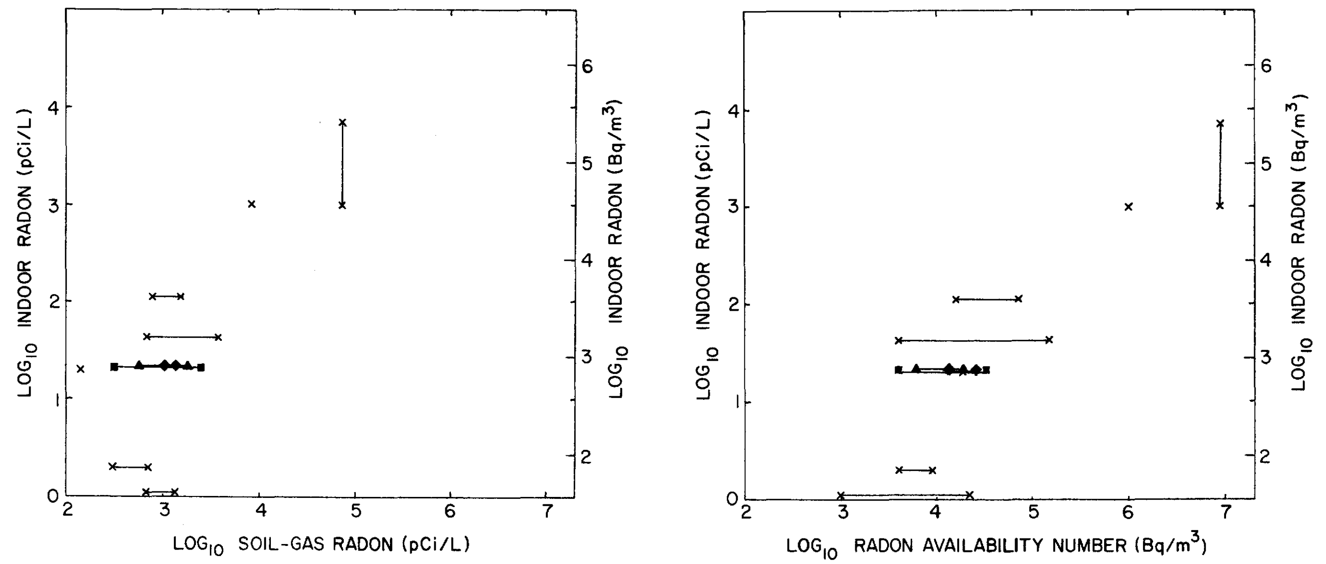

Figure 4. Comparisons of reported indoor radon concentrations with concentrations of radon in soil gas from boreholes near 10 houses (left graph) and with radon availability numbers calculated from data from the same boreholes (right graph). Like symbols joined by lines denote data from multiple boreholes at a given site. pCi/L, picocuries per liter; Bq/m^3, becquerels per cubic meter.

Table 1. Calculated interstitial mean migration distances for radon-222

[Unless otherwise noted, porosity, permeability, and diffusion coefficient values have been selected by the author as representative of the soil types indicated. An arbitrary driving pressure difference of 5 Pa (pascals) is assumed. The mean migration distances have been calculated by the method given by Tanner (1988) and divided by porosity to yield the interstitial mean migration distances]

Soil type	Permeability (*k*, in square meters)	Diffusion coefficient (*D**, in square meters per second)	Porosity (ϵ)	Interstitial mean migration distance (in meters)
Clay, wet	1×10^{-16}	2×10^{-10}	0.45	0.01
Silty clay, moist	1×10^{-15}	7×10^{-8}	.45	.18
Silty sand	1×10^{-13}	3×10^{-7}	.40	.39
Silt	2×10^{-13}	2×10^{-7}	.40	.33
Well-graded silty sand and gravel	2×10^{-12}	4×10^{-7}	.35	.59
Gravelly sandy loam, 4 weight percent water[1]	1.1×10^{-12}	2.7×10^{-6}	.35	1.2
Uniform fine sand:				
17 weight percent water[2]	1×10^{-11}	5×10^{-7}	.30	1.1
8.1 weight percent water	2×10^{-11}	5×10^{-6}	.30	2.1
Well-graded sand and gravel	1×10^{-10}	4.5×10^{-6}	.20	4.0
Uniform medium sand	1×10^{-10}	6×10^{-6}	.25	3.8
Uniform coarse sand	2×10^{-9}	6×10^{-6}	.25	15
Clean gravel	10^{-9}–10^{-7}	7×10^{-6}	.20	10^{1}–10^{2}

[1] Data from Schery and others (1984).

[2] Diffusion coefficients from Grammakov (1936).

nificantly to radon availability. If the undisturbed ground is well-sorted sand or coarse material and is well drained, the backfill should be a minor source of radon.

RADON MEASUREMENT METHODS

The soil-probe methods discussed thus far use "active" methods of radon detection and counting, in that they extract the soil gas and measure its radon content. Another method that should be classified as active is the prompt counting of the gamma rays from bismuth-214 (^{214}Bi), a radon decay product, in hermetically sealed soil samples (Stieff and others, 1987); the method permits determination of both concentration and mobility of radon in the sampled soil. The active methods have the disadvantages of being expensive, requiring equipment costing

$4,000 or more, and (except for the method of Stieff and others, 1987) of indicating the radon concentration and mobility in soil at the measurement time only. The equipment cost may be an acceptable capital cost to State and local government agencies, consultants, and engineering firms; moreover, having immediate answers is preferable to having to wait weeks to months for long-term measurements. However, many investigations of radon in soil are made with inexpensive "passive" methods, which include the etched-track (Alter, 1980), charcoal-adsorption (George, 1984; Hesselbom, 1985), and electret detectors (Kotrappa and others, 1988); methods of absorbing radon from soil gas into a liquid scintillation counting medium (Taipale and Winqvist, 1985; Fukui, 1987); and continuous gamma-ray monitoring of the space in a cylindrical cavity 1 m in diameter by 1 m high filled with coarse rock of low radium activity (Rudakov, 1985a,b).

Users of the passive methods have assumed that radon concentrations at the detectors are equal to the interstitial radon concentrations in undisturbed soil, but decay of radon during its movement to the detector cavity can reduce the concentration at the detector significantly (Tanner, 1991). The concentration of radon available to a passive detector can be estimated by considering diffusion of radon into an air-filled cavity from an infinite homogeneous ground. If diffusion in the ground is isotropic, the diffusion is as efficient as possible and proceeds along radii of an approximately spherical cavity. It can be shown that the ratio of the radon concentration at the center of the cavity to the undepleted radon concentration at infinite radius into the medium is the following function of the cavity radius, r, the porosity, ϵ, the pore water fraction, m, the reciprocal of the radon diffusion length in air, a(0.42 m^{-1}), and the reciprocal of the interstitial effective radon diffusion length in the medium, b:

$$\frac{C_0}{C_\infty} = \frac{\epsilon(1-m)a^3r(br+1)}{b^2ar\cosh(ar)+[\epsilon(1-m)a^2(br+1)-b^2]\sinh(ar)}$$

where cosh and sinh are the hyperbolic cosine and hyperbolic sine, respectively. A lower limit to the diffusion, as might prevail in an uncracked, highly anisotropic layered soil, can be estimated by diffusion along the radii of a vertical cylinder of the same radius. It can be shown that the ratio of the concentration in the cavity to that where it is undepleted follows the relation

$$\frac{C_0}{C_\infty} = \frac{\epsilon(1-m)\sqrt{D_m}}{\epsilon(1-m)\sqrt{D_m}I_0(ar)+\sqrt{D_c}I_1(ar)[K_0(br)/K_1(br)]}$$

where D is the interstitial effective diffusion coefficient, the subscript m referring to the medium and the subscript c referring to the cavity; I and K refer to modified Bessel functions of the first and second kinds, respectively; and the subscripts 0 and 1 refer to the orders of the Bessel functions. Figure 5 shows graphs of these relations for several different cavity radii and for interstitial effective diffusion lengths corresponding to the indicated degrees of pore water saturation. The graphs show that passive detectors in small cavities do not yield significant errors until the surrounding ground is nearly saturated but that meter-sized cavities yield errors even in moderately dry ground. Because of the rapid diffusion of radon in a continuous air medium, the concentration ratios for cavities of the radii shown differ by no more than 2 percent between the cavity centers and the cavity walls. The position of the detector in the cavity is therefore not important.

Although the radon concentration in the soil gas in a cavity must be less than that in the soil itself, an increase in the pore water fraction can cause an increase in the radon concentration in the cavity, because radon is concentrated in the gas phase of a gas-water system. It is reasonable to assume that the rate of radon emanation into the pores of the medium is not reduced as the water fraction in the pores is increased. Reduction of the rate of emanation of radon from samples with increasing moisture content, as observed by Lindmark and Rosen (1985) and by Damkjaer and Korsbech (1985), is likely due to the diminished rate of diffusion from the sample; Bates (1980) showed by computation that a period of 10 hours is needed to reach a steady-state flux of radon from a cylindrical sample of 9.2-cm length, 3.843-cm radius, 0.2 porosity, 1×10^{-7}-cm^2 permeability, and 10^{-2}-cm^2/s diffusion coefficient (equivalent to about 90 percent saturation) at 300 K. Because radon follows Henry's Law (Boyle, 1911), the concentration of radon in the water phase of the pore fluid is in temperature-dependent proportion to the partial pressure (or concentration) of radon in the gas phase. If it is assumed that equilibrium is quickly established between the phases and that there is no gain or loss of radon from the pores, the ratio of the concentration of radon in the gas phase to the (constant) average concentration of radon in the pores can be shown to be

$$C_{\mathrm{gas}}/C_{\mathrm{pores}} = 1/[m(R-1)+1],$$

where m is the pore water fraction, and R is the ratio of the radon concentration in the water phase to that in the gas phase. R is dependent only on temperature in dilute solutions (Köfler, 1913). A composite curve showing determinations of R by several authors was presented by Rogers (1958, fig. 44); slightly different values can be derived from Harley and Terilli (1986). Figure 6 shows the radon concentration in the gas phase, normalized to unit average concentration in the pores, as the pore water fraction increases.

The combination of enrichment of radon in the gas phase and increased decay of the radon along its path to a passive detector can result in apparent radon concentrations that are higher than, equal to, or lower than the actual average radon concentration in the pores, depending on the cavity size, the soil temperature, the pore water fraction,

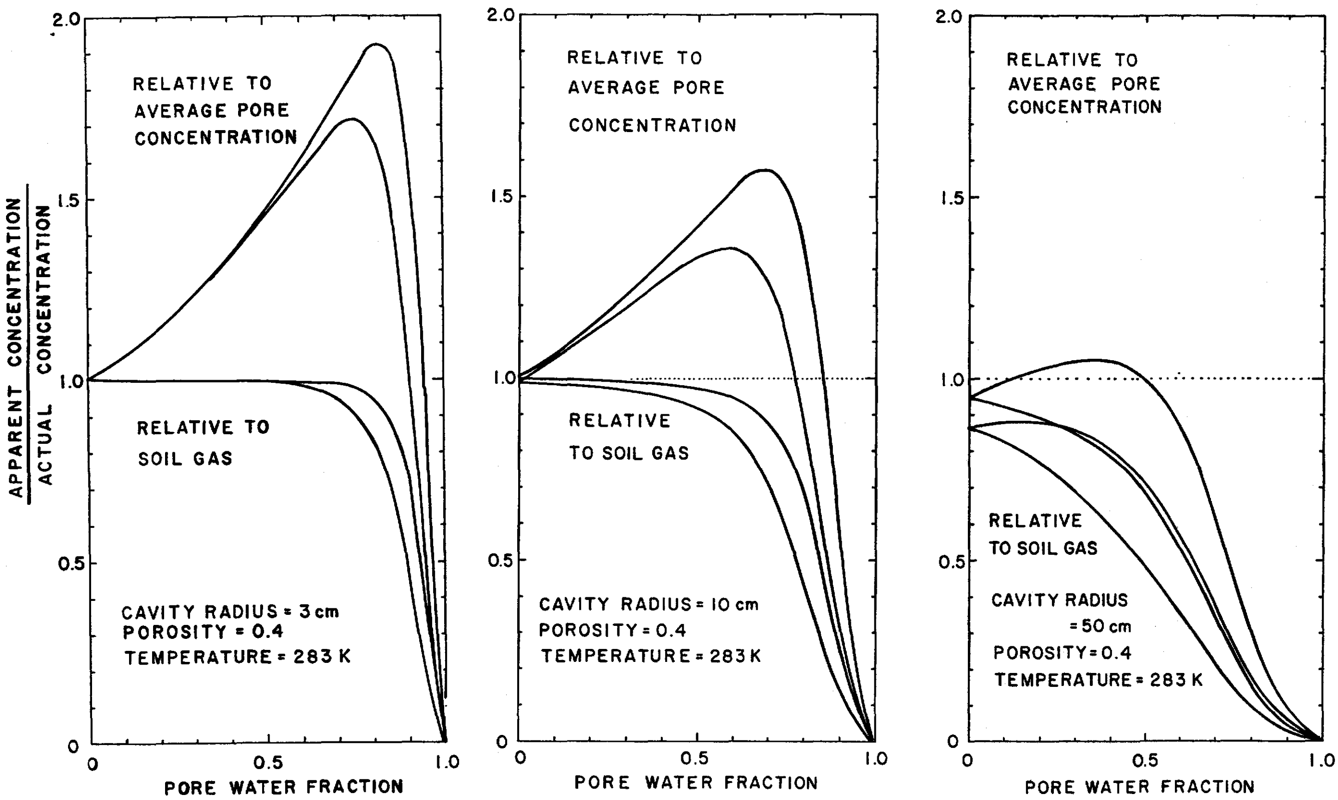

Figure 5. Curves showing the error in estimation of radon concentration by passive detectors. The curves are based on diffusion models. All curves relate the radon concentration at the center of the passive-detector cavity to the radon concentration at a sufficient distance from the cavity (typically less than a few meters) to be unaffected by radon diffusion toward the cavity. The lower pair of curves in each frame relates the radon concentration at the cavity center to the undepleted radon concentration in the soil gas. The upper pair of curves in each frame relates the radon concentration at the cavity center to the amount of radon in both gas and water phases per unit pore volume. For each pair of curves, the upper curve was computed from a spherical diffusion model, and the lower curve was computed from a cylindrical diffusion model. The regions between the curves of each pair indicate the range of uncertainty due to variability of diffusion anisotropy.

and the range and anisotropy of the diffusion coefficient. The active methods are affected by the enrichment of radon in the gas phase, but decay is negligible in transit to the sampling device; in principle the measurements can be corrected if the temperature and pore water fraction in the volume of ground sampled are known.

SHORT-TERM METEOROLOGICAL EFFECTS

Meteorological effects on radon concentrations in soil gas are discussed at length by Asher-Bolinder and others in another paper of this Bulletin, entitled "A Preliminary Evaluation of Environmental Factors Influencing Day-to-Day and Seasonal Soil-Gas Radon Concentrations." Rainfall and snowmelt have several effects on the concentration of radon in soil pores and on the ease of radon migration. The short-term effects are greatest near the surface in most soils: if the ground surface is sealed, exhalation of radon to the atmosphere is nearly halted, the near-surface radon concentration builds up, and the ability of radon to migrate is drastically reduced. At depths greater than 1 m in most fine-grained soils, the immediate effects are that exhalation to the atmosphere is reduced but mobility is not reduced significantly. As the water percolates downward, the effects of enrichment of the gas phase and shortening of the mean migration distance should take place. Harley and Terilli (1986) have noted radon "events" taking place in a house that are due to changing water vapor pressure in soil, as caused by temperature pulses, which can be induced by infiltration of rainfall that is warmer than the soil. These factors indicate that radon assessment procedures should not be conducted at depths much less than 1 m.

SEASONAL EFFECTS

Rudakov (1985b) reported a quasi-sinusoidal variation of mean monthly radon concentrations in 1-m-diameter cylinders filled with low-activity coarse rock in cavities between 1 and 2 m below the ground surface at two locations in central Asia, Soviet Union, over faults. The

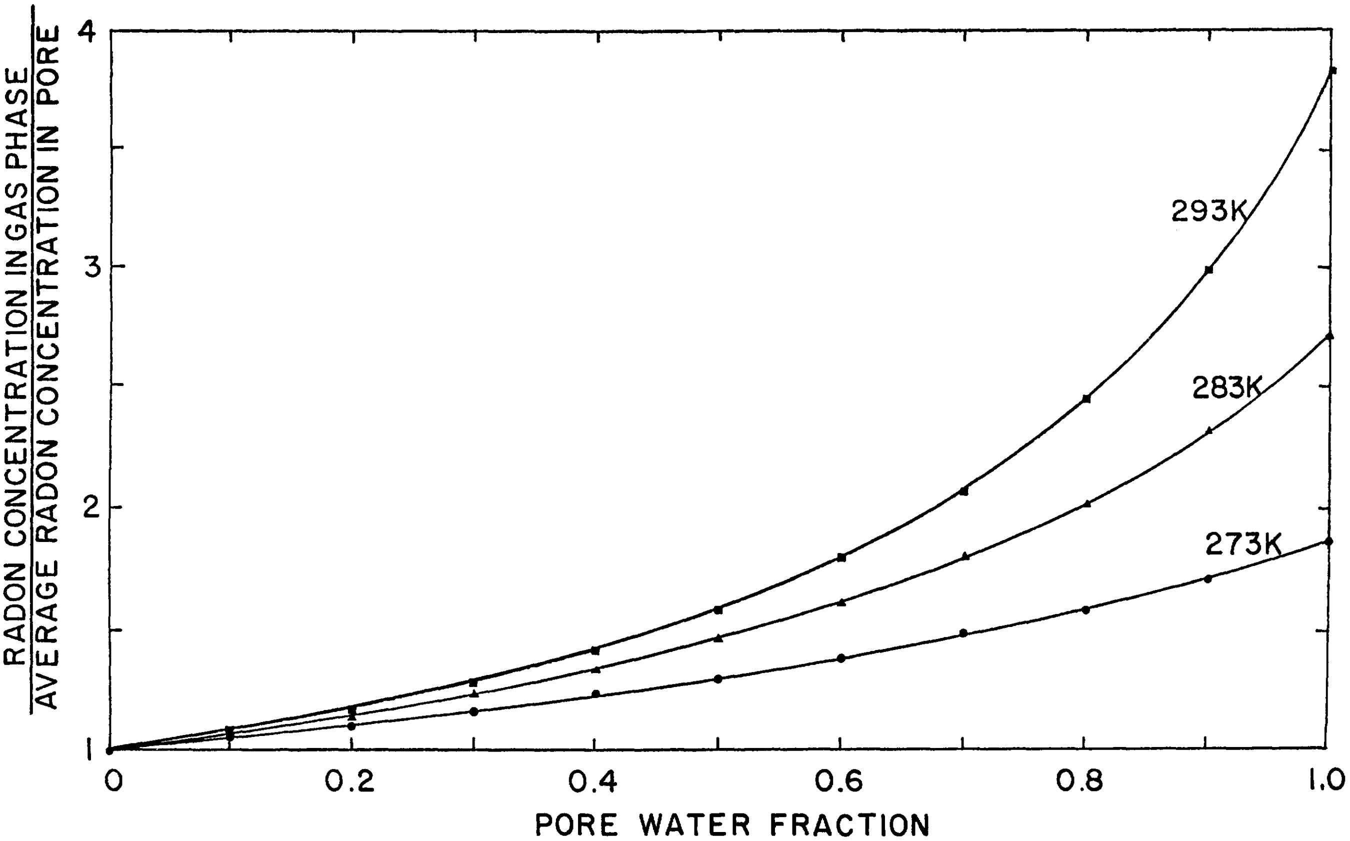

Figure 6. Curves showing the calculated ratio of the radon concentration in the gas phase of the pore space in soil to the average radon concentration in the pore space as a function of pore water fraction for 273 K (0 °C), 283 K (10 °C), and 293 K (20 °C).

maximum and minimum values occurred in January and July, respectively, and differed by a factor of four. The radon concentrations were inversely related to surface temperature and thermoplastic deformations and directly related to barometric pressure; they were inferred from gamma radiations from a radon decay product measured by detectors at the centers of the cylinders and were influenced by the in-transit radon decay shown in figure 5.

Rose and others (1988) observed seasonal changes of radon concentration of a similar fourfold range, but of opposite phase, in a well-drained silt loam soil in central Pennsylvania. The measurements were made with passive etched-track detectors but appear to have been verified by soil-gas extraction and counting in an alpha scintillation cell.

Using the soil-probe instrumentation of figure 3, I measured the radon concentration in soil gas from a hole 1.2 m deep in a moderately drained silt loam in Fairfax County, Va. Table 2 shows the variation of soil characteristics measured during different seasons. The fact that the radon concentration in the soil gas did not vary significantly may be a recommendation for the use of active measurement methods. At this site radon migration was dominated by diffusion, which is strongly affected by the pore water fraction. The pore water fraction and porosity were determined by packing a sample of the soil from the hole bottom in a canister and measuring the bulk density of the soil as collected and after desiccation. An indeterminate error is caused by failure to achieve the same bulk density in the canister as in the ground. The high porosity calculated for the measurements of October 31, 1988, may have resulted from insufficient pressure in packing the canister, which could have led to an underestimate of the pore water fraction and an overestimate of the diffusion coefficient, mean migration distance, and radon availability number.

The ranges and qualitatively different results of measurements reviewed in this paper and by Asher-Bolinder and others in this Bulletin indicate that we cannot yet closely characterize soils for their indoor radon potential from measurements made at a single time. The principal difficulty is estimating the average pore water fraction of the soil over a few years' time. Alternatives to instantaneous, in situ measurements would be desirable.

ACKNOWLEDGMENTS

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Table 2. Characterization of soil at 1.2 m in depth at a site in Fairfax County, Va.

[Data were obtained by means of procedures and calculations given in the protocol proposed by Tanner (1988); n.d., not determined]

Soil characteristic	Dates of measurements			
	July 9, 1987	Jan. 19, 1988	April 14, 1988	Oct. 31, 1988
Radon concentration (in nanocuries per liter)	1.02	0.95	1.19	1.01
Pore water fraction	n.d.	.76	.66	.30
Porosity	n.d.	.51	.43	.60
Diffusion coefficient (10^{-2} m^2/s)	n.d.	.27	.5	3.2
Permeability (10^{-13} m^2)	8.7	6.0	8.2	4.6
Mean migration distance (in meters)	n.d.	.20	.26	.74
Radon availability number (in kilobecquerels per square meter)	n.d.	7	12	40

Agreement No. DW14931927–01–0. Investigation of gas transport in soils and its relation to radon availability was partly supported by the Office of Health and Environmental Research of the U.S. Department of Energy under grant DE-AI05-87ER60578.

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APPENDIX A. DERIVATION OF THE RADON AVAILABILITY NUMBER (RAN)

1. Quantities, symbols, units, and dimensions

Quantity	Symbol	Value	Units	Dimensions
Activity concentration in interstitial fluid	C	variable	Bq/m^3	$L^{-3}T^{-1}$
Diffusion coefficient (effective diffusion coefficient in interstitial fluid), $D^*=D_e/\epsilon$	D^*	variable	m^2/s	L^2T^{-1}
Diffusion coefficient (effective diffusion coefficient for geometrical area), $D_e=\epsilon D^*$	D_e	variable	m^2/s	L^2T^{-1}
Flux density	J	variable	Bq/m^2 · s	$L^{-2}T^{-2}$
Porosity	ϵ	variable	none	none
Decay constant, Rn-220 (thoron)	λ_{Tn}	1.25×10^{-2}	/s	T^{-1}
Decay constant, Rn-222 (radon)	λ_{Rn}	2.10×10^{-6}	/s	T^{-1}
Mean life, Rn-222 (5.52 days)	τ	4.77×10^{5}	s	T
Depth (positive upward)	z	variable	m	L
Velocity of convection across geometrical area (Darcy velocity)	v	variable	m/s	LT^{-1}
Rate of radon generation into interstitial fluid	G	variable	Bq/s · m^3	$L^{-3}T^{-2}$
Viscosity of air (absolute or dynamic) at 290 K	μ	1.82×10^{-5}	Pa · s	$ML^{-1}T^{-1}$
Permeability	k	variable	m^2	L^2
Mean migration distance in geometrical volume	M	variable	m	L

2. Assumptions

a. The ground can be modeled as a homogeneous, isotropic permeable medium.

b. The interstitial (pore) spaces are large compared with the mean free path of the radon isotope.

c. Absorption of radon into liquids in the pores and adsorption of radon by surfaces of the pores can be ignored.

d. The gravitational term in Darcy's Law may be neglected; v is proportional to the pressure gradient in the interstitial gas and inversely proportional to the dynamic viscosity of the gas, the permeability being defined as the proportionality constant.

e. Prior to the calculation of mean migration distance, the porosity, diffusion coefficient, and permeability have been determined. Procedures for obtaining these data were given by Tanner (1988).

3. Calculation of mean migration distance

W.E. Clements' dissertation (1974) introduced the parameters N_t and γ, defined as follows:

$$N_t=v/(2(\epsilon\lambda\ D_e)^{1/2}) \qquad (1)$$

and

$$\gamma=N_t+(N_t^2+1)^{1/2}. \qquad (2)$$

Clements' equation 2–24 gave the steady-state flux density of radon at the Earth's surface as

$$J=G\gamma\ (\epsilon D_e/\lambda)^{1/2}. \qquad (3)$$

We multiply the left side of equation 3 by the mean life of radon, τ, and divide the right side by its reciprocal, λ, to obtain the amount of radon exhaled during one mean life:

$$J\tau=(G/\lambda)\gamma\ (\epsilon\ D_e/\lambda)^{1/2}. \qquad (4)$$

The left side corresponds to the radon availability number (RAN). The expression G/λ is the concentration of radon in the interstitial fluid at infinite depth (that is, where there is insignificant depletion by exhalation), and the remaining factor is the mean migration distance averaged over the geometrical volume (that is, the mean interstitial migration distance multiplied by the porosity). Then

$$M=\gamma(\epsilon D_e/\lambda)^{1/2} \qquad (5)$$

and

$$\mathrm{RAN}=C_{-\infty}\,M, \qquad (6)$$

where $C_{-\infty}$ is the radon concentration in soil gas at sufficient depth to be undepleted by loss to the atmosphere and M must be computed from equation 5. M cannot be calculated directly from the observed or estimated quantities, however, because the value of v, the Darcy velocity, is not directly known. It could be calculated from the Darcy relation

$$v=-(k/\mu)(dp/dz) \qquad (7)$$

if the pressure gradient were known, but the known quantity is the overall pressure difference, 5 Pa, to some distance from which migration is inconsequential (assumed to be 4.6 times the interstitial mean migration distance, or the distance over which hundredfold diminution of a radon flux takes place); this depth is dependent on M, the quantity sought. The approach used is to assume that the pressure gradient, dp/dz, is numerically equal to the pressure difference, to compute M, to compute the quantity

$$(dp/dz)(4.6\ M/\epsilon) \qquad (8)$$

and to recompute M using a larger or smaller value of pressure gradient until the above quantity approaches the value of −5 Pa to within 1 percent.

APPENDIX B. DERIVATION OF RELATIONS AMONG PERMEABILITY, FLOW RATE, AND PRESSURES IN THE HEMISPHERICAL GEOMETRY OF THE SAMPLING HOLE

1. Quantities, symbols, units, and dimensions

Quantity	Symbols	Value	Units	Dimensions
Pressure	p	variable	Pa	$ML^{-1}T^{-2}$
Atmospheric pressure	p_a		Pa	
Pressure in sampling hole	p_w		Pa	
Volume	V	variable	m^3	L^3
Viscosity of air (absolute or dynamic) at 290 K	μ	1.82×10^{-5}	Pa · s	$ML^{-1}T^{-1}$
Permeability	k	variable	m^2	L^2
Flow rate, volume basis	Q_v	variable	m^3/s	L^3T^{-1}
Velocity of fluid flow across geometrical area (Darcy velocity)	v	variable	m/s	LT^{-1}

2. Assumptions

a. The ideal gas law is sufficiently accurate to describe soil gas (air) in the absolute pressure range from near zero to atmospheric.

b. Soil gas moves into the sampling hole isothermally; contact with soil grains is sufficient to furnish heat to the soil gas during the 10-minute sampling period to minimize cooling of the soil gas.

c. Gravitational effects over the vertical range of a few meters are negligible.

d. The ground is isotropic and of uniform porosity.

e. The equation of continuity applies to the square of the pressure (or density); that is, the Laplacian of pressure squared (or gas density squared) equals zero. See, for example, Greenkorn (1983, eq. 119 and 120), Bird and others (1960, eq. 4.J–7), or Muskat (1937, sec. 11.2, eq. 1).

f. The flow of soil gas into the waste and sampling cavities is separated by a horizontal plane passing through the midpoint of the lower packer; the flow is approximately hemispherical.

g. The flow of soil gas is adequately represented by Darcy's Law. In very fine material, Darcy's Law becomes inaccurate, but the permeability is then negligibly small.

h. Except for the disturbance of the pressure field created by the sampling, pressure in the ground is equilibrated with that in the near-surface atmosphere. Propagation of atmospheric pressure changes is slow in very fine materials, but their permeabilities are negligibly small.

3. Derivation of the pressure function.

Under the above assumptions,

$$\nabla^2 p^2=0. \tag{1}$$

For spherical flow in the radial direction in spherical coordinates the above equation may be reduced (see Muskat, 1937, sec. 5.2, eq. 2) to

$$\frac{1}{r^2}\frac{d}{dr}\left(r^2\frac{d(p^2)}{dr}\right)=0\ , \tag{2}$$

which implies that

$$r^2\frac{d(p^2)}{dr}=\text{constant}=c_1 \tag{3}$$

$$\int d(p^2)=c_1\int\frac{1}{r^2}\,dr \tag{4}$$

$$p^2=c_2-\frac{c_1}{r} \tag{5}$$

$$p=\left(c_2-\frac{c_1}{r}\right)^{1/2}\ . \tag{6}$$

At r = infinity, the pressure is assumed to be that of the atmosphere; hence,

$$c_2=p_a^2\ . \tag{7}$$

At r = radius of sampling hole, r_w,

$$p_w=\left(p_a^2-\frac{c_1}{r_w}\right)^{1/2}\ , \tag{8}$$

whence

$$c_1=r_w(p_a^2-p_w^2)\ . \tag{9}$$

Let the difference of the squared pressures be represented by Y. Then

$$p=\left(p_a^2-\frac{r_wY}{r}\right)^{1/2}\ . \tag{10}$$

In order to obtain the velocity of radial flow into the sampling hole we must obtain the pressure gradient, dp/dr, and evaluate it at the spherical surface of the hole:

$$\frac{dp}{dr}=\frac{d}{dr}\left(p_a^2-\frac{r_w Y}{r}\right)^{1/2} \tag{11}$$

$$=\frac{1}{2}\left(p_a^2-\frac{r_w Y}{r}\right)^{-1/2}\frac{d}{dr}\left(p_a^2-\frac{r_w Y}{r}\right)$$

$$=\frac{1}{2}\left(p_a^2-\frac{r_w\, Y}{r}\right)^{-1/2}\left(\frac{r_w\, Y}{r^2}\right) . \tag{12}$$

From equations 6 and 12,

$$\frac{dp}{dr}=\frac{r_w\, Y}{2p\, r^2} \tag{13}$$

or

$$\frac{dp}{dr}=\frac{r_w}{2p\, r^2}\,(p_a^2-p_w^2) . \tag{14}$$

4. Calculation of permeability from flow into the sampling hole

Note that Darcy's Law applies to flow across a geometrical surface. The radial flow velocity is then proportional to the pressure gradient,

$$v=\frac{-k}{\mu}\,\frac{dp}{dr} . \tag{15}$$

From equations 14 and 15,

$$v=-\frac{k\, r_w(p_a^2-p_w^2)}{\mu\; 2\, p\, r^2} . \tag{16}$$

The volumetric flow into the sampling hole is the product of the radial flow velocity and the spherical surface area of the sampling hole:

$$Q_v=4\pi r_w^2 v . \tag{17}$$

Combining equations 16 and 17 and evaluating v at $r=r_w$,

$$Q_v=\frac{-4\,\pi\, r_w^2\, k\, r_w\,(p_a^2-p_w^2)}{\mu\; 2\, p_w\, r_w^2} \tag{18}$$

or

$$Q_v=\frac{-2\,\pi\, k\, r_w\,(p_a^2-p_w^2)}{\mu\, p_w} . \tag{19}$$

For hemispherical flow, Q_v is reduced by a factor of two, yielding

$$Q_v=\frac{-\pi\, k\, r_w\,(p_a^2-p_w^2)}{\mu\, p_w} \tag{20}$$

where the minus sign indicates flow in the direction of decreasing radius (that is, inward to the sampling cavity). The permeability is then

$$k=\frac{Q_v\,\mu\, p_w}{\pi r_w(p_a^2-p_w^2)} . \tag{21}$$

Equation 21 may be modified by expanding the difference of squared pressures,

$$(p_a^2-p_w^2)=(p_a-p_w)\,(p_a+p_w) \tag{22}$$

and observing that (p_a-p_w) is the pressure difference between the atmosphere and the sample space and that for small pressure differences (p_a+p_w) is approximately equal to $2p_w$:

$$k=\frac{Q_v\,\mu\, p_w}{\pi\, r_w(p_a-p_w)2p_w} , \tag{23}$$

which simplifies to

$$k=\frac{Q_v\,\mu}{2\,\pi\, r_w(p_a-p_w)} \tag{24}$$

for hemispherical geometry or to

$$k=\frac{Q_v\,\mu}{4\,\pi\, r_w(p_a-p_w)} \tag{25}$$

for spherical geometry.

APPENDIX C. DERIVATION OF RELATIONS AMONG PERMEABILITY, FLOW RATE, AND PRESSURES FOR RADIAL FLOW INTO A CYLINDRICAL SAMPLING HOLE

1. Quantities, symbols, units, and dimensions

Quantity	Symbol	Value	Units	Dimensions
Pressure	p	variable	Pa	$ML^{-1}T^{-2}$
Atmospheric pressure	p_a			
Pressure in sampling hole	p_w			
Volume	V	variable	m^3	L^3
Viscosity of air (absolute or dynamic) at 290 K	μ	1.82×10^{-5}	$Pa \cdot s$	$ML^{-1}T^{-1}$
Permeability	k	variable	m^2	L^2
Flow rate, volume basis	Q_v	variable	m^3/s	L^3T^{-1}
Velocity of fluid flow across geometrical area (Darcy velocity)	v	variable	m/s	LT^{-1}
Radius of sampling cavity	r_w	variable	m	L
Height of sampling cavity	h	variable	m	L

2. Assumptions

a. The ideal gas law is sufficiently accurate to describe soil gas (air) in the absolute pressure range from near zero to atmospheric.

b. Soil gas moves into the sampling hole isothermally; contact with soil grains is sufficient to furnish heat to the soil gas during the 10-minute sampling period to minimize cooling of the soil gas.

c. Gravitational effects over the vertical range of a few meters are negligible.

d. The ground is isotropic and of uniform porosity.

e. The equation of continuity applies to the square of the pressure (or density); that is, the Laplacian of pressure squared (or gas density squared) equals zero. See, for example, Greenkorn (1983, eq. 119 and 120), Bird and others (1960, eq. 4.J–7), or Muskat (1937, sec. 11.2, eq. 1).

f. The flow of soil gas into the sampling cavity is bounded by horizontal planes passing through the midpoints of the packers above and below the sampling cavity; the flow is along radii of the cylindrical cavity.

g. The flow of soil gas is adequately represented by Darcy's Law. In very fine material, Darcy's law becomes inaccurate, but the permeability is then negligibly small.

h. Except for the disturbance of the pressure field created by the sampling, pressure in the ground is equilibrated with that in the near-surface atmosphere. Propagation of atmospheric pressure changes is slow in very fine materials, but their permeabilities are negligibly small.

3. Derivation of the pressure function

Under the above assumptions,

$$\nabla^2 p^2 = 0. \tag{1}$$

For flow in the radial direction in cylindrical coordinates the above equation may be reduced (see Muskat, 1937, sec. 4.2, eq. 3) to

$$\frac{1}{r}\frac{d}{dr}\left(r\frac{d\,(p^2)}{dr}\right)=0, \tag{2}$$

which implies that

$$r\,\frac{d(p^2)}{(dr)}=\text{constant}=c_1 \tag{3}$$

$$\int d(p^2)=c_1\int\frac{1}{r}dr \tag{4}$$

$$p^2=c_1\ln(r)+c_2\ . \tag{5}$$

Equation 5 may be evaluated at the boundary conditions $r=r_w$, $p=p_w$ at the surface of the cylindrical cavity and $p=p_e$ at some radius r_e into the ground:

$$p_e^2=c_1\ \ln(r_e)+c_2 \tag{6}$$

$$p_w^2=c_1\ \ln(r_w)+c_2\ . \tag{7}$$

Subtraction of equation 7 from equation 6 yields

$$c_1=\frac{p_e^2-p_w^2}{\ln(r_e/r_w)}\ , \tag{8}$$

and substitution of the right side of equation 8 for c_1 in equation 7 yields

$$c_2=\frac{p_w^2\ln(r_e)-p_e^2\ln(r_w)}{\ln(r_e/r_w)}\ . \tag{9}$$

By taking the square root of equation 5, we obtain the pressure distribution as a function of radius,

$$p=[c_1\ \ln(r)+c_2]^{1/2} \tag{10}$$

from which the pressure gradient is

$$dp/dr=c_1/2r\ [c_1\ \ln(r)+c_2]^{1/2} \tag{11}$$

or

$$dp/dr=\frac{(p_e^2-p_w^2)}{2r(\ln(r_e/r_w)[p_e^2(\ln(r/r_w))-p_w^2(\ln(r_e/r_w))])^{1/2}}\ . \tag{12}$$

If equation 12 is evaluated at the boundary of the sample cavity ($r=r_w$),

$$dp/dr=\frac{(p_e^2-p_w^2)}{2r_w(\ln(r_e/r_w)[p_e^2(\ln(r_w/r_w))-p_w^2(\ln(r_e/r_w))])^{1/2}} \tag{13}$$

$$=\frac{(p_e^2-p_w^2)}{2r_w(\ln(r_e/r_w)[p_w^2(\ln(r_e/r_w))])^{1/2}} \tag{14}$$

$$=\frac{(p_e^2-p_w^2)}{2r_wp_w(\ln(r_e/r_w)} \ . \tag{15}$$

4. Calculation of permeability from flow into the sampling hole

Note that Darcy's Law applies to flow across a geometrical surface. The radial flow velocity is then proportional to the pressure gradient,

$$v = -(k/\mu)(dp/dr) \ . \tag{16}$$

From equations 15 and 16,

$$v=-\frac{k \quad (p_e^2-p_w^2)}{\mu 2r_wp_w\ln(r_e/r_w)} \ . \tag{17}$$

To obtain the total volumetric flow rate across the boundary, we multiply the flow velocity by the cylindrical surface area, $2\pi r_wh$, obtaining

$$Q_v=-\frac{k\pi h(p_e^2-p_w^2)}{\mu p_w\ln(r_e/r_w)} \ , \tag{18}$$

where the minus sign indicates flow in the direction of decreasing radius (that is, inward to the sampling cavity). The permeability is then

$$k=\frac{Q_v\mu p_w\ln(r_e/r_w)}{\pi h(p_e^2-p_w^2)} \ . \tag{19}$$

Equation 19 may be simplified by expanding the difference of squared pressures,

$$(p_e^2-p_w^2)=(p_e-p_w)(p_e+p_w) \tag{20}$$

and observing that (p_e-p_w) is approximately equal to the pressure difference between the atmosphere and the sampling space and that for small pressure differences (p_e+p_w) is approximately equal to 2 p_w:

$$k=\frac{Q_v\mu p_w\ln(r_e/r_w)}{\pi h(p_e-p_w)2p_w} \ , \tag{21}$$

which simplifies to

$$k=\frac{Q_v\mu\ln(r_e/r_w)}{2\pi h(p_e-p_w)} \ . \tag{22}$$

Equation 22 corresponds to the expression for flow of an incompressible fluid. In either case, the radius r_e is not easily established. It cannot be extended to infinity, because $\ln(r_e/r_w)$ also becomes infinite. However, because r_e is not dependent on soil type, and because the effect of the sampling becomes inconsequential at a distance of the order of 10 m or less, an arbitrary selection of $r_e=10$ m is reasonable. At such distance, p_e is very close to atmospheric pressure under assumption h, above, and (p_e-p_w) may be referred to atmospheric pressure, as is the practice. The factor $\ln(r_e/r_w)$ is not sensitive to variation of r_w in a practical range.

SIMPLE TECHNIQUES FOR SOIL-GAS AND WATER SAMPLING FOR RADON ANALYSIS

G.M. Reimer

Abstract

Various devices and techniques are available for sampling soil air and ground water for analysis of their gas content. A simple approach to collection is presented here and can be useful as a screening technique for radon when analyses are desired at the field location. For soil-gas samples, a small-diameter probe is pounded into the ground to a depth of at least 0.75 meter. For water, a plastic bottle containing the sample is shaken so headspace air purges the gases from the water. Once the gas samples are extracted from the soil or water, they can be analyzed with whatever field technique is desired.

INTRODUCTION

Measured soil-gas and ground water concentrations of radon (Rn) are important parameters needed to evaluate the radon potential in any location. Preliminary field measurements often are important when designing a larger program to assess the radon potential. Simple techniques and materials for soil-gas sampling and water sampling that can be used for screening are described. Although these techniques are not part of any protocol for sampling, they are in general use by different programs within the U.S. Geological Survey. These methods can be used for some aspects of research to understand the distribution and migration of radon. They also can be easily modified to suit a variety of analytical schemes and instruments.

SOIL-GAS SAMPLING TECHNIQUE

Soil-gas sampling is an integral part of the scientific methodology used to understand the distribution, migration, and availability of radon gas. Many factors affect the concentration of radon in soil, such as the radium concentration, the emanation characteristics of the soil materials, the soil porosity, permeability, and moisture content, and, of course, meteorological conditions. Similarly, many factors come into play when collecting a sample of the soil gas for analysis. Samples typically are collected by augering a hole or driving a probe into the soil. A detector is set in the hole for a time-integrated analysis to measure radon concentrations emanated from the soil, or pumping is performed in some fashion to obtain a sample for a more instantaneous analysis of pore-space radon. By their very nature, all these methods disturb the soil environment to some degree, and they sample different regimes of the soil air. Therefore, the selection of a specific soil-gas sampling method is based upon some compromises such as requirements for a special sample size, the need for unique pumping or pressure differential requirements, or the need for sampling at a particular soil horizon or depth.

The sampling method described here has many advantages for a typical sampling program where the relative distribution of radon concentrations is sought for the surficial environment. This method has evolved, like so many others, from the uranium exploration programs of the past. It has advantages and limitations, but both can be defined.The technique involves pounding a small-diameter hollow steel probe into the ground and using a hypodermic syringe for collecting the sample. This approach is very accommodating for samples to be analyzed on an electronic alpha-particle scintillometer. The advantages of this method are that it provides for fast sampling, only a small quantity of sample needs to be collected, and there is minimal disturbance to the soil environment. The same collection system can be used for determining the soil permeability of the sample site. It also measures the actual in situ pore-space concentration of radon that can then be used more directly for radon availability measurements. The primary disadvantage is that it is a "grab sample," which defines the concentration only for the environmental and meteorologic conditions that exist at the time of collection.

Description of Soil-Gas Sampling Equipment and Technique

The basic equipment is of simple design made from easily obtainable supplies. Size and grade of the materials used are not critical, but some guidelines are offered. Considerations concerning the type of sampling to be performed contribute to the specific design of the sampling equipment. In this case, for example, there are criteria for minimum disturbance to the sampling environment. The probe should be durable and should not clog easily when used in a variety of soil types; the sampling system should be portable to permit fairly rapid sampling at remote areas.

In such a system, as shown in figure 7, the probe is small-diameter, thick-walled carbon steel tubing. The outside diameter can range from 6 to 9 mm (millimeters) and the inside diameter from 2 to 3 mm. Sizes in this range give the best strength and ease of emplacement. The length of the probe can also vary, but typically 0.75 to 1.0 m (meter) is chosen for the depth from which samples are taken. This depth is one below which major influences from meteorological variables are found and one that frequently encounters the upper B or lower C soil horizon (Reimer, 1980; Hesselbom, 1985). The carbon alloy used is somewhat limited because of availability in the thick-walled tubing; usually the available grades are 1015, 1018, or 1020. Other

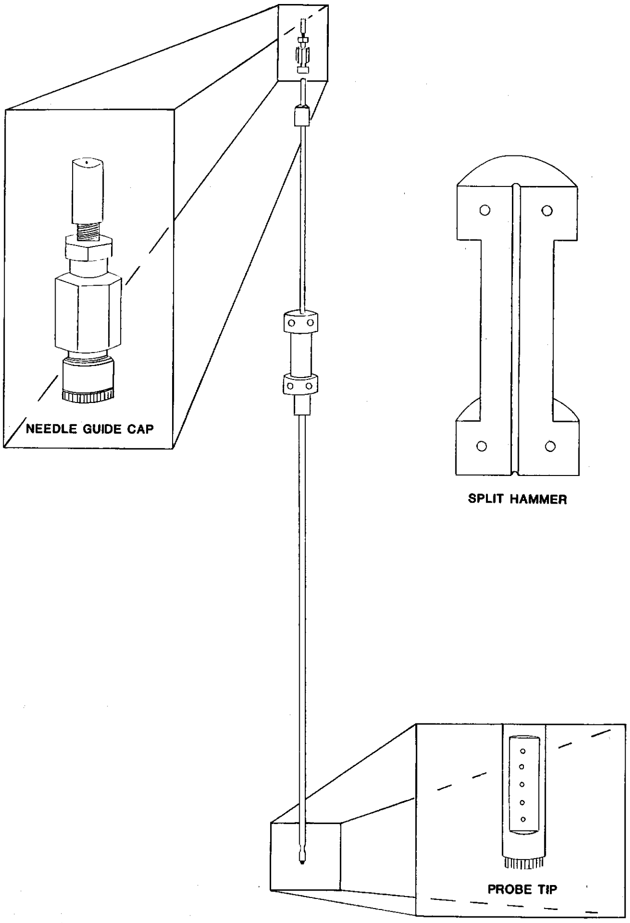

Figure 7. Soil-gas sampling probe showing the tip and needle guide components.

alloys would provide greater strength but are available only in quantities that make an acquisition expensive. Stainless steels are softer than the carbon alloys but are used for applications where a permanent probe for long-term monitoring is desired.

The probe is emplaced into the ground by pounding. To accomplish this, two pounding collars 3 cm (centimeters) in diameter and 5 cm in length are soldered onto the probe. They are separated by a distance of about 60 cm to permit a split, barbell-shaped deadweight to be used as a sliding hammer. The hammer is made from machined steel about 18 cm in length, 6 cm in diameter, and weighing about 2 kg (kilograms). The split shape allows it to be fitted over the probe, and it is held together with machine screws.

Many designs of the probe tip are possible, and two are described. In the first, a small machine screw is threaded into the lower end of the probe to prevent dirt from entering. There is no need to use a pointed end on small-diameter probes because several uses would quickly dull the end. About 2 cm above the tip, the opposite sides of the probe are ground flat by removing 0.5 mm of material for a 4-cm length to provide a zone of reduced diameter. Four or five sets of holes are drilled through this flat area; the diameter of the holes should be about equal to the inside diameter of the probe. A wire can be inserted into the probe to prevent dirt from entering the side holes. It serves to reduce the dead volume as well when collecting the sample but can be removed, if desired. A second probe tip design is one in which no flats or side holes are used. A rivet is put in the sampling end of the probe, and the probe is pounded into the ground. At the desired sampling depth, the probe is retracted a few centimeters by upward pounding. This action creates a cavity at the end of the probe. This particular design is sometimes useful for obtaining a sample when very high soil-moisture contents or fine silts are encountered. It also is a way to reutilize a side-hole probe on which the end has fatigued and broken.

The sample is taken from the open end of the probe. Tubing can be slipped over the end and a sample pumped to a detector, or an O-ring fitting containing a septum can be secured over the end to permit a discrete volume to be collected in a hypodermic syringe. Either extraction system should be able to purge the probe of air before the sample is collected. Because of the thick-walled steel, a 1-m probe has an internal volume of only 3 cm^3 (cubic centimeters), so extracting 10 cm^3 from it is sufficient for a thorough purge. Samples can be stored in the syringes as long as the needle is capped to prevent gas loss or exchange. Tests with helium have shown that the syringes are capable of retaining the gas for a period of several days before diffusion would cause dilution (Reimer and others, 1979). Fittings and tubes commonly are supplied in English dimensions. The tube size typically available is $^5/_{16}$ in (inch) outside diameter, 0.092 in inside diameter, and the O-ring fitting is $^1/_4$ in. The top end of the probe must be machined to a $^1/_4$-in diameter for the O-ring fitting.

Discussion

The probe design described above had been developed in 1974 and was subsequently modified for testing the possibility of sampling soil gases as a means of detecting energy resources. It has proven to be durable and reliable in a variety of soil environments. The small diameter causes minimal disturbance to the soil environment and provides self-sealing without the need for a packer system. The depth to which soil gases are sampled to minimize atmospheric influences is a matter of debate. Strength of the steel limits the practical length of the probe that can be pounded into the ground, usually to 1.5 m. For greater depths, a pilot hole could be made or the probe could be drilled into the ground. This basic design can be adapted to permit a number of modifications or alternate uses. For example, the choosing of the tip design, as mentioned previously, is such a modification, and determining the permeability from differential flow and pressure rates is an additional use. It has also been used for injecting gases in the soil for gas migration studies (Reimer, this volume).

WATER SAMPLE COLLECTING TECHNIQUE

Because natural concentrations of radon in water extend over many orders of magnitude (Graves, 1987), precise analyses are not necessary to categorize the general distribution of radon. A method has been developed for the rapid extraction and determination of the radon concentration in water. This technique is an ideal screening method for field operations where the qualitative determination of the radon concentration is acceptable and the results are desired at the field sampling site. Obtaining a representative sample is not a trivial procedure, however. The method described here involves collecting a sample that has been exposed to the atmosphere, such as from a spring or from ground water that has been pumped from a well. Agitation or natural effervescence can strip gases from the water. When water is obtained from wells, the sample should be secured from the water line before it has entered a pressurized holding tank. Even with the greatest of care, some gas loss is probably inevitable, and the concentrations should be considered as minimum values.

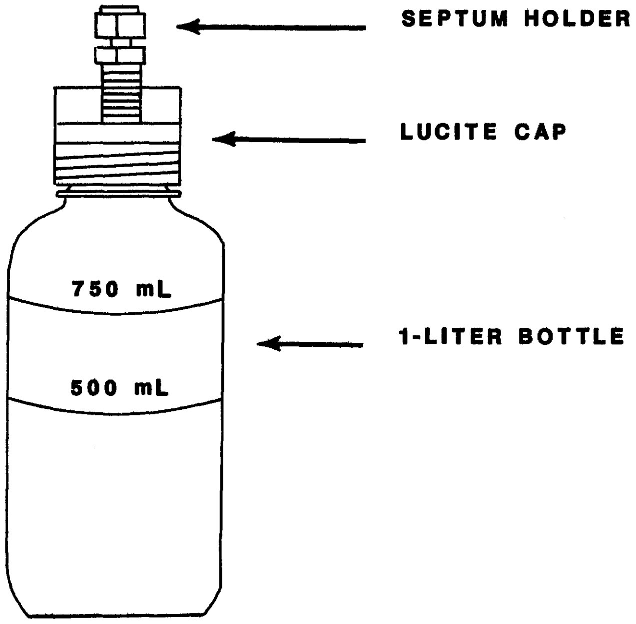

Figure 8. Water sampling container showing the cap and septum holder.

Description of Water Sample Collecting Equipment and Technique

The equipment for gas extraction consists of a standard 1-L (liter) plastic bottle and a modified cap (fig. 8). The cap is constructed of a hard plastic and is machined and threaded to fit the top of the bottle. The only critical step in machining is to ensure that the cap creates a seal with the opening of the bottle and not the shoulder or neck of the bottle. The cap is also machined to accommodate a fitting that contains a septum through which a hypodermic needle can be inserted to extract a sample from the closed bottle.

The bottle should be marked in the fashion of a graduated flask or beaker so that various quantities of water introduced can be measured. Typically, water samples of 750 cm^3 are collected in the bottle. The exact quantity is not critical, and volumes within 30 cm^3 of this nominal amount are sufficient. Continuous addition or removal of water to achieve the desired volume should be avoided because turbulence causes degassing of the water. The ratio of the water to headspace can be determined later. The bottle is then capped and shaken vigorously for 30 seconds. The agitation strips the dissolved gases from the water into the headspace air of the bottle. The bottle should then stand for 2 minutes to allow most of the bubbles in the water to combine with the headspace air. A hypodermic syringe is inserted through the septum on the cap, the bottle is squeezed to force some headspace air into the syringe, and the syringe is then removed and the needle capped. The quantity of gas introduced into the syringe can be selected to suit the type of analytical equipment used and the required accuracy of the measurement. Typically, sample sizes in the range of 10 to 50 cm^3 are sufficient. The sample can be analyzed immediately after collection if a field-portable instrument is available. If, indeed, the analysis can be performed in the field, a short delay time, less than 1 or 2 minutes, from collection to analysis, usually permits ^{220}Rn (thoron) to be analyzed.

Field analytical instruments are commonly alpha scintillometers that contain a phosphor-coated cell. The cells should be sealed and capable of holding a vacuum. Evacuating the cell to a fixed pressure before sample injection permits a small sample to be quantitatively introduced and, in turn, minimizes contamination of the cell with the radon daughters, especially when high concentrations of radon are encountered. Such a system has been described by Reimer (1977). It allows the repeated use of a single cell for an extended period before the cell background becomes significant. Each scintillometer should be calibrated, and the efficiency of the gas extraction system should also be compared to a standard such as can be prepared from a known amount of radium salts dissolved in water. The concentration of the radon in the sample is directly related to the quantity of water collected and the output of the analytical device. For first-order approximations of the concentration, even the temperature of the water can be ignored but should be considered for the final determinations.

Summary

The sampling system described here is a simple version and is based on the same principle of the bubbler technique commonly used to strip gases from liquids. The advantages of this system are its low cost, simplicity of use, rapid availability of the results, ease of modification for collecting waters from a variety of sources, and the capability to analyze for dissolved thoron. The primary disadvantage is the sacrifice of analytical precision and accuracy to limits of perhaps 20 percent. This disadvantage is usually not a restrictive factor as long as order-of-magnitude variations are considered sufficient resolution.

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A PRELIMINARY EVALUATION OF ENVIRONMENTAL FACTORS INFLUENCING DAY-TO-DAY AND SEASONAL SOIL-GAS RADON CONCENTRATIONS

Sigrid Asher-Bolinder, Douglass E. Owen, and R. Randall Schumann

Abstract

A long-term study of radon concentrations in a smectitic soil of the semiarid Colorado Piedmont reveals that interaction of meteorologic, climatic, and pedologic factors affects radon concentrations on both a daily and seasonal time scale. Seasonal variations are generally of greater magnitude than day-to-day fluctuations. Day-to-day radon fluctuations in this soil are controlled largely by changes in barometric pressure and by shallow capping effects resulting from precipitation. Change in gas permeability due to variations in soil moisture is the major control on seasonal soil-gas radon variation. Radon concentrations correlate with wind speed, relative humidity, air and soil temperatures, and the difference between those temperatures. Because these factors are interrelated, it is difficult to determine the magnitudes of their individual influences. Therefore, the nature of a soil and its response to climate must be understood if short-term measurements are to be used to predict long-term ranges of soil-gas radon concentrations.

INTRODUCTION

Soil-gas radon concentrations at a given locale are controlled largely by the interaction of geologic, pedologic, climatic, and meteorologic factors through time. Field studies (Gundersen and others, 1988; Schumann and Owen, 1988) have shown that a strong relation exists between geology and radon in soil gas. Field measurements for such studies are usually carried out within a few weeks or one season, so measurements are not affected by seasonal variations. From site to site, however, mineralogy, chemistry, structure, and moisture content of soil affect radon levels in soils developed on a single geologic unit. In addition, radon levels measured at a site in July are not necessarily predictive of radon levels in January. If prediction of the yearly range of soil-gas radon values from short-term sampling is to become possible, the soil and climatic factors controlling soil-gas radon must be understood adequately.

This report provides a preliminary interpretation of climatic, soil moisture, and meteorologic controls on radon-222 concentrations in soil gas in the semiarid environment of the Colorado Piedmont. We did not distinguish between the radon-generation and radon-transport *processes* that influence radon concentrations in the soil gas at the site, but we have attempted to identify the day-to-day (short-term) and seasonal (long-term) *controls* on those measured soil-gas radon concentrations. To identify and describe these controls, climatic, moisture, and weather effects on soils developed in this geologic setting were measured and studied.

THE IMPORTANCE OF TIME SCALES

In assessing the effects of climatic and weather-related factors on soil-gas radon concentrations, it is important to note that different factors are dominant, depending on the time scale under consideration. In this discussion we will consider three time scales: geologic time, seasonal time, and daily time.

Geologic time is here defined as the time period during which soil-forming processes take place. These processes commonly require hundreds to thousands of years. On this scale, climatic and geologic characteristics are of primary importance in determining the amount and distribution of radon source material (radium), particle size and distribution, emanating power, porosity, moisture retention characteristics, whether or not leached mineral horizons or soil-grain coatings are present, and other weathering characteristics that control soil-gas radon concentrations. Because our observations span a short period of geologic time, these factors can be assumed to be constant for the purposes of this study.

Seasonal time refers to one complete cycle of seasonal changes, generally equivalent to 1 year. Shorter term (day-to-day) fluctuations may be superimposed on the larger scale seasonal variations that occur during the span of a year. Likewise, the daily time scale refers to changes that can take place over time periods of less than an hour to several days. This is the period of time during which weather phenomena including precipitation, barometric and temperature changes, and wind effects take place. Changes that occur during daily time may be referred to as day-to-day fluctuations.

The concept of daily time scales, as used in this paper, must not be confused with the concept of diurnal variation. Diurnal variations in soil-gas radon concentrations have been noted by several researchers (Wilkening and Hand, 1960; Tanner, 1964; Schery and others, 1984) and are related to the regular variations in air and soil temperature, air pressure, and humidity caused by insolation during day-night cycles. However, because this phenomenon is relatively well known and well documented, we focused our investigation on identifying and describing those factors that cause day-to-day fluctuations and seasonal variations in soil-gas radon concentrations.

PREVIOUS STUDIES

Although geologic factors are primary controls of radon concentrations in soil gas, there are a number of climatic and meteorological controls as well. A survey of published literature indicates that the dominant nongeologic variables influencing radon soil-gas concentration are soil moisture, precipitation, barometric pressure, soil temperature, air temperature, wind, and capping effects. A summary of the historical findings on the effects of each of these factors follows.

Soil Moisture and Precipitation

Previous investigators observed that soil moisture has a major impact on observed soil-gas radon concentrations. Radon emanation rates increase with an increase in soil moisture of up to 15 to 17 percent by weight; at greater moistures, the observed emanation rate decreases (Damkjaer and Korsbeck, 1985; Lindmark and Rosen, 1985). Water is the most important agent in enabling radon to escape from solid material, because the water surrounding the soil particles absorbs the kinetic energy of the radon atom's recoil and prevents the radon atom from burying itself in an adjacent soil grain (Tanner, 1980). Further increases in soil moisture beyond 15 to 17 percent cause decreasing diffusion lengths of the radon atoms, because they must travel through thicker films of pore water (Damkjaer and Korsbeck, 1985; Lindmark and Rosen, 1985).

Lindmark and Rosen (1985) found that a twofold to fivefold increase in soil-gas radon concentration was common between dry and optimal (~15 percent) water content, but they observed an increase as great as 30 times in some soils. Baranov and Novitskaya (1960; cited by Mattsson, 1970) observed an increase in emanation of radon from uranium minerals when the humidity of the air increased.

Barometric Pressure

Low barometric pressure causes an increase in soil-gas radon concentration in the upper few meters of soil. Low pressure causes soil gas to rise toward the surface from greater depths, where the soil-gas radon concentrations are generally higher because of less dilution by atmospheric air. High pressure has the opposite effect and pushes atmospheric air into the ground, diluting the soil gas (Kovach, 1945; Kraner and others, 1964; Miller and Ostle, 1973; Klusman and Webster, 1981; Jaacks, 1984; Schery and others, 1984; Hesselbom, 1985; Lindmark and Rosen, 1985).

Kraner and others (1964) noted barometric influences on soil-gas radon concentrations to a depth of 96 in (inches). Kovach (1945) observed variations caused by barometric pressure to a depth of 2 m (meters). Jeter and others (1977) calculated that influences of barometric pressure would be noticeable at a depth of 20 m in highly permeable material and that these fluctuations would be smaller than, and lag behind, those at shallower depths. Holkko (1987) found that changes in barometric pressure can cause convective flow of soil gases. Barometrically induced soil-gas motion may be equally as effective as molecular diffusion in transporting radon in soil (Robertson, 1969). Bakulin (1971) found that a decrease in pressure increases radon exhalation "proportional to the square of the pressure drop rate, [and to the] square of the gas permeable soil layer depth, and [it] increases linearly with time." Clements and Wilkening (1974) found that pressure changes of 1 to 2 percent associated with the passage of weather fronts could produce changes of 20 to 60 percent in the radon flux, depending on the rate of change of pressure and its duration.

Soil Temperature and Air Temperature

Observations concerning the effect of soil temperature on radon concentrations conflict. Some authors suggest that soil temperature has little or no effect on soil-gas radon content (Kovach, 1945; Lindmark and Rosen, 1985). However, according to Ball and others (1983), "Radon concentration correlates with soil temperature and to a lesser extent with air temperature." Kovach (1945) observed higher radon emanation during temperature lows. Jaacks (1984) observed negative correlations between both soil and air temperature and radon concentrations and noted that temperature gradients within the soil, or between the soil and air, can induce convective soil-gas transport.

The conflicting observations of temperature effects may be due in part to the complexity of the relationships between meteorologic factors. A particular temperature change may be accompanied by changes between dry and wet conditions, a stable or unstable pressure regime, or variable winds. Thus, the effects of temperature may be difficult to separate from the effects of other factors that exert a greater influence on the soil-gas radon concentration.

Wind

Gusting winds cause a decrease in soil-gas radon concentrations because soil gas is being diluted or removed at the surface (Smyth, 1912; Kovach, 1945; Kraner and others, 1964; Miller and Ostle, 1973; Jaacks, 1984; Hesselbom, 1985; Lindmark and Rosen, 1985). Kovach (1945) observed wind effects to a depth of 150 cm (centimeters). Kraner and others (1964) noted wind effects at 70 in but none at 90 in. In addition to wind velocity, the following factors affect the magnitude and depth to which the wind can exert an influence: soil permeability, the amount of moisture in the soil, the type and amount of ground cover, extent of snow and ice cover, and water frozen in the ground. Holkko (1987) suggests that winds can cause soil-gas flow in coarse-grained soil. Hesselbom (1985) and Pearson and Jones (1966) attribute the pumping effect of the wind to its turbulence or to fluctuations in wind speed. Wind turbulence can be caused by updrafts and downdrafts produced by atmospheric convection or by wind shear (Neiburger and others, 1973). The turbulence and fluctuations cause pumping by alternating between pressurization and depressurization of the soil. This effect is similar to that caused by barometric pressure changes. Venturi effects may draw gases out of the soil when the wind blows across desiccation cracks or open pores.

Capping

High soil moisture can lead to the swelling of expandable lattice clays (smectites), and this swelling lowers the permeability of the soil in those zones. As pores fill with water, the gas permeability is progressively reduced. As the top layer of the soil becomes saturated, soil-gas flow is restricted, and a cap that prevents mixing of soil gas with the atmosphere forms. This capping may reduce or block the effects of barometric pressure or wind on soil-gas radon concentrations. Kraner and others (1964) found that heavy precipitation could produce a cap that caused radon concentrations to reach levels in the near surface equal to those found at greater depths.

Also, water frozen within the soil can produce an effective cap that retards radon flux to the atmosphere; an increase in soil-gas radon concentrations results (Kovach, 1946; Jaacks, 1984; Hesselbom, 1985; Lindmark and Rosen, 1985; Taipale and Winqvist, 1985). Kovach (1945), for instance, found that the highest soil-gas radon concentrations of the year occur when the ground is frozen, an example of the effectiveness of capping in the upper layers of the soil.

SITE DESCRIPTION

A long-term radon monitoring site was established on the grounds of the Denver Federal Center in Lakewood, Colo. (fig. 9). The site is located on flatlying, undisturbed, and unirrigated land subject only to the foot traffic of the investigators and a twice-yearly mowing of the sparse grasses that constitute the ground cover. The site is typical of many areas of the Western United States on which housing is constructed. The soil developed on a lower terrace or pediment deposit of the Verdos Alluvium (Lindvall, 1978). This Pleistocene alluvium is derived largely from the weathering and transport of material from the Denver and Arapahoe Formations, which, in turn, were derived from granitic and gneissic terrane. The soil, a Denver-Urban land complex, "formed in calcareous clayey material derived from mudstone and shale" (Soil Conservation Service, 1983, p. 44) and is smectitic, hard, and pebbly; it contains calcium carbonate streaks and is subject to deep cracking when dry. "Permeability of this Denver soil is slow. The available water capacity is high. . . . The shrink-swell potential is high. Rock fragments make up 0 to 15 percent of the volume" (SCS, 1983, p. 45). Denver series soils commonly have a "well developed argillic B horizon and commonly have secondary accumulations of calcium carbonate, which indicate relative stability in respect to erosion and to further deposition. This parent material was deposited primarily as transported mantle over older surfaces as alluvium, colluvium, and loess" (SCS, 1983, p. 234).

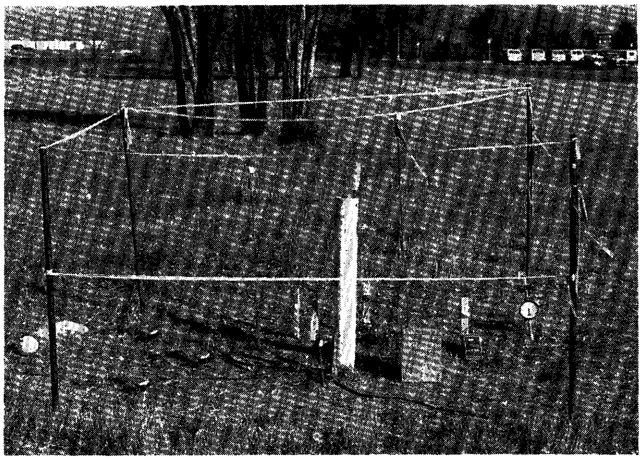

Figure 9. Study site at the Denver Federal Center. Three soil-gas probes are in the center; the 100-cm probe is encased in PVC pipe.

The site is 1,715 m above sea level and is located in the semiarid, temperate, continental climatic zone (Hansen and others, 1978). The area of the study site has a mean low January temperature of −9 to −7 °C (degrees Celsius) and has a mean high July temperature of 23 to 27 °C. Mean dates of first and last frosts are October 4 and May 11. Mean annual precipitation is 36 to 41 cm, including 125 to 140 cm of snowfall, and mean annual evaporation is 125 to 150 cm. About 70 percent of the annual precipitation falls between April and September. Mean annual relative humidity is 50 to 55 percent. Sixty to 62 percent of the days are defined as clear (less than 30 percent of the sky is cloud covered). Wind speeds of 3.5 to 5.5 meters per second occur more than 35 percent of the time in the Denver area, with highest velocities occurring in May and November. In summary, there are "strong seasonal variations for most weather elements" (Hansen and others, 1978, p. 58) in the area that includes this study site.

METHODS

Soil gas was generally collected once each weekday from stainless steel probes permanently installed at 50, 75, and 100 cm below the study site's soil surface. The technique and equipment described by Schumann and Owen (1988) were used. During the first 3 months of the study, only the 100-cm probe was installed. The 50- and 75-cm probes were later installed to provide a more complete radon-depth profile. Soil gas was usually collected within the time interval of 10 o'clock a.m. to 2 o'clock p.m. to limit diurnal effects on radon concentrations. Radon concentrations in the soil gas were measured by use of an EDA RDA–200 radon detector, and the counts recorded by the instrument were converted to picocuries per liter (pCi/L) by use of a calibration equation. Barometric pressure, soil temperature at 60 cm, air temperature in the shade 5 to 10 cm above the soil surface, wind speed and direction, and soil-gas extraction times were measured also. The extraction times represent an approximation of gas permeability at each depth. Less quantifiable factors that were noted were cloud cover, recent rain, snow cover, snowmelt, standing water, and whether or not a frozen or water-saturated layer of soil was present at the surface. Width and depth of soil cracks were recorded also; the cracks add a significant amount of permeability to the soil and indicate the extent of soil drying.

Our soil-gas radon and meteorological measurements were integrated with meteorological data collected by the U.S. Bureau of Reclamation's weather station about 0.4 km away. The radon measurements were inserted into the data base at the nearest whole hour for comparison with weather station data. However, all 24 daily weather measurements were used for interpretation of the radon data.

DISCUSSION

Soil-gas radon concentrations vary progressively and markedly with season. A progressive drop in soil-gas radon concentration at 100 cm can be seen in a plot of radon concentration over time (fig. 10). Radon concentrations were highest from March through June. Spring was the wettest period of the year in the study area and was characterized by unstable weather, with large fluctuations in air temperature, barometric pressure, relative humidity, and wind. During early spring, much of the precipitation fell in the form of wet snows that melted quickly and thus saturated the shallow soil layers and maintained a relatively consistent moisture cap that kept soil-gas radon concentrations elevated (fig. 10). The moisture cap was commonly frozen when nighttime temperatures dropped below freezing for extended periods of time. The soil-gas radon concentrations reached a minimum in August, and, except where affected by short-term weather events, remained low through the remainder of the year as a result of the relatively dry, stable weather conditions that existed during this period.

Almost 75 percent of the precipitation recorded during the study period fell between March and mid-July (fig. 10). Because temperatures were cooler and precipitation events were relatively evenly spaced during this period, the soil surface remained damp much of the time, and a moisture cap was apparently maintained until about mid-June. The importance of moisture in maintaining the cap, especially in the semiarid climate typical of much of the Western United States, is illustrated by the drop in radon concentrations that occurred during the first part of April, an approximately 2-week period of fair, windy weather during which there was little precipitation, and daytime temperatures were generally above freezing. The surface of the soil began to dry, and surficial desiccation cracks appeared. Soil-gas radon concentrations rose again immediately following the next precipitation event. In soils with smectitic (swelling) clays such as those at the study site, less moisture may be needed to create and maintain capping conditions.

Between mid-June and mid-July, the warmer daytime temperatures allowed the soil to dry between rains. As a result, the moisture cap was depleted, and exchange of soil gas with atmospheric air occurred. During summer and fall, more complete drying occurred, and desiccation cracks up to 1 cm wide and 45 cm deep appeared in the soil surrounding the study site. From August through September, a relatively free exchange between atmospheric air and soil gas in the near-surface layers was possible, and soil-gas radon concentrations at both 50 and 100 cm were at their lowest. A slight increase in soil-gas radon concentrations at 50 and 100 cm at the beginning of November coincided with a drop in daily air and soil temperatures and greater fluctuations in barometric pressure. These higher radon levels were maintained through the remainder of the study period.

In general, the highest radon concentrations occurred during wetter seasons. In the Denver area, this period, late winter through spring, is typically one of unsettled, highly variable weather in which snowfall and freezing temperatures may be followed the next day by warm, sunny weather, which tends to maximize percolation of snowmelt into the soil. In areas that have distinct wet and dry seasons, soil-gas radon values may vary by an order of magnitude or more. In areas that have wetter climates, radon values may be generally higher most of the year, except during periods of complete soil saturation.

Whereas the major long-term trends in soil-gas radon concentrations are related to seasonal climatic variation, the shorter term, usually lower magnitude, fluctuations are related to changes in weather—storms, frontal passages, and movements of air masses. The dominant weather factors influencing day-to-day radon concentrations are

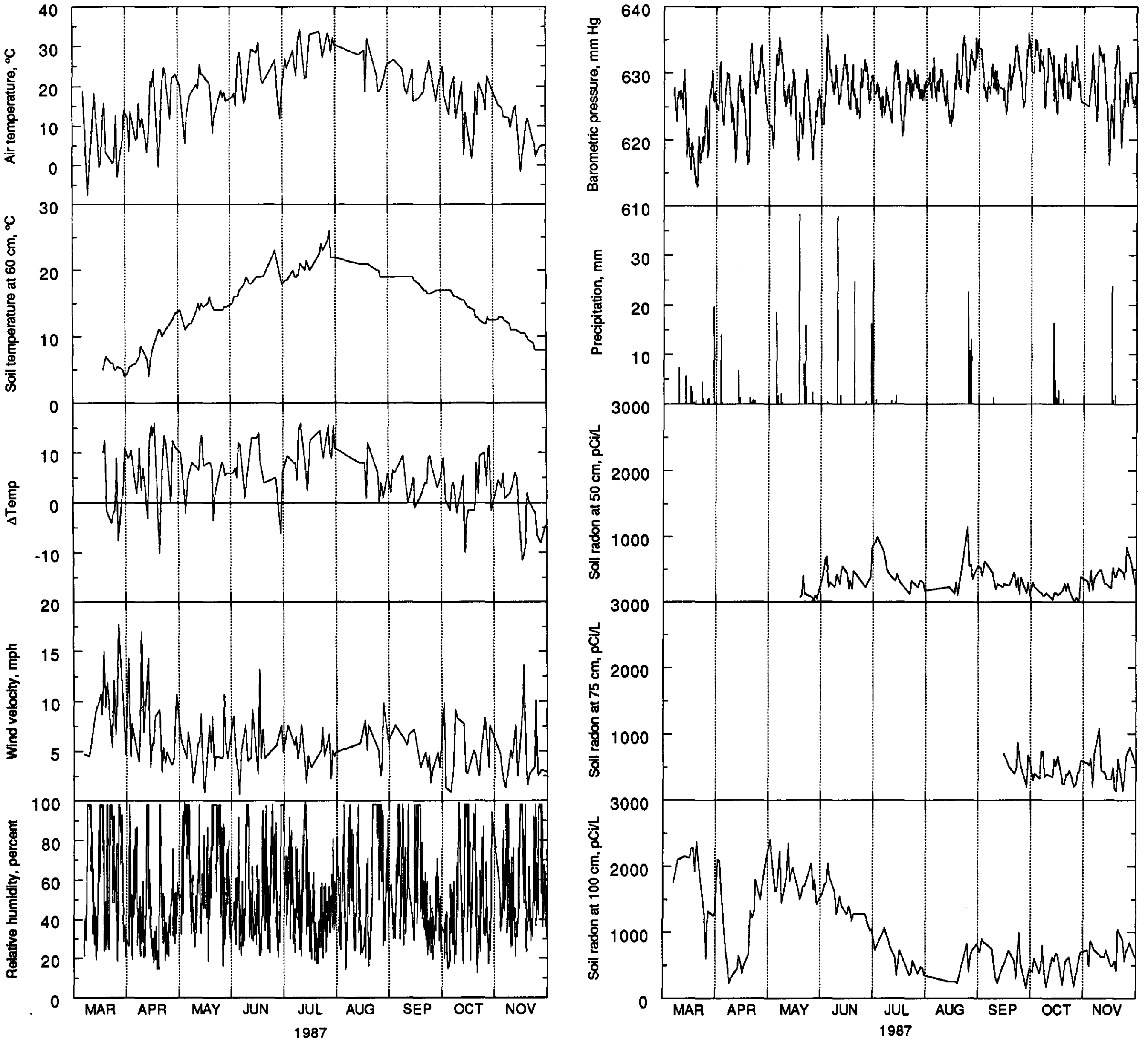

Figure 10. Weather parameters and radon in soil gas, Denver Federal Center study site. ΔTemp, delta temperature—a positive value when the air temperature is greater than the soil temperature and a negative value when the soil is warmer than the air; °C, degrees Celsius; mph, miles per hour; mm Hg, millimeters of mercury; pCi/L, picocuries per liter.

barometric pressure changes and precipitation. An increase in barometric pressure is usually accompanied by, or followed by, a decrease in radon concentrations. It appears that the actual value of the barometric pressure is less important than whether a change has occurred. The magnitude and direction of the change determine the extent of the effect on soil-gas radon concentrations. It is important to note that effects of barometric pressure are stronger and more pronounced in highly permeable soils. The soils in the study area have a relatively low gas permeability (between 10^{-8} and 10^{-9} cm^2, measured at 100 cm; R.T. Peake, oral commun., 1987), so that the effect of barometric pressure is probably somewhat inhibited; nevertheless, the data suggest that barometric pressure is one of the two primary influences on daily radon fluctuations. Much of this effect may be due to the periodic, and sometimes prolonged, presence of numerous desiccation cracks in the soil that considerably enhanced the effective permeability of the soil. A lag of from several hours to over a day between changes in barometric pressure and corresponding changes in soil-gas radon concentrations was noted (fig. 10).

Precipitation, and its effect on soil moisture, is the second major cause of day-to-day fluctuations in soil-gas radon concentrations. A comparison of precipitation and radon concentration at 100 cm (fig. 10) shows that the elevated soil-gas radon concentrations encountered during the spring and early summer continued as long as precipitation was sufficient to maintain a surface capping layer in the soil. When a prolonged dry period was encountered at the beginning of July, soil-gas radon concentrations dropped (fig. 10). Two of the three precipitation events in the latter part of the study period, the one near the end of August and the one in mid-November, correspond to increases in radon concentrations in the soil at both 50 and 100 cm. The precipitation in mid-October does not appear to have had a noticeable effect on the soil radon level at 100 cm, and just a slight increase in the radon level at 50 cm followed the event. Perhaps the total amount of rainfall from that storm, about 20 mm, was insufficient to completely saturate the surface soil layers; this is a likely explanation in light of the almost 2 months of dry weather that preceded it. In contrast, the order-of-magnitude increase in radon concentration at 100 cm that followed the rainfall in early May suggests that the amount of soil moisture in the near-surface layers was just below a threshold level for forming a moisture cap during late April, and this threshold was exceeded with the additional input of precipitation in early May (fig. 10). These examples point out the importance of antecedent conditions in determining the magnitude of the effects of weather on soil-gas concentrations.

Changes in relative humidity appear to correlate fairly well with changes in soil-gas radon concentrations, but this correlation is because nearly every large peak on the relative humidity plot (fig. 10) corresponds to a precipitation event. Therefore, although a positive correlation exists between relative humidity and soil-gas radon concentrations, it is of a secondary nature, as the relative humidity values are generally indicators of precipitation when examined in the context of day-to-day fluctuations.

Wind may have a less pronounced effect on the concentration and movement of soil-gas radon at the study site. A correlation between winds and soil-gas radon concentrations could not be positively identified. There are three possible explanations for this lack of correlation: (1) as discussed previously, the soil may not have been permeable enough to allow free exchange of atmospheric and soil air, (2) high winds usually preceded or accompanied storms, and the concurrent changes in barometric pressure, temperature, and soil moisture may have masked any wind effects, or (3) no identifiable correlation exists between winds and radon movement in soils.

Air- and soil-temperature data show a seasonal negative correlation with soil-gas radon values. Radon values in the soil at the study site were highest during winter and spring, when temperatures were cooler (fig. 10). Day-to-day fluctuations in air temperature also show a negative correlation, because cooler-than-normal air temperatures usually result from storms. Note that all unseasonably low daily air temperatures correspond to low barometric pressures and precipitation.

Temperature may exert an influence on soil-gas radon concentrations at the study site. However, the effect of temperature appears to be masked by pressure changes and precipitation that accompany storms. For example, a drop in soil or air temperature may accompany rainfall that effects a cap responsible for higher soil-gas radon levels. Although temperature would have a negative correlation with radon concentration, it would be difficult to determine a cause-and-effect relationship. In contrast, radon concentration could correlate positively with temperature when a high pressure system with attendant lower temperatures moves into an area, and the higher barometric pressure forces atmospheric air into the ground, where it dilutes the soil gas and reduces soil-gas radon concentrations near the surface. This positive correlation of radon with temperature could be misleading, because it may be coincident with another event.

Because soil acts as a heat sink, fluctuations in soil temperature are small and gradual; thus, no identifiable correlation exists between soil temperature (at 60 cm) and soil-gas radon concentrations in the context of daily time. However, in highly permeable soils, temperature-induced convection could enhance radon transport to a considerable degree (Kraner and others, 1964; Mogro-Campero and Fleischer, 1976; Jaacks, 1984). A plot showing the difference between air temperature at the surface and soil temperature at 60 cm indicates those periods when conditions are favorable for temperature-induced convective transport (fig. 10). This difference, which we refer to as "delta temperature," or "ΔTemp," has a positive value when the air temperature is greater than the soil temperature and a negative value when the soil is warmer than the air. The sign (+or−) of ΔTemp indicates the direction of temperature gradients between soil air and atmospheric air—upward when ΔTemp is positive, downward when it is negative. Although a correlation exists between ΔTemp and changes in soil-gas radon concentrations, this may be because ΔTemp is a derivative of the more basic factors of soil and air temperature.

In an attempt to quantify the relationships between weather factors and soil-gas radon concentrations, linear correlation coefficients were calculated for each of the parameters in the data set (table 3). These correlations are defined as significant at the 95-percent confidence level. As shown in table 3, the radon concentrations at 100 cm show significant negative correlations with "month/day" (indicating that the highest radon values occur seasonally) and with "barometric pressure" and "soil temperature." The lack of significant correlations between radon at 50 and 75 cm and barometric pressure may be explained by the fact that fewer measurements were made of radon concentrations at these depths, so the data are not sufficient to establish significant correlations.

Significant positive correlations between radon concentrations at 100 cm and both ΔTemp and relative humid-

Table 3. Linear correlation coefficient matrix of weather and radon parameters at the Denver Federal Center study site

[Values in boldface are significant at a 95-percent confidence level; number of pairs for each correlation is equal to the smaller number of sample factors in each pair; ΔTemp, difference between air temperature at the surface and soil temperature at 60 cm]

	Number of samples	Month/ day	Barometric pressure	Air temper- ature	Soil temper- ature	ΔTemp	Relative humidity	Wind speed	Precipi- tation	Radon at 50 cm	Radon at 75 cm	Radon at 100 cm
Month/day	156	1.000	**0.358**	0.076	**0.293**	**−0.329**	**−0.162**	**−0.340**	−0.104	−0.092	−0.109	**−0.642**
Barometric pressure	152		1.000	**.162**	**.306**	−.084	−.067	**−.492**	−.025	.078	−.018	**−.342**
Air temperature	152			1.000	**.746**	**.779**	**−.561**	**−.161**	**−.166**	−.001	.106	−.126
Soil temperature	150				1.000	**.235**	−.015	**−.246**	.021	.061	.121	**−.231**
ΔTemp	149					1.000	**−.610**	−.019	**−.169**	−.080	−.055	**.177**
Relative humidity	152						1.000	.019	**.388**	.151	.011	**.257**
Wind speed	152							1.000	−.003	−.047	−.151	.130
Precipitation	156								1.000	**.230**	−.123	.134
Radon at 50 cm	109									1.000	**.297**	.121
Radon at 75 cm	50										1.000	**.418**
Radon at 100 cm	153											1.000

ity exist. Both of these correlations appear to illustrate a common problem in the interpretation of correlation data: "Frequently, two variables may appear to be highly correlated when, in fact, they are not directly associated with each other but are both highly correlated with a third variable" (Huntsberger and Billingsley, 1981, p. 396). ΔTemp is calculated from air and soil temperatures, as discussed previously. Relative humidity acts as an indicator of precipitation and is statistically more significant than precipitation itself, because relative humidity generally increases to a level in the range of 80 to 100 percent during and after rains, whereas precipitation can occur as one large magnitude event or as a number of closely spaced smaller events, both having a similar effect on soil moisture but not on statistical correlations.

Radon concentrations at 100 cm also correlate positively with radon concentrations at 75 cm but not with radon at 50 cm. Radon at 75 cm correlates with radon at both 50 and 100 cm. These correlations indicate that radon concentrations at 50 cm and 100 cm vary in a dissimilar manner (at least statistically) and that conditions at 75 cm are transitional between those at shallower and greater depths. Radon concentrations at 50 cm also correlate significantly with precipitation; however, the lack of any other significant correlations with radon at either 50 or 75 cm is disturbing, because the soil gas nearest the surface is expected to be most sensitive to meteorologic effects. A possible explanation for this disparity is that the variability in radon concentrations at depths shallower than 100 cm is due to the complex interaction of a number of factors that cannot be separated statistically, at least by the method of statistical analysis employed here.

Table 4 summarizes what we consider to be the fundamental factors influencing radon concentrations in soil gas, considered in the context of different time scales. The table was developed from our analysis of this study's data and from our interpretations of previous researchers' work. These relationships should be generally applicable in any geologic, pedologic, or climatic setting. A positive influence (signified by a +) indicates that an increase in the factor correlates with an increase in soil-gas radon concentrations. In contrast, a negative influence (indicated by a −) means that an increase in that factor correlates with a decrease in soil-gas radon concentrations.

Note that we do not imply an implicit cause-and-effect relationship for each factor; the seemingly complex interaction of weather factors makes a definitive determination of cause and effect difficult. Theoretical discussions of the physical processes associated with weather and soil-gas radon generation and migration provide a foundation for understanding these relationships. However, empirical data as yet are not sufficient to definitively confirm and quantitatively describe those relationships.

Table 4. Summary of factors influencing soil-gas radon concentrations

[+, generally positive influence (that is, an increase in the factor generally causes an increase in soil-gas radon concentration); −, generally negative influence; ±, may have either positive or negative influence, because influence is interrelated with or dependent on other factors; blank indicates that the influence of that factor is not significant during that time scale]

Factor	Time scale of influence		
	Geologic	Seasonal	Daily
Radium content of soil	+		
Soil permeability[1]	+	+	+
Precipitation/soil moisture[2]		±	±
Barometric pressure		−	−
Wind speed			−
Air temperature		−	±
Soil temperature		−	±

[1] Effective permeability is influenced by variations in soil-moisture content.

[2] At the Denver Federal Center site, increasing soil moisture caused capping effects that increased soil-gas radon concentrations; in more permeable soils or soils with lower clay contents, however, high soil-moisture contents would generally cause soil-gas radon contents to decrease as soil pores become occluded with water.

SUMMARY

Statistical analysis and visual inspection of quantitative data from our study site suggest that no single factor controls radon concentrations on either a daily or a seasonal basis. However, precipitation, because it increases soil moisture, appears to be the most important control of seasonal soil-gas radon content and variation. On a day-to-day basis, however, weather factors such as barometric pressure, precipitation's capping effects, temperature, and winds interact to control radon fluctuations.

We have shown that radon concentrations in soil at a single site are variable through time and that these variations are due primarily to the effects of changes in meteorological variables. An understanding of the interactions of meteorologic variables with soil gases is necessary if short-term soil-gas sampling is to be used to predict long-term ranges of radon concentrations. In addition, because radon sampling programs typically consist of measurements taken at multiple sample sites, often covering several soil and rock units, an understanding of geologic and pedologic processes and parameters, as they relate to radon generation, concentration, and transport, is also necessary. Although geologic and meteorologic controls on soil-gas radon have often been studied separately, it is only by integration of these data that effective site or area evaluations can be made.

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DERIVATION OF RADON MIGRATION RATES IN THE SURFICIAL ENVIRONMENT BY USE OF HELIUM INJECTION EXPERIMENTS

G.M. Reimer

Abstract

Helium injection into the subsurface provides a means of determining the rate of radon migration in a particular area. The passage of a concentration front is monitored at outlying stations, and an effective diffusion coefficient can be determined. From experiments in a portion of the Reading Prong near Boyertown, Pa., it is possible to explain the observations with a model in which diffusion is the primary transport mechanism, and convective mass tranport is influential. This technique permits determination of both horizontal and vertical migration components. The rate of migration is highly variable from area to area, but measurements can set realistic limits on models being developed for assessing the potential for radon accumulation in dwellings.

INTRODUCTION

The rate of radon migration in the surficial environment can greatly influence the indoor radon concentration. The movement of radon through soils has been the subject of numerous theoretical and empirical evaluations (Tanner, 1964; Jeter, 1980; Wilkening, 1980; Kristiansson and Malmqvist, 1982; Nazaroff and others, 1985); yet, study continues in order to determine a value or develop a unified model to predict the rate. A major difficulty in designing a model is the highly complex environment that is being investigated and the interrelationship of all the variables that can affect the movement of gases. A general consensus has been reached, however, on the major influences, which are meteorologic, those pertaining to soil chemistry and physics, concentration and nature of the source, and hydrologic intervention. In any one fixed location, the rate of radon migration is variable with time because it depends on the relative "strengths" of the influences, which are themselves variable.

A technique that can be used to measure the migration of gases is the direct injection of gas into the subsurface and the monitoring of the passage of the gas at various distances from the injection site. This approach was used in a number of studies to trace underground migration pathways (Turk and others, 1968; Robertson, 1969a,b; Herring and others, 1985; Reimer and Been, 1985). This technique was used with radon by Bulashevich and Kartashov (1967) to determine the radon diffusion coefficient in different natural surroundings. They demonstrated that the radon diffusion coefficient could be estimated from the helium diffusion data. In this paper, diffusion refers to dynamic molecular diffusion, which does not require that equilibrium conditions be achieved. Convective transport refers to movement due to bulk flow that has been induced by other driving mechanisms such as changes in barometric pressure or temperature gradients active in the near-surface soils.

Injection methods have the unequaled advantage of being the most realistic for the in situ determinations but have the disadvantage of being time consuming if large areas are being evaluated. Helium has several advantages for determining diffusion coefficients in surficial settings. It is safe to handle, has the fastest rate of diffusion of the noble gases, and, with mobile equipment, can be detected at part-per-billion concentrations. A demonstration study was performed in an area near Boyertown, Pa., where indoor and soil-gas radon concentrations in adjacent homes differed by more than an order of magnitude. Helium was injected through probes pounded into the ground (Reimer and Bowles, 1979), and the mobile helium analyzer developed by the U.S. Geological Survey was used for analysis (Reimer and others, 1979). The helium analyzer has a sensitivity of 10 ppb (parts per billion) for helium.

STUDY SETTING

The site for injection in the Boyertown area was within soil overlying a localized mylonitic zone in a quartz-feldspar and biotite gneiss that had been identified by Gunderson and others (1987) as being the host rock for locally higher uranium concentrations. Homes built within this geologic unit had indoor radon concentrations in excess of the Environmental Protection Agency guideline of 4 pCi/L (picocuries per liter) (Environmental Protection Agency, 1986). Soil-gas radon concentrations in this zone were 5,000 to 10,000 pCi/L. At the sampling site, the mylonitic zone trends N. 70° E. and is about 40 m (meters) wide. A series of probes was set within the zone to sample both parallel and perpendicular to this trend. The injection probe was at a 2-m depth, and monitoring probes were at a 1-m depth. The measured rate would be a combination of horizontal and vertical migration components. The monitoring probes were 2, 4, and 6 m in distance from the injection probe (fig. 11). The soil was saprolitic, and the upper surface was reworked to an average depth of 0.2 m from the construction of the nearby residence. Some free moisture was present in the soil from moderate precipitation that had occurred 2 days earlier. The study ran for a period of 4 hours on August 19, 1986. At the beginning of the study, the sky was overcast, the air temperature was 25 °C (degrees Celsius), the barometric pressure was 94.0034 kPa (kilopascals) (705.202 mm Hg, millimeters of mercury),

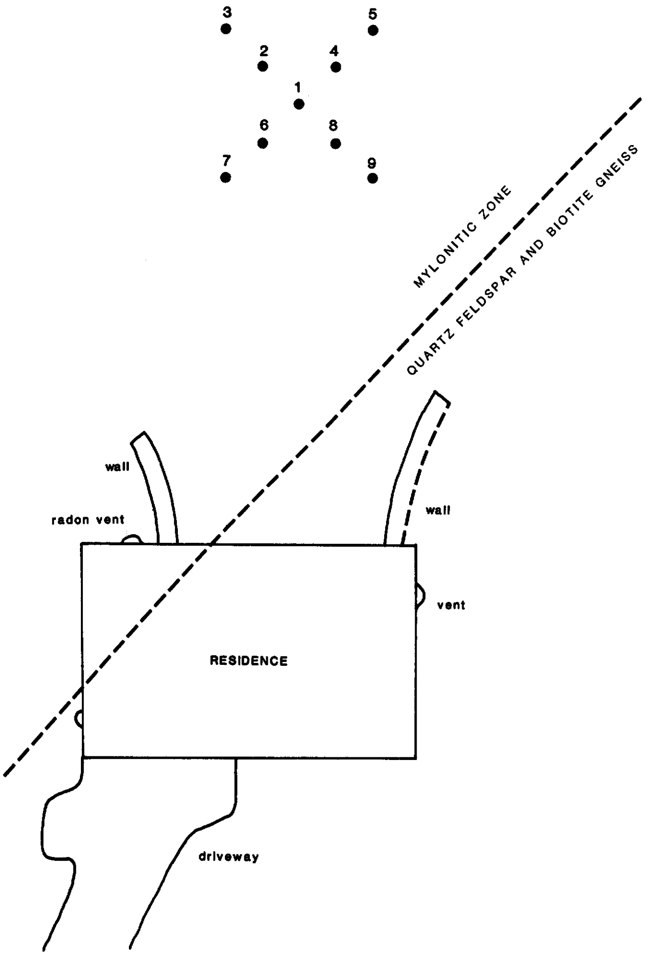

Figure 11. The location of the injection probe (1) and the monitoring probes (2–9) for the helium migration study. All probes are within the soils above the mylonitic zone. Even-numbered stations are 2 m distant and odd-numbered stations 4 m distant from the injection site. The zone trends N. 70° E., and the northern limit is approximately located by the dashed line.

and the wind was from the northwest at 8 to 15 km/h (kilometers per hour). Barometric pressure decreased by 0.0207 kPa (0.155 mm Hg), temperature increased to 27 °C during this period, and the wind remained the same throughout the study period.

A total of 14,000 cm^3 (cubic centimeters) STP (standard temperature and pressure) high-purity helium was injected during a 7-min (minute) period (a rate of 35 cm^3/s, cubic centimeters per second). A pressure monitor, having a maximum pressure limit of 175 kPa, was set on one of the monitoring probes 2 m distant. During the injection, the monitor showed no increase in pressure at the sensitivity limit of the pressure transducer, 2 parts in a million. Initially, samples were collected from every outlying probe. The sampling interval increased from a few minutes to 15 min after a response to the injected helium was observed at the closest monitoring probes.

RESULTS

Figures 12 to 19 show the results of the helium analyses. Even-numbered stations were 2 m distant and odd-numbered stations were 4 m distant from the injection site. All inner stations showed a response in about 30 to 45 min, and the outer stations responded in about 90 to 150 min, with the exception of stations 4 and 5. These 2 probes were located parallel to the mylonitic trend but topographically lower than the other probes. This area also contained the most soil moisture. In fact, probe 4 was relocated a few centimeters along the sampling pattern radius before injection because free water was drawn from it with the syringe when a pre-injection sample was collected. Gas was easily drawn from the remaining probes. Probe group 2 and 3 and group 8 and 9 showed a response slightly later than probe group 6 and 7. This delayed response may result from the lamination of the mylonitic zone that permits gas to migrate more freely in the N. 70° E. direction. In addition, stations 4 and 6 showed a decrease in helium after about 200 min that probably represents the passage of the maximum concentration available. The open circle on figures 12, 13, 18, and 19 is the helium soil-gas concentration 18 hours after injection. The inner stations showed a decrease, but the outer stations had higher values than they had the previous day.

DISCUSSION

Although the migration of gases in the soil is controlled by a number of complex and interrelated factors, it is possible to explain a large amount of the migration observed in this study by use of a simplified diffusion model. This approach is based on studies of environmental hazards presented by Thibodeaux (1979). It is a plausible model because the near-surface environment has high permeability and porosity. Because we are dealing with a dynamic system, diffusion in this model does not mean equilibrium conditions must be achieved. While diffusion can explain most of the observed migration rate, the remainder is probably from convective transport.

The helium injection represented a point source in both time and space. The sensitive analytical capabilities permit the advancing gradient to be detected. A helium background concentration for the area was determined by analyzing the soil gas for helium at each monitoring probe site. After injection, helium was detected at the 2-m monitoring stations in approximately 40 min and at the 4-m stations in approximately 160 min. In the presence of a concentration difference, there appears to be an r^2 function, or mean square displacement, for the time-distance relationship, where r is the radius of the system being evaluated. From the helium data, it is possible to develop a mathematical model using Fick's second law to describe the rate of molecular diffusion or penetration in a stagnant air mass and

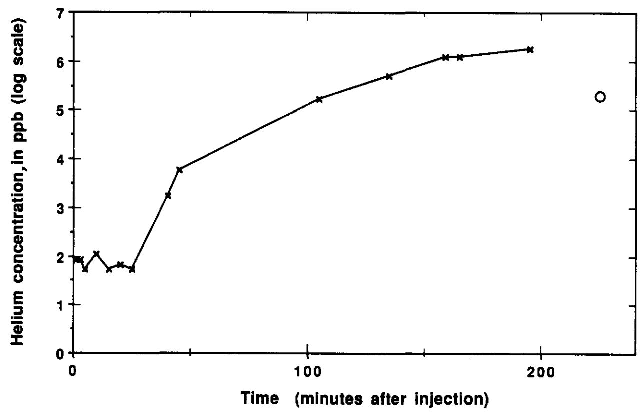

Figure 12. Variation in soil-gas helium concentration at station 2 as a function of time after injection at probe site 1. Open circle is the helium soil-gas concentration 18 hours after injection. ppb, parts per billion (figs. 12 through 19).

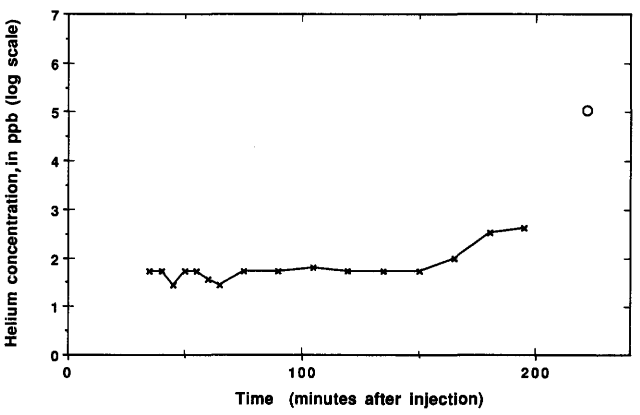

Figure 13. Variation in soil-gas helium concentration at station 3 as a function of time after injection at probe site 1. Open circle is the helium soil-gas concentration 18 hours after injection.

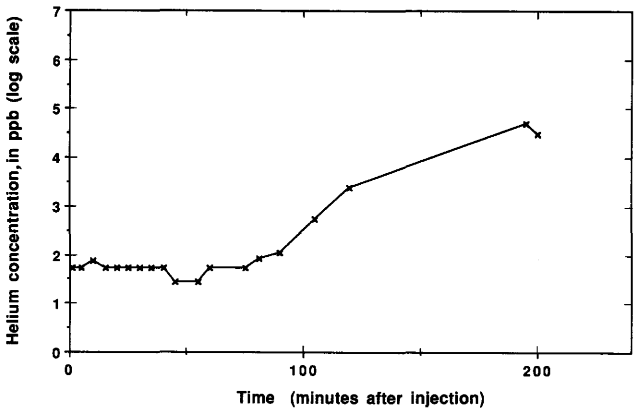

Figure 14. Variation in soil-gas helium concentration at station 4 as a function of time after injection at probe site 1.

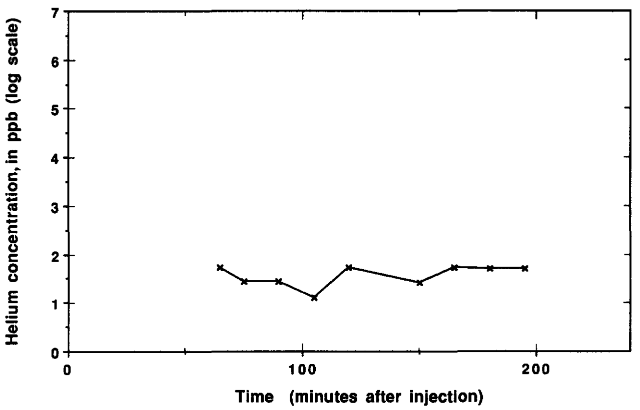

Figure 15. Variation in soil-gas helium concentration at station 5 as a function of time after injection at probe site 1.

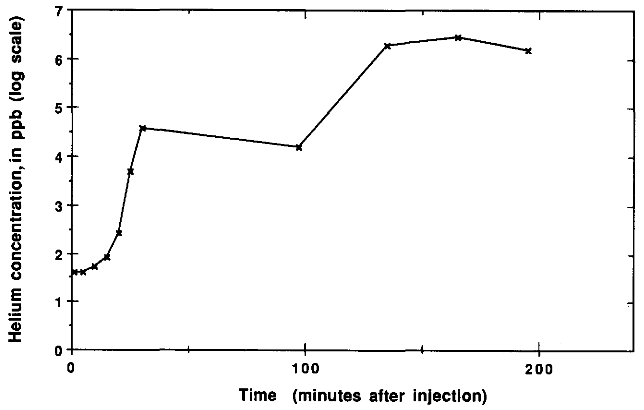

Figure 16. Variation in soil-gas helium concentration at station 6 as a function of time after injection at probe site 1.

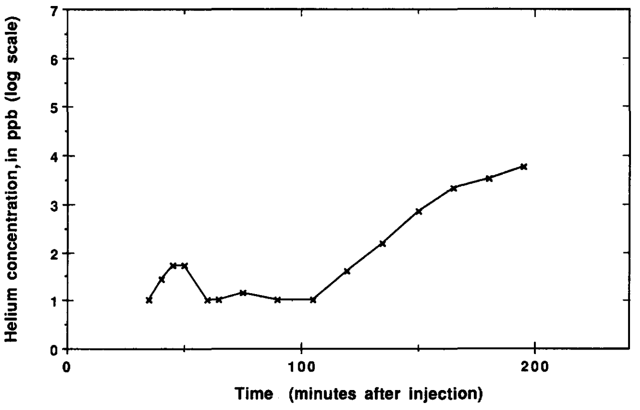

Figure 17. Variation in soil-gas helium concentration at station 7 as a function of time after injection at probe site 1.

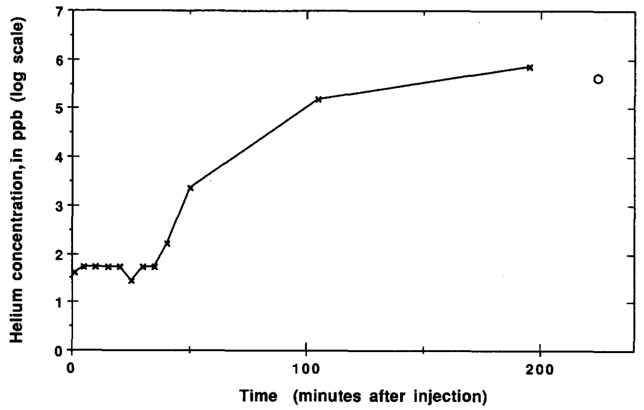

Figure 18. Variation in soil-gas helium concentration at station 8 as a function of time after injection at probe site 1. Open circle is the helium soil-gas concentration 18 hours after injection.

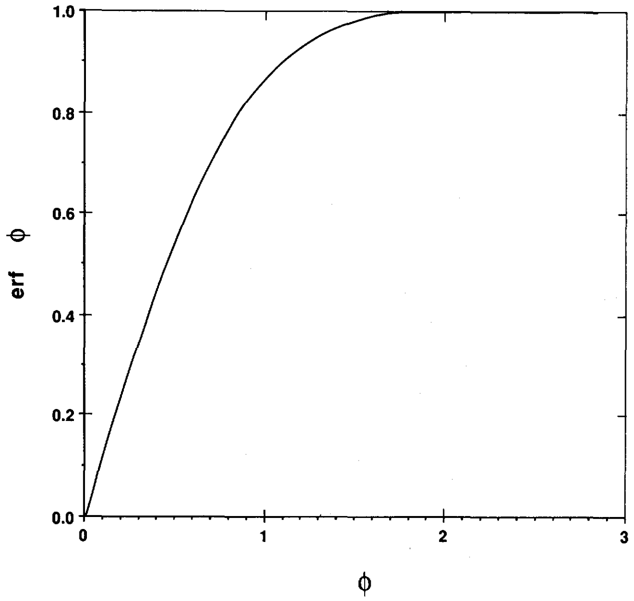

Figure 20. Plot of the probability function used for equation 1. erf is the gauss error integral, or probability function, which is obtained from a calculated value, represented by phi (ϕ), of parameters in equation 1.

then apply this finding to the more complex and varied soil system. The relationship among concentration, distance, and time can be expressed with the following formula:

$$\frac{C_z-C_s}{C_b-C_s}=\mathrm{erf}\left(\frac{z}{\sqrt{4Dt}}\right), \qquad (1)$$

where C_z = mole fraction of helium to be determined,
C_s = mole fraction of helium at the source,
C_b = background mole fraction of helium,
z = distance,

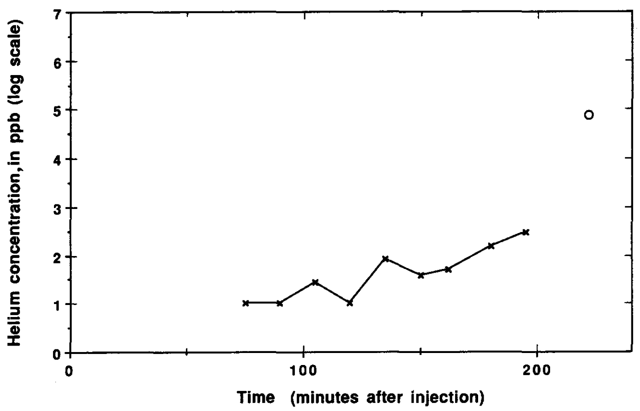

Figure 19. Variation in soil-gas helium concentration at station 9 as a function of time after injection at probe site 1. Open circle is the helium soil-gas concentration 18 hours after injection.

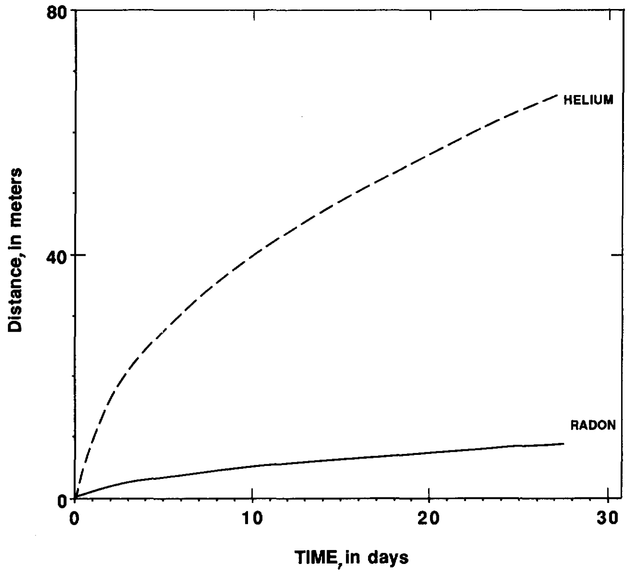

Figure 21. Plot of a probable migration distance with time for helium and radon in soils at the Boyertown site. The distances are the advancing front or penetration distances of gas from a source and are not expressing an equilibrium condition.

D = diffusivity of helium in air,
t = time, and
erf = the gauss error integral, or probability function.

Figure 20 is a plot of the error function. The equation is valid where the concentration gradients are within a few orders of magnitude of each other and the range of time and distance are within about 5 orders of magnitude.

The absolute helium concentration in the soil at the injection site after injection is not known, because the concentration exceeded the upper limit of analytical resolution. A controlled process to dilute the sample before analysis was not available in the field.

An alternate method is to select 2 monitoring probes as the points of evaluation for the equation. If the probe pair 6–7 is selected, the concentration of helium after 40 min is 35,240 ppb at station 6 and 5,270 ppb (which is considered background) at station 7. A diffusion coefficient of 0.6 cm^2/s for helium in air is used (Pogorski and Pogorski, 1982). Other values to determine C_z are C_s=35,240 ppb, C_b=5,270, z =200 cm, D =0.6 cm^2/s, and t =7,200 s. The calculated value of helium at station 7 is 6,220 ppb for 2 hours after the station 6 concentration was measured. After background concentrations (5,270 ppb) are subtracted, the measured concentration for station 7 is 7,340 ppb, only about twice what would be expected from diffusion alone. Several conditions exist that should be applied to the derivation to make the transition from an air system to a soil system. The first is that the source concentration at station 6 is continually increasing. This continual increase would have the effect of increasing the concentration at station 7 during the time period of 2 hours. A second condition is that the hindrance factor for gases diffusing through the soil must be considered. Typical hindrance factors are 2 to 7, if tortuosity, particle shape, and the soil void fraction are combined (Thibodeaux, 1979). The effective diffusion coefficient would be lower by a proportional amount. A third consideration is that convective mass transport is also acting in the near-surface environment. Because the measured concentration was greater than calculated, mass transport and increasing helium concentration at the source had a greater influence than the hindrance. Other station pairs show different responses. For example, the station pair 4–5 was known to be in a zone of higher moisture content. Station 4 showed an increase in helium 75 min after injection, but station 5 showed no response after 200 min. Diffusion coefficients for water (Boerboom and Kleyn, 1969) are on the order of 10^{-4} to 10^{-5} cm^2/s; thus, even if the zone is not saturated, the decrease in permeability caused by water in the pore spaces reduces the migration rate.

From these measurements for helium, a general migration rate can be derived. The detectable helium front migrates at a rate where time is related approximately to the square of the distance from the source. Figure 21 shows this function for helium as determined from the empirical data. At the end of a 28-day period, the helium would be capable of migrating 64 m. If it is assumed that the helium measurement is an indication of permeability and the approximation that diffusion of gases is proportional to the square root of the masses is used, radon would migrate 7 times more slowly and would traverse only about 9 m in the same time. That time period is a practical limit for radon because of the 3.8-day half-life. Interestingly, this distance is one that has been determined theoretically for radon in systems involving other types of overburden (Tanner, 1964). Naturally, when convective transport is involved because of atmospheric pumping or ground-water movement, radon and other gases can move more rapidly. Convective transport probably is always working in the near-surface environment. For example, the measured elevated radon content of soil-gas extends 20 m from the contact with the mylonite in this area (Reimer and Gundersen, 1989).

As a check on the practicality of the diffusion model, the diffusion coefficient for helium was calculated following the method of Bulashevich and Kartashov (1967), whereby the passage of the concentration maximum at a monitoring site is used. If the values for station 4 are used, where the maximum helium concentration was encountered at 200 min, a gross diffusion coefficient of 0.69 cm^2/s was determined. This compares favorably with the value of 0.6 cm^2/s used for the calculations in this study and indicates a close balance between hindrance factors and the opposing influences of mass transport.

CONCLUSIONS

Gaseous migration in the soil is a very difficult parameter to measure empirically or to determine theoretically. The soil is not a homogeneous medium; permeability, porosity, grain shape and size, composition, moisture content, and abundance of cracks and fractures are all variables. If the meteorologically induced effects that contribute to mass transport in the near-surface soil environment are included, the range of the variables becomes too large to define a universal, simplified model adequately. Even the rudimentary injection experiment performed in this study shows large variations over a distance of only 4 m. The results, however, set realistic constraints on the distance of gas migration in the Boyertown area and demonstrate that diffusion can contribute significantly to the migration. Rates of gaseous migration in the soil then can be explained by applying hindrance factors to a model characterizing diffusion in stagnant air. Rates of gaseous migration in the surficial environment are probably in the transition area between definitions of diffusion and convective mass transport, and the dominant mode will be controlled by the variable external influences.

Specific geologic units are responsible for the higher radon content in homes in the Boyertown area. Houses constructed on or near those units are more likely to have elevated indoor radon concentrations than those built 10 to 50 m away. The radon soil-gas measurements confirm the limited dispersion from the surface (or near-surface) contacts of the geologic units and suggest that the vertical and horizontal migration of radon is constrained. Where no unique geologic source of higher radon supply is found for dwellings having a radon buildup, there is the implication that overall gross permeability may be sufficiently large to allow a larger volume of rock or soil to contribute radon. It may be possible to establish criteria based on soil-gas radon measurements and permeability that would identify zones of high radon availability that contribute to indoor accumulation. Similarly, if the measurements were below a certain limit, lower concentrations would be anticipated. The limits would have a substantial and perhaps variable area of uncertainty between them, the size of which would depend on other external parameters such as construction techniques.

ACKNOWLEDGMENTS

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PART 1: STUDIES OF RADON IN ROCKS AND SOIL

SECTION 2: GEOLOGIC FIELD STUDIES

RADON IN SHEARED METAMORPHIC AND IGNEOUS ROCKS

Linda C.S. Gundersen

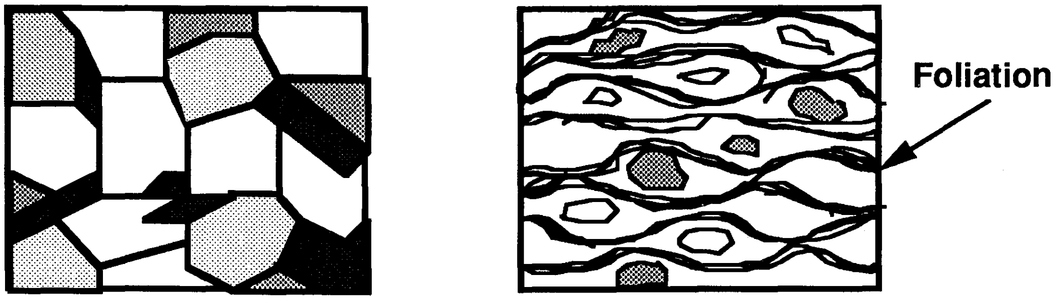
Abstract

Sheared fault zones in the Appalachian region of the Eastern United States have the potential for creating anomalously high amounts of indoor radon. These fault zones, many of which have known uranium occurrences, are usually characterized by high aerial radioactivity. Factors controlling the radon concentrations at these locations are bedrock uranium concentration, high permeability, and high radon emanation. These factors may be directly attributed to the deformation process of ductile shear, known as mylonitization of the rock. During mylonitization, the uranium concentration is increased by (1) the introduction of uraniferous fluids into the shear zone or (2) volume loss, which leaves the rock relatively enriched in uranium. Grain-size reduction of uraniferous accessory minerals common to metamorphic and igneous rocks such as titanite, zircon, monazite, and apatite makes uranium available for redistribution into the foliation. This process increases the emanation of radon from the rock dramatically. The texture imparted to the rock during shear also increases its permeability. Oxidation of iron during deformation and subsequent weathering results in the distinctive iron "staining" characteristic of many shear zones. Iron oxides and other metal oxides scavenge uranium and radium available through the weathering processes, increase the radon emanation from the rocks and soils, and make radon readily available to local ground waters. Shear zones in Pennsylvania, Virginia, New Jersey, and Maryland show anomalously high radioactivity and uranium, indoor radon, and soil radon concentrations that set them apart statistically from their unsheared host rocks.

INTRODUCTION

Many of the major shear zones in the Eastern United States have aerial radioactivity anomalies or have published uranium occurrences (Grauch and Zarinski, 1976; Carpenter, 1981; Baillieul and Daddazio, 1982; Baillieul and Dexter, 1982; Sargent and others, 1982; Madson and others, 1983). Shear zones have been identified as the cause of some of the highest indoor radon concentrations ever recorded in the United States. Two of the most notable of these occur at Boyertown, Pa.(Gundersen and others, 1987, 1988a; Smith and others, 1987), and Clinton, N.J. (Henry and others, this volume). This paper documents the Boyertown radon occurrence, as well as three other case studies, to illustrate the geologic processes responsible for making shear zones a source of high radon concentrations.

An introduction to the geologic processes that form mylonites and the resulting textural and mineralogical characteristics is essential to understanding the radionuclidic and geologic data presented in this paper. Mylonite is a rock type that develops as a result of ductile shear in which temperatures are generally above 250 °C. During ductile shear, the rock behaves plastically, rather than by fracturing, which is considered brittle shear. The development of a mylonite involves changes in the microstructure, porosity, permeability, and chemical composition of the parent rock. These changes can influence the mobility and concentration of uranium and may promote the emanation of radon (radon available to pore space). Grain-size reduction of minerals is characteristic of mylonites and is achieved by plastic processes (the crystals undergo ductile deformation, breaking down and becoming smaller in size). Granulation and fracturing play a minor role, especially with more resistant minerals such as uraniferous titanite and zircon. The grain-size reduction of these minerals may free uranium and make it available for chemical reaction and redistribution. Mylonitic rocks have a strong anisotropy, or foliation, due to the stretching and alignment of minerals during progressive deformation. This anisotropy may channelize fluids, which dissolve or replace minerals and thus change both the volume and composition of the deformed rock. Fluids also may interact with elements freed during grain-size reduction. As a result, the bulk composition of a mylonite often differs from that of its parent rock. Figure 22 shows a homogeneous granite before and after deformation, the foliation that develops, and the mineral changes that take place. The change in chemistry and mineralogy of a rock through mylonitization is documented by Beach and Fyfe (1976), Beach (1980), Sinha and others (1986), and Sinha and Glover (1986). In the mylonite zones described below, there is evidence of redistribution of uranium during deformation by oxidizing fluids into the developing foliation of the deforming rock. This redistribution and the development of foliation increase the radon emanation from the rock, its uranium concentration, and its permeability. Figure 23 is a general locality map for the mylonite zones described here.



Undeformed

Composed of quartz, sodium feldspar, potassium feldspar, biotite, titanite, and zircon.

Deformed

Composed of quartz, potassium feldspar, muscovite, epidote, hematite, and rutile. Biotite, titanite, and zircon breaking down and reacting. Sodium feldspar gone.

Figure 22. Physical and mineralogical difference between undeformed and deformed granodiorite.

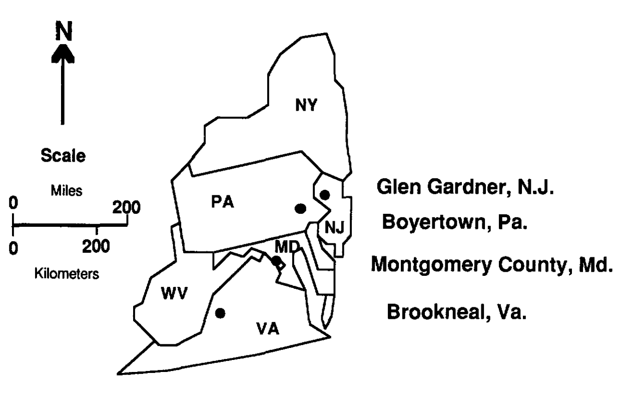


Figure 23. Location of shear zones in the Appalachian region.

METHODS

In each study area, techniques to assess the radionuclidic content of the soil and rocks, as well as their physical and chemical geologic characteristics, were used. Radon in soil gas was measured by use of a technique developed by Reimer (this volume). A carbon steel probe is inserted into the soil to a depth of 75 to 100 cm and 10 to 20 cm^3 of gas is withdrawn with a syringe. Samples are then measured in an alpha-sensitive scintillometer adapted with a Lucas cell and counted for 2 minutes. Three to five counts are made and then averaged. In the Eastern United States, this sampling depth corresponds approximately to the lowermost B and uppermost C soil horizons. All soils sampled in this study are residual, or saprolitic, soils derived from the underlying bedrock. Sampling was conducted in the summer during periods of dry, stable weather. In general, soils were dry to moderately moist. Radon concentrations are expressed in picocuries per liter (pCi/L).

Permeability measurements are from the Soil Conservation Service County Reports and indicate water permeability. Slow permeability is defined as less than 0.6 in/h (inches per hour), moderate permeability is 0.6 to 2 in/h, and moderately rapid permeability is 2 to 6 in/h.

Indoor radon measurements were supplied by State agencies and homeowners. Data from Boyertown, Pa., are 3-month alpha-track, winter measurements. The remaining data are 2- to 3-day charcoal cannister, winter measurements. All data are from homes that have basements and are proprietary information.

Uranium concentration was determined by fluorimetry. Thorium and radium concentrations were determined by gamma spectroscopy. Equivalent uranium was measured at the soil surface by use of a portable gamma spectrometer.

BOYERTOWN, PENNSYLVANIA

Radon concentrations in soil gas and their relation to geology were studied in Colebrookdale Township on the west side of Boyertown, Pa. The Boyertown occurrence was one of the first well-publicized discoveries of severe levels of indoor radon in the United States. The Proterozoic geology of Boyertown is described in detail by Gundersen and others (1987) and is only summarized here. Three major rock types underlie the area of interest shown on the map in figures 24 and 25. In the rock descriptions below, minerals are listed in order of decreasing abundance.

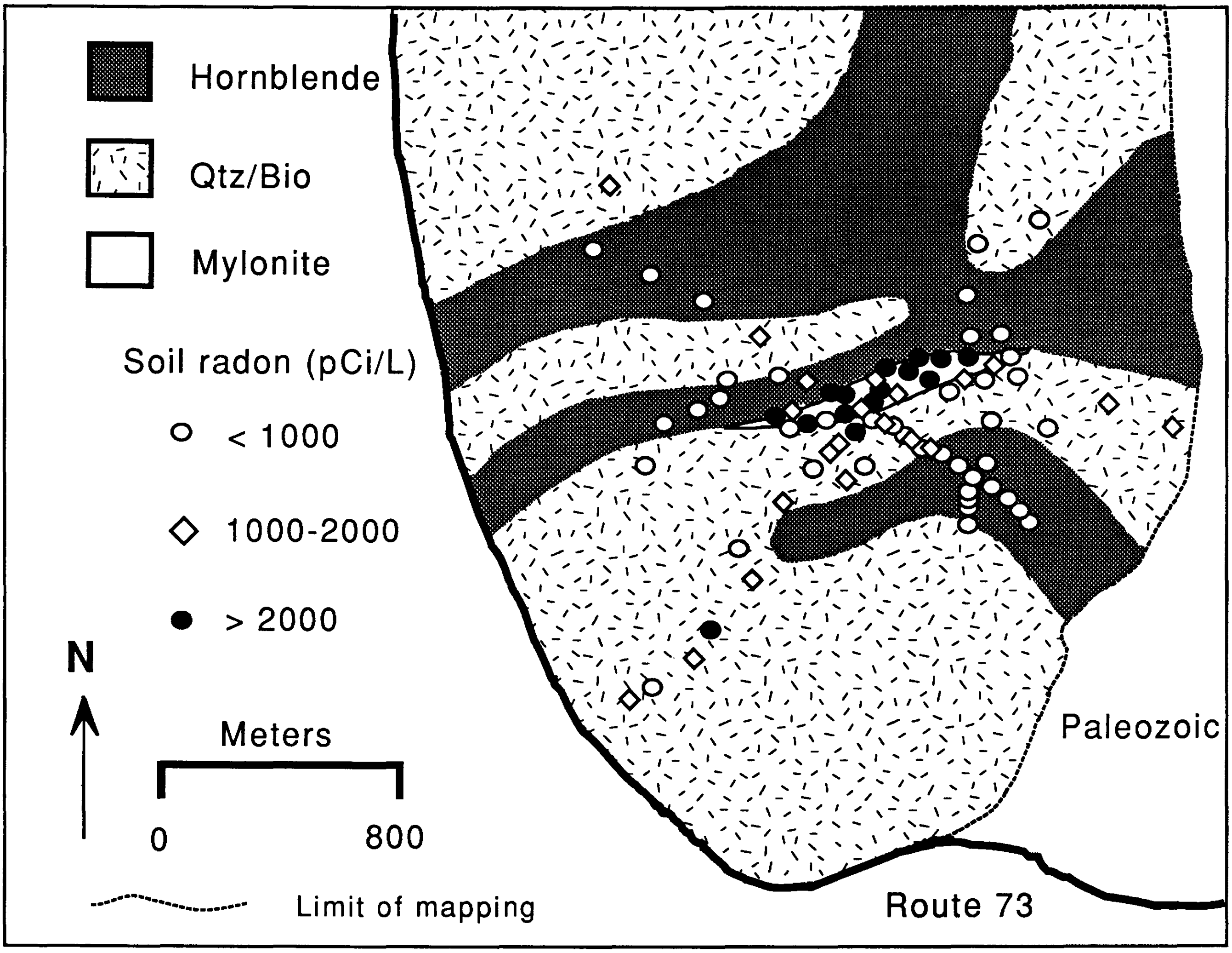


Figure 24. Radon measured in soil gas for the Boyertown, Pa., area. The shaded area is underlain by hornblende gneiss. The stippled area is underlain by interlayered quartz-feldspar and biotite gneiss. pCi/L, picocuries per liter.

Hornblende Gneiss

This rock is composed of quartz, plagioclase feldspar, and at least 20 percent hornblende. Pyroxene, biotite, and iron-sulfides occur in variable amounts. The maximum uranium concentration analyzed in this rock was 5 ppm (parts per million). Soil-gas and indoor radon values are low. Soils derived from this rock type are slowly permeable.

Interlayered Quartz-Feldspar and Biotite Gneiss (QFB)

Quartz-feldspar gneiss is composed of quartz, potassium feldspar, and sodium plagioclase. Biotite, titanite, magnetite, allanite, monazite, and rare garnet are accessories. Uranium concentrations as high as 25 ppm were measured. Biotite gneiss contains quartz, sodium plagioclase, and biotite and has potassium feldspar, titanite, magnetite, and monazite as common accessories. Uranium concentrations can be as high as 10 ppm. In both units, uranium-bearing titanite, allanite, monazite, and uraninite are sometimes concentrated in migmatite. These anatectic zones are marked by very large crystals of quartz and feldspar and locally produce high soil radon concentrations. In general, radon levels in soil and indoor radon levels are low to moderate for this rock unit. Soils derived from this rock type are moderately permeable.

Mylonite

Part of the QFB has been mylonitized near its contact with the hornblende gneiss. The mylonite differs mineralogically from the unsheared QFB by its abundant garnet. The mylonite's texture, defined by two foliation bands, is characterized by hematite and dark, fine-grained, highly uraniferous material. Late brittle shear or cataclasis is

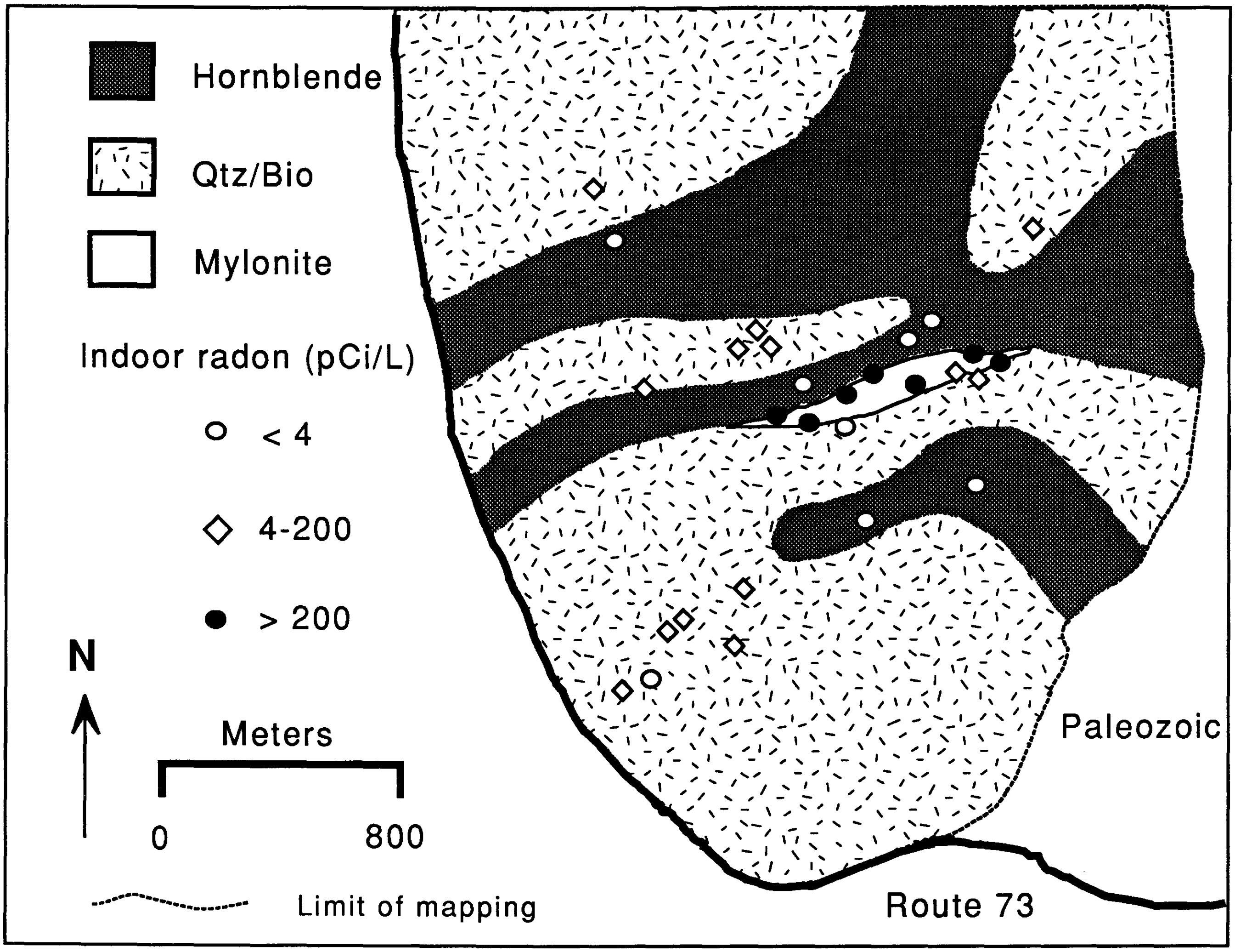


Figure 25. Indoor radon for the Boyertown, Pa., area. Geology is the same as in figure 24. pCi/L, picocuries per liter.

evident. Uranium concentrations as high as 50 ppm have been measured. Soils derived from this rock type are rapidly permeable.

The highest indoor radon and soil radon measurements are found over the mylonite developed in the QFB. Uranium in the QFB is located in titanite, monazite, zircon, and allanite. These minerals have low radon emanation; much of the radon produced does not escape the mineral or surrounding grains. When the QFB was mylonitized, the uraniferous minerals were broken down, and uranium was made available for reaction with hot, oxidizing fluids present in the shear zone. During deformation, the uranium was redistributed into the developing foliation along with hematite. Uranium is a much more effective producer of mobile radon when it is in the foliation. The mylonite weathers primarily along the foliation, exposing the uraniferous surfaces. Chemical analyses of the Boyertown rocks show uranium and radium to be in equilibrium (Agard and Gundersen, this volume), which is due in part to radium being scavenged by iron oxides as it is produced by uranium in the foliation. Radon emanation also is increased by this process. The Boyertown mylonite zone is structurally complex, and the source of the uranium enrichment in the zone is equivocal. The uranium/thorium ratio is highly variable in fresh rock samples of the Boyertown mylonite, and because veinlets of quartz are common in small, sheared fractures throughout the zone, it is suggested that fluid has been introduced into the zone, perhaps bearing uranium.

BROOKNEAL ZONE, VIRGINIA

The Brookneal mylonite zone near Brookneal, Va., bounds the western edge of the Danville basin in the southern Virginia Piedmont (Gates and others, 1986). The mylonite has developed in a homogeneous, Cambrian granodiorite composed of quartz, sodic plagioclase, minor hornblende, biotite, and titanite. Soil-radon concentrations show a positive correlation with the amount of shear measured in the underlying mylonitic outcrop (Gates and

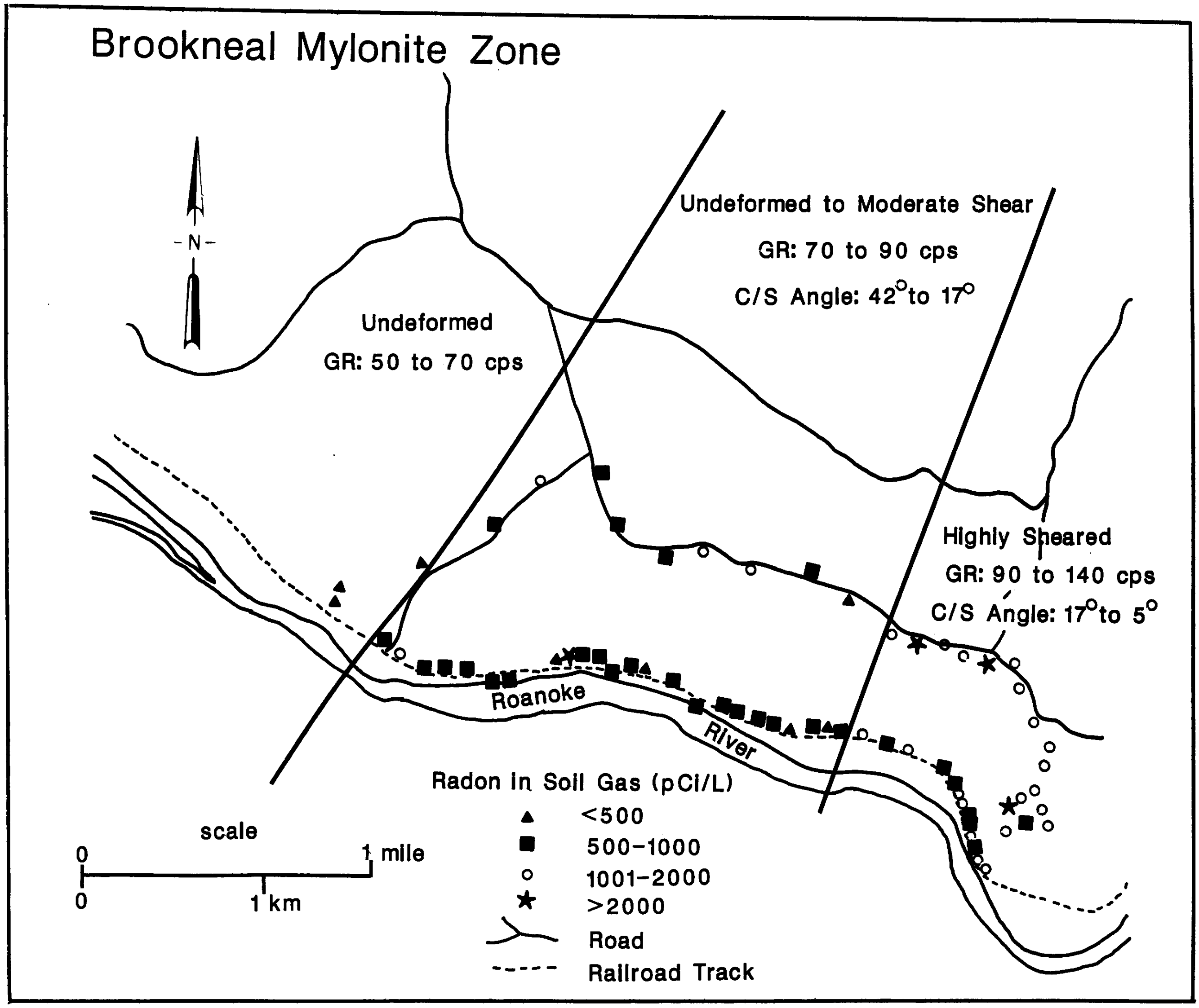


Figure 26. Summary of data for the Brookneal mylonite zone. Surface gamma radiation (GR) was measured in counts per second (cps) with a scintillometer, and the angle between the C and S bands was measured in degrees. Uranium and radon both increase as the C/S angle becomes smaller. pCi/L, picocuries per liter.

Gundersen, 1988). In the Brookneal zone, the amount of shear was measured by determining the angle between the two foliation bands developed during deformation. These are known as C and S foliation bands (Berthe and others, 1979; Simpson and Schmid, 1983; Simpson, 1984). The smaller the angle between the C and S bands, the greater the amount of shear strain. The C band forms initially at an angle of 45° to the S band. This angle becomes increasingly smaller with progressive shearing. Therefore, the smaller the C/S angle the greater the shear strain. Figure 26 is a summary map of the area showing the geology, the angle between C and S bands, and soil-radon concentrations. The angle between the C and S bands was measured on the outcrop exposed along the railroad track. Radon in soil was measured in the moderately permeable soil directly overlying the outcrop and along a road on top of the ridge that is above the outcrop. Uranium was measured in 10 of the rocks for which the C and S angles were measured. The correlation among uranium, soil radon, and the C/S angle is shown in figures 27 and 28. These results clearly show that soil-radon and uranium concentration in the rock increases with increasing shear. Uranium and radon both increase as the C/S angle becomes smaller. The progressive breakdown of the rock is directly linked to changes in chemical and physical composition with increasing shear.

The Brookneal zone is a good example of the relative enrichment of uranium by volume loss. Figure 27*A* shows that the uranium-thorium ratios change little with increasing shear. Thorium is considered an immobile element; thus, it appears that new uranium has not moved into the system to

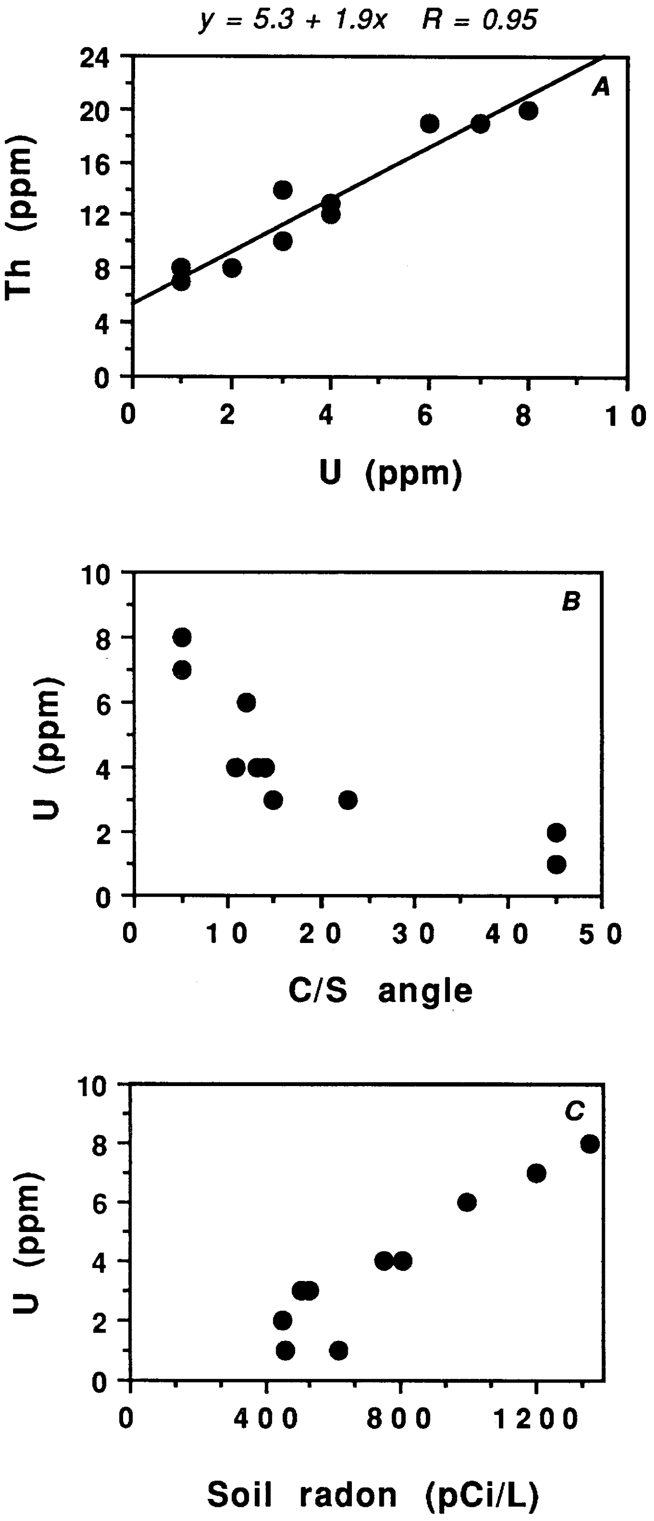


Figure 27. *A*, Uranium versus thorium for samples taken from outcrop along the railroad track. *B*, Uranium versus the angle between the C and S bands, as measured in the same samples. *C*, Uranium measured in outcrop samples versus radon measured in the overlying soils (along the railroad track). pCi/L, picocuries per liter; ppm, parts per million.

cause the increase in uranium concentration. Instead, other elements have moved out, especially silica and sodium, creating a relative enrichment of uranium in the mylonite as a result of volume loss.

GLEN GARDNER, NEW JERSEY

The Glen Gardner area was originally mapped by Markowitz (1975) and shows a number of mylonite zones occurring at the contacts between different rock types or crossing several different rock types. Indoor radon and soil-radon concentrations are high within the mylonite zone shown in figures 29 and 30. Four major lithologies are associated with the mylonite.

Hornblende Granite

Most of the Proterozoic gneiss in the area has been mapped as hornblende granite. This slightly variable rock type is commonly composed of quartz, potassium feldspar, sodium feldspar, and hornblende. Accessories include titanite, monazite, zircon, epidote, magnetite, allanite, and biotite. Permeability in the amphibolite is moderate. In the unsheared gneiss, soil radon concentration is moderate to low as is the indoor radon concentration. Uranium concentration in the rock varies between 2 and 8 ppm.

Amphibolite

A small body of hornblende amphibolite flanks the mylonite zone to the northeast. Hornblende, biotite, quartz, and sodic plagioclase are the principal minerals present. Abundant titanite, zircon, and magnetite occur as accessories. Permeability is slow in soil derived from the amphibolite. Soil radon is low to moderate, and indoor radon is low. Uranium concentration in the rocks varies between 1 to 3 ppm.

Magnetite Ore

Three abandoned magnetite pits are located along the northern edge of the mylonite zone. The magnetite ore is made up of magnetite and pyrrhotite interlayered with sillimanite and quartz gangue. Uranium concentration varies from 2 to 5 ppm in the magnetite ore, and radon concentrations over the ore were low to moderate. The immediate host of the orebody is a more amphibolitic facies of the hornblende granite and a biotite-sillimanite gneiss containing local layers of monazite. The biotite-sillimanite gneiss yields moderate to high radon concentrations and has uranium concentrations as high as 28 ppm where it has been

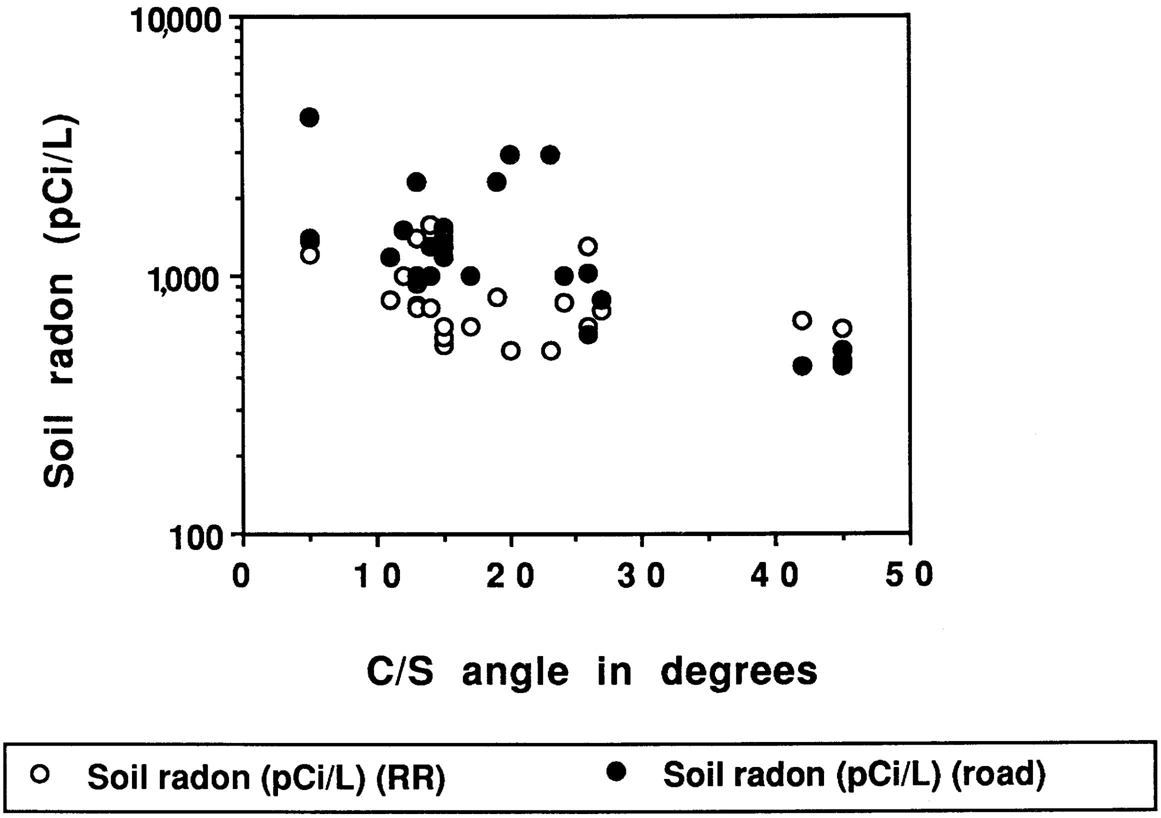


Figure 28. Radon in soil along the railroad track (RR) and radon in soil measured along the road on top of the ridge versus the angle between the C and S bands. pCi/L, picocuries per liter.

sheared. Soil derived from the magnetite varies in permeability from slow to moderate.

Mylonite

Deformed hornblende granite, minor hornblende gneiss, and minor magnetite ore are found in the main mylonite zone. Where mylonite is developed in the hornblende granite, iron oxides are common on the surface of the rock, and the mylonite is composed of quartz and sericitized feldspar. Foliations are composed of minor biotite, epidote, hematite, and muscovite. Soil derived from the mylonite is moderately permeable. Uranium appears to be amorphous in association with hematite in the foliation and varies in concentration from 5 to 20 ppm. Processes similar to those that occurred in the Boyertown mylonite are the reason that uranium and radon concentrations in the mylonite are high here.

MONTGOMERY COUNTY, MARYLAND

In a recent study of Montgomery County, the radon potential of Piedmont metamorphic rocks was found to be substantially lower than their sheared counterparts. Figure 31 shows a generalized version of the radon potential map developed for the county (Gundersen and others, 1988b). The map units are rated 1, 2, and 3 in order of increasing radon potential. A map unit rated 1 indicates that less than 40 percent of the homes in this rating area will have indoor radon screening levels greater than 4 pCi/L. All indoor radon screening levels will be in the range of 1 to 20 pCi/L. A map unit rated 2 indicates that about 50 percent of the homes will have indoor radon screening levels greater than 4 pCi/L, and as many as 10 percent of these will be over 20 pCi/L. A radon potential rating of 3 indicates that 60 percent or more of the homes in this area will be greater than 4 pCi/L and as many as 30 percent of the homes will be greater than 20 pCi/L. These ratings are based on 567 measurements of soil radon, 278 measurements of surface equivalent uranium, and 319 charcoal cannister measurements of indoor radon. These data were then compared with available geologic maps by Cloos (1964) and Froelich (1975) and permeability data from the Soil Conservation Service for Montgomery County.

The high-potential area in the western part of the county (3p) is underlain by phyllite that is characterized by high uranium concentrations not caused by shearing of the rock. Uranium in the phyllite is associated with graphite and is probably of depositional origin. However, the remaining

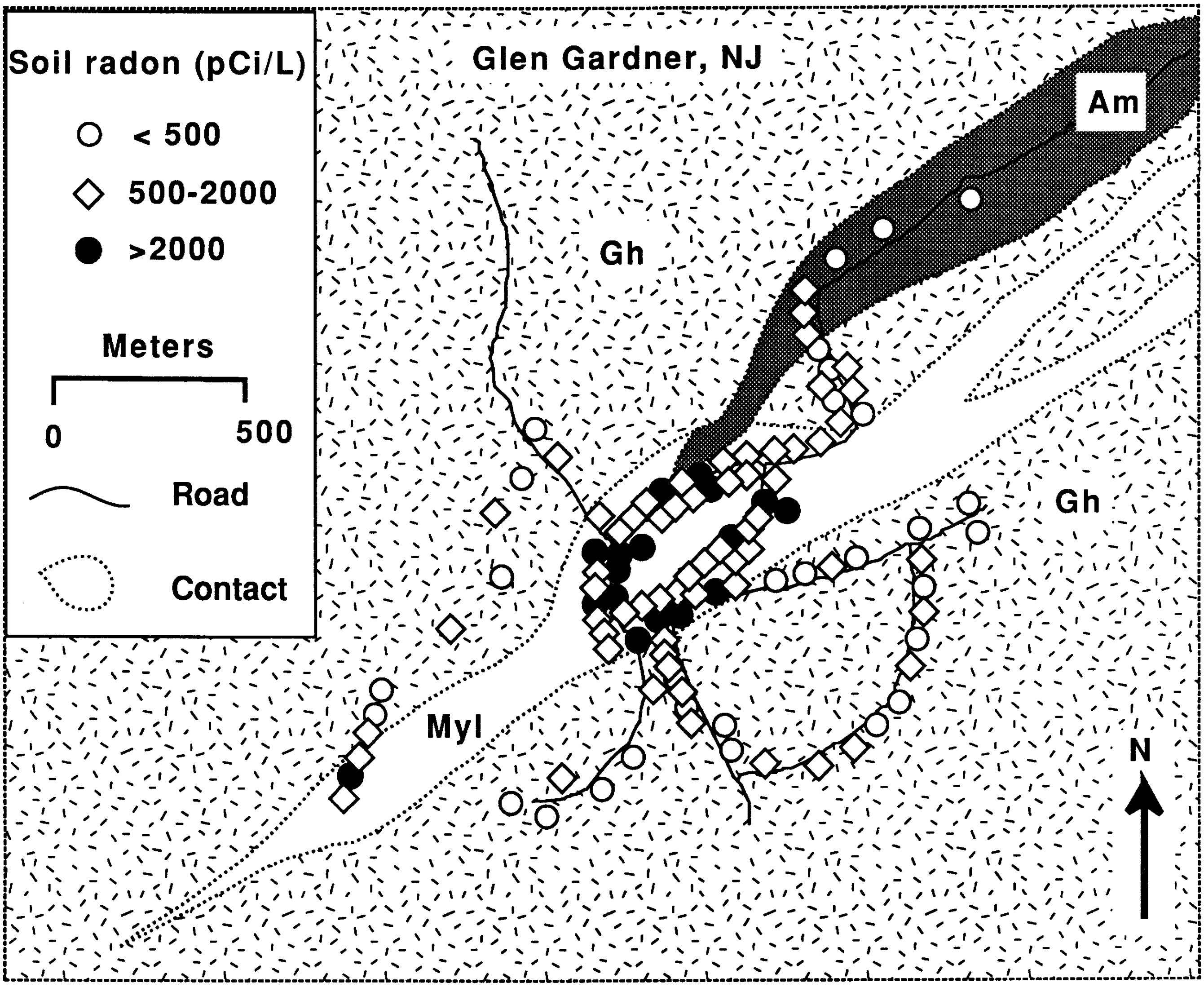


Figure 29. Radon in soil gas in the Glen Gardner area. Am, amphibolite; Gh, hornblende granite; and Myl, mylonite.

areas rated 3 have high indoor radon and soil radon levels caused by mylonitic shear zones. The shear zones in Montgomery County yielded the highest average soil radon (2,300 pCi/L) and the highest average indoor radon (24 pCi/L) when compared with the unsheared rocks. The three rock types in which shears are developed are described below.

Schist, Map Units 2s, 3s, and 3p/s

This rock unit underlies a significant portion of the central and eastern part of the county. It is composed of quartz, feldspar, and biotite with varying amounts of muscovite, chlorite, magnetite, staurolite, and garnet. Allanite, apatite, titanite, monazite, and zircon commonly occur as accessories and are the source of uranium in the rock. Interlayers of quartzite, metagraywacke, and phyllite are common. Equivalent uranium measurements average 2.7 ppm and have a maximum of 6.7 ppm where the schist is sheared. Permeability of soils developed from this rock type is moderate. Most of the schist is rated 2, but where it is sheared it is rated 3. In general, soil radon ranges between 500 and 1,500 pCi/L in unsheared schist. Where the schist is sheared, the range is between 2,000 and 3,000 pCi/L. Indoor radon ranges between 5 and 30 pCi/L in the sheared schist and between 1 and 10 pCi/L in the unsheared schist.

Gneiss, Map Units 2g and 3g

Numerous bodies of gneiss occur within the schist. They are composed of quartz, potassium feldspar, sodium feldspar, and variable amounts of hornblende and biotite. Granodiorite and pegmatite are common. Equivalent uranium measurements average 2.4 ppm and have a maximum of 3.7 ppm in sheared gneiss. Permeability of soils is

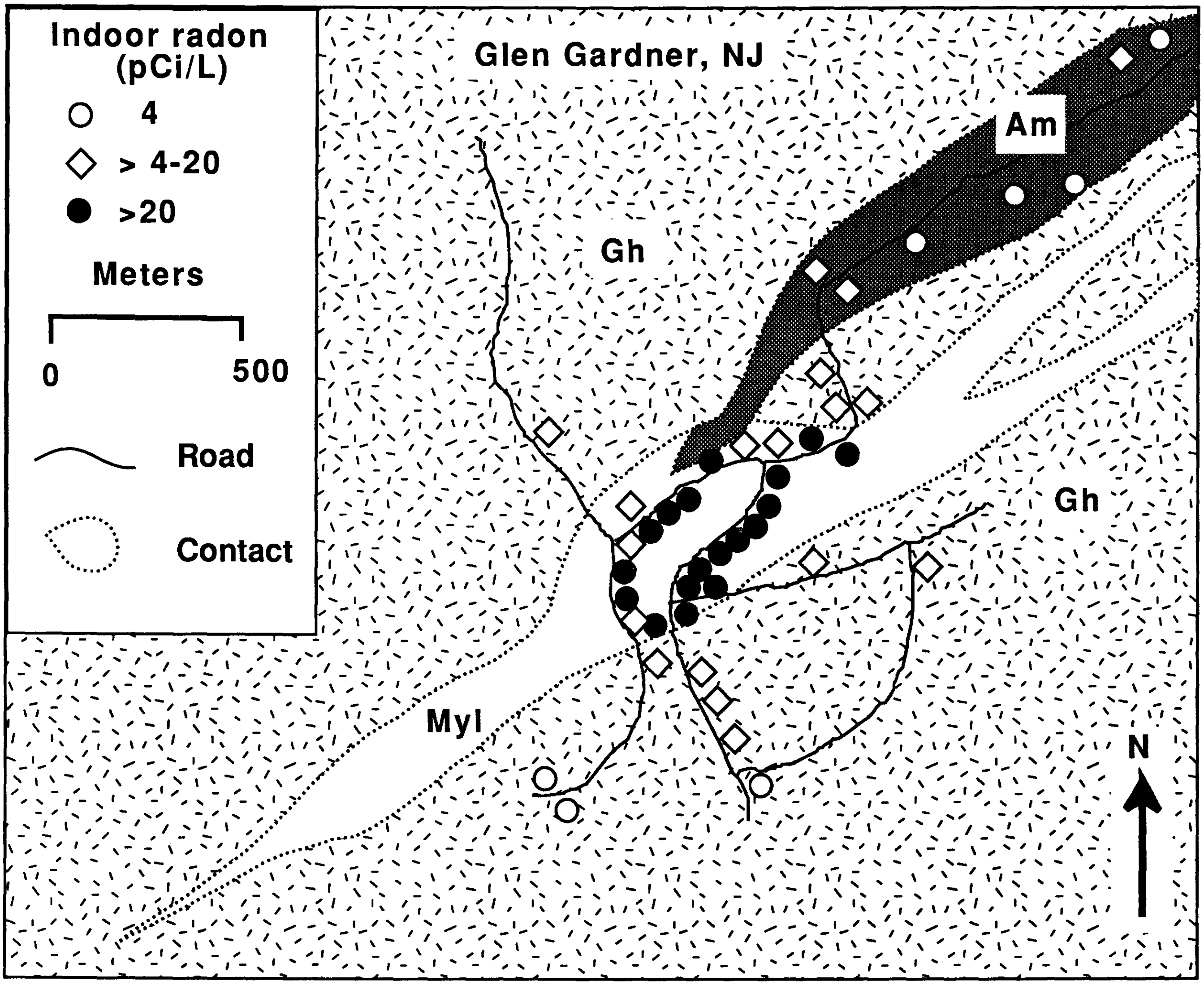


Figure 30. Indoor radon for the Glen Gardner area. Am, amphibolite; Gh, hornblende granite; and Myl, mylonite.

moderate. In the southern part of the county, the gneiss is characterized by a well-defined mylonitic texture. The northern outcrops of gneiss have a few scattered, narrow (1–10 m) mylonite zones. High indoor radon and soil-radon concentrations are found in all mylonite developed in gneiss. In general, soil-radon concentrations range between 2,000 and 3,000 pCi/L in the sheared gneiss and in the unsheared gneiss range between 250 to 1,500 pCi/L. Indoor radon concentrations range between 5 and 30 pCi/L in sheared gneiss and in the unsheared gneiss range between 1 and 10 pCi/L.

Mafic and Ultramafic Rocks, Map Units 1m, 2m, and 3m/s

Mafic and ultramafic bodies commonly occur within the schist and are typically bordered by shear zones. The rocks vary and include greenschist, quartz diorite, tonolite, amphibolite, metagabbro, serpentine, and chlorite-actinolite-talc schist. Permeability is slow in soils developed on this rock type. The igneous mafic rocks typically have a low uranium concentration, with equivalent uranium averaging 2.1 ppm and having a maximum of 4 ppm in the sheared mafic rocks. The source of uranium is probably from the adjacent sheared schist, and in a few rare cases, gneiss. In general, soil-radon concentrations in the unsheared mafic rocks range between 200 and 1,000 pCi/L and in the deformed mafic rocks range between 2,000 and 3,000 pCi/L. Indoor radon measurements of 4,000 pCi/L have been found in the map unit 3m/s, and, in general, indoor radon measurements range between 10 and 30 pCi/L in the sheared mafic rocks.

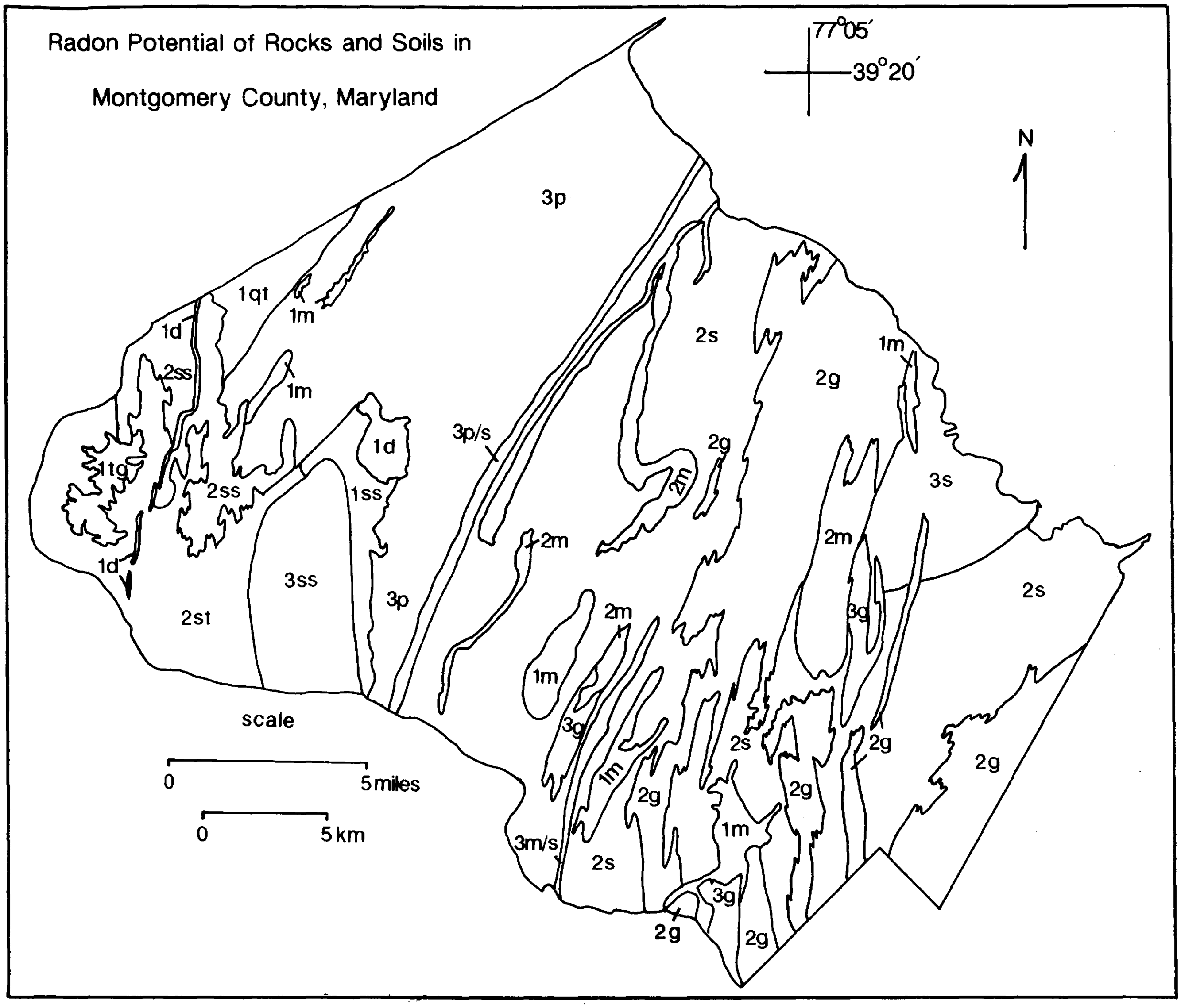


Figure 31. Radon potential for Montgomery County, Md. The numbers in each map unit label indicate low (1), moderate (2), and high (3) radon potential. The letter in each map unit label indicates the rock type: phyllite (p), schist (s), gneiss (g), sandstone (ss), diabase (d), siltstone (st), mafic (m), mylonite along the phyllite schist contact (p/s), mylonite along the mafic schist contact (m/s), quartzite (qt), or terrace gravel (tg). The following areas also have mylonite zones: 3g, 3s, and 2m. Modified from Gundersen and others, 1988b.

DISCUSSION AND CONCLUSIONS

The results of this study indicate that mylonites create anomalously high indoor radon and soil radon concentrations. From the rock descriptions and data presented, it is evident that each rock type has a distinctive radionuclidic signature. A plot of the average indoor radon versus the average soil-radon concentrations for the mylonite zones and associated rock types discussed in this paper is shown in figure 32. Data from studies in Easton, Pa., have also been included (Gundersen and others, 1988a). Note that the rocks cluster in three catagories. These catagories are controlled by the rock composition, the radionuclidic concentration, and the permeability. Rocks composed chiefly of dark-colored minerals such as hornblende, pyroxene, and biotite (commonly called mafic rocks) seldom have uranium concentrations of more than 5 ppm. Permeability is slow in mafic rocks because the dark minerals often weather to clay. The mafic rocks in all of the case studies generally have soil radon values less than 500 pCi/L and indoor radon values less than or equal to 4 pCi/L, except where they are sheared.

Uranium concentrations are higher in rocks of granitic composition, such as the QFB, schist, gneiss, and hornblende granite, because of the occurrence of minerals such as allanite, monazite, zircon, and titanite. These uraniferous minerals are common in metamorphosed sedi-

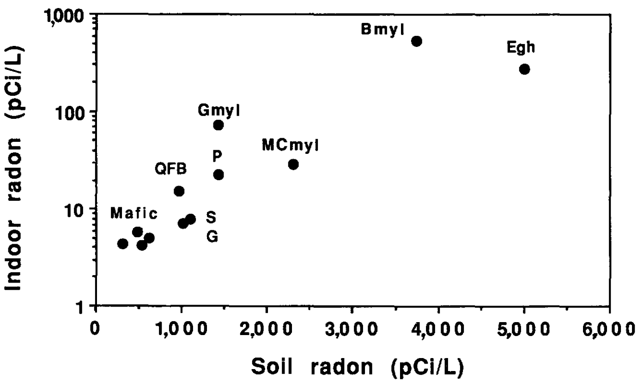


Figure 32. Average indoor radon versus average soil-radon concentrations for the areas described in this paper and for Easton, Pa. The correlation coefficient, or *R* factor, is 0.82. The four points labeled "Mafic" are two biotite schists from Easton and Boyertown, ultramafic rocks from Montgomery County, and an amphibolite from Glen Gardner. S, quartz-feldspar-biotite schist from Montgomery County; G, quartz-feldspar gneiss from Montgomery County; QFB, interlayered quartz feldspar and biotite gneiss from Boyertown; P, phyllite from Montgomery County; Gmyl, mylonite from Glen Gardner; MCmyl, mylonite from Montgomery County; Bmyl, mylonite from Boyertown; and Egh, sheared granite gneiss from Easton. pCi/L, picocuries per liter.

mentary rocks and granites. Soils derived from them are sandier and more permeable because of the abundance of quartz. Most of the rocks have moderate amounts of uranium (5 to 25 ppm) and moderate amounts of soil radon (500 to 2,000 pCi/L).

Mylonites formed in the various rock types are consistently moderate to high in radon and uranium concentrations. Several factors contribute to the high radon values in mylonites. First, uranium increases in concentration relative to the parent rock. Uranium is removed from its crystalline source in minerals such as titanite and reacts with oxidizing fluids present in the developing mylonite. As shear strain increases, grain-size reduction (the breaking down of the minerals) increases, and more uranium is mobilized and concentrated in the foliation. Concentrations increase by two processes: (1) new uranium is brought into the zone by fluids and (2) uranium is left in the zone as other elements leave the system and thus cause volume loss and relative enrichment of uranium.

Second, emanation of radon from uranium increases in the foliation. Uranium is removed from a site of low radon emanation (the crystal) to a site of high radon emanation (the foliation). Weathering of mylonites occurs mostly along foliations and exposes the source of uranium even further.

Third, the foliation developed during mylonitization increases the permeability of the rock dramatically. This pervasive foliation is also mimicked in the soil profile and provides paths for water and air movement.

In conclusion, shear zones can be local as well as regional areas of high radon concentrations, especially in mountain belts such as the Appalachians. Shear zones are not the only cause of high indoor radon levels, and some shear zones will not create radon problems. Shears developed in rocks having less than 1 ppm uranium may not produce sufficient radon to cause a problem unless uranium is introduced from another source during deformation. However, shear zones developed in rocks having higher uranium concentration, such as granitic rocks, have a high probability of causing an indoor radon problem.

High radon levels in water accompany high soil radon in Boyertown, Pa. (Wanty and Gundersen, 1988). Preliminary water data from two other shear zones, the Hylas fault in Virginia and a brittle shear fault in the Silver Plume Quartz Monzanite near Conifer, Colo., indicate that many domestic wells have radon concentrations greater than 10,000 pCi/L. High radon emanation, especially along fracture surfaces, contributes significantly to radon concentrations in water. Mylonites tend to be aquifers because of the increased permeability caused by the shear bands formed during deformation, whereas the same rock, when not sheared, may not be an aquifer.

ACKNOWLEDGMENTS

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THE GEOLOGY AND GEOCHEMISTRY OF SOILS IN BOYERTOWN AND EASTON, PENNSYLVANIA

Sherry S. Agard and Linda C.S. Gundersen

Abstract

Soils derived from a wide variety of metamorphic rocks in the Reading Prong, Pennsylvania, exhibit secular equilibrium between uranium and radium. Direct correlations between uranium, radium, radon in soil gas, and indoor radon indicate that radon in soil gas is attributable to the uranium content of the soils and can be used predictively to estimate levels of indoor radon. Permeability of the soils does not vary enough to affect the radon availability except where fractured and jointed bedrock is close to the surface, in which case convective flow, rather than diffusion, becomes the dominant radon transport mechanism. Radon emanation of soils, as measured in the laboratory, correlates well with radium, radon in soil gas, and indoor radon. Careful examination of the physical and chemical properties of the soil must be made in order for radon in soil gas to be used as an effective predictor of indoor radon levels. In addition, predictions of indoor radon levels based on radon in soil gas and geology are limited to ranges of values because of the inherent variability in soil makeup.

INTRODUCTION

The Reading Prong in the Northeastern United States is underlain by a complex sequence of Proterozoic metasedimentary and metaigneous rocks, many of which contain abundant uranium. As uranium-238 decays, it produces radium, which in turn decays to radon. Radon migrates into buildings under the influence of negative pressure differentials between the structure and the underlying bedrock or soil. When radon concentrates in indoor air, it can become a health problem (Environmental Protection Agency, 1986). Although the uranium and radium content of the bedrock is ultimately responsible for the presence of radon, it is often the porous and permeable nature of the overlying "soil" that allows the radon to migrate. As the uranium-bearing bedrock weathers both physically and chemically, it produces residuum and soils that differ, often significantly, from their parent material.

Two areas in the Reading Prong, Boyertown and Easton, Pa. (fig. 33), were selected for detailed study to examine the relation of geology, radon in soil gas, and indoor radon (see Gundersen, this volume, for a more detailed discussion of the geology). This paper discusses some preliminary data on the physical properties of surface soils as they may influence radon availability and migration. Radon in soil gas was measured by use of the methods of Reimer (this volume) at a depth of 75 cm, which corresponds to the lowermost B horizon or the upper C horizon in these soils. Indoor radon measurements were volunteered by the homeowners and are proprietary information. In general, the indoor radon measurements were made in basements during a 3-month period during the winter of 1984–85 by use of alpha-track devices. All the houses used in this study have basements.

GEOLOGY OF BOYERTOWN AND EASTON

Studies of radon in soil gas and geology were done in Colebrookdale Township on the west side of Boyertown, Pa., where one of the first cases of severe indoor radon in the United States was discovered (Smith and others, 1987). The following is a brief description of the three major rock types shown on the map in figure 34, which was derived from original mapping by Gundersen.

Hornblende gneiss.—This rock is composed of quartz-plagioclase feldspar and hornblende (>20%). Pyroxene, biotite, and iron sulfides occur in variable amounts. The rock is moderately foliated and jointed, but folds are not evident on the scale of outcrops. In general, both the soil-gas and indoor radon values are low for this unit (<1,000 pCi/ L (picocuries per liter) and <4 pCi/L, respectively).

Interlayered quartz-feldspar and biotite gneiss (QFB).—The quartz-feldspar gneiss comprises quartz, potassium feldspar, and sodium plagioclase. Biotite, titanite, magnetite, allanite, monazite, and rare garnet are accessories. The biotite gneiss contains quartz, sodium plagioclase, and biotite, with potassium feldspar, titanite, magnetite, and monazite as common accessories. This unit is weakly to moderately foliated and well jointed. Folds are not evident on the scale of outcrops. In both units, uranium-bearing titanite, allanite, monazite, and uraninite are sometimes concentrated in local anatectic zones. These zones of local melting are marked by very coarse crystals of quartz and feldspar and can produce local radon anomalies. In general, soil-gas radon and indoor radon for this unit is in the low to moderate range (200–2,000 pCi/L and 1–20 pCi/L, respectively).

Mylonite.—Near its contact with the hornblende gneiss, part of the QFB has been mylonitized. The mylonite is composed of deformed quartz-feldspar gneiss interlayered with minor biotite gneiss. It differs mineralogically from the undeformed QFB because it contains abundant garnet. The mylonitic texture, defined by C and S foliation bands, is characterized by dark, fine-grained, highly uraniferous material. The unit is highly foliated, and folds are

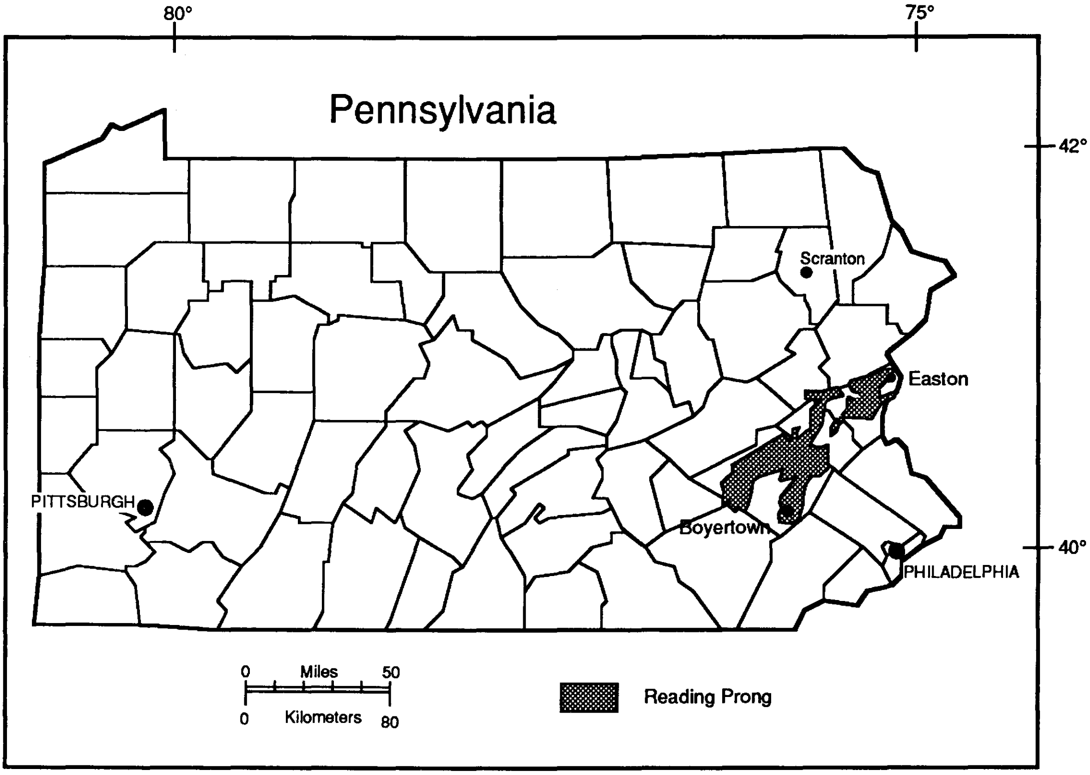


Figure 33. Locations of study areas in the Reading Prong.

abundant on the scale of outcrops. The strong anisotropy of the rock gives it a distinct weathering profile. In general, soil-gas radon and indoor radon measurements in this unit are high (1,000–12,000 pCi/L and >20 pCi/L, respectively).

Chestnut Hill in Easton, Pa., is underlain by Proterozoic metasediments that possibly represent an ancient reef assemblage. It is fault bounded on either side by the Cambrian Leithsville Formation. The sequence is tightly to isoclinally folded at map scale. The geology of this area was compiled from Drake (1969), and additional fieldwork was done by the authors. A geologic map of the area is shown in figure 35. The following is a description of the three main rock types.

Marble.—This unit is composed of dolomite, calcite, serpentine, tremolite, phlogopite, and talc. Pyroxene gneiss is interlayered with the marble along its northern contact. Uraninite and exotic uranium- and thorium-bearing minerals are concentrated where the marble is locally intruded by albitic pegmatite. In general, radon in soil gas is moderate to high (1,000 to 5,000 pCi/L).

Sillimanite gneiss.—This unit is composed of quartz, potassium feldspar, sodium plagioclase, and sillimanite. Uranium content is low in the sillimanite gneiss, except where it is intruded by tourmaline-bearing pegmatite near its contact with the quartz-feldspar gneiss. Soil-gas radon levels vary from low to moderate; some high values (>2,000 pCi/L) were recorded along contacts with other units.

Quartz-feldspar gneiss.—Potassium feldspar, quartz, biotite, and magnetite are the major constituents of this unit. Accessories include uranium-bearing titanite, monazite, and zircon. Uraninite occurs in quartz-filled shears, as much as 0.7 m long, in a discontinuous zone subparallel to strike. In the eastern part of Chestnut Hill, biotite gneiss forms a mappable layer within the quartz-feldspar gneiss. It is composed of quartz, potassium feldspar, sodium plagioclase, biotite, and rare magnetite layers. Local anatexis has produced coarse-grained quartz-feldspar knots with abundant uranium-bearing titanite and monazite similar to those observed in Boyertown. In general, the biotite gneiss has low radon levels in soil gas (<1,000 pCi/L). The quartz-feldspar gneiss generally has moderate to high radon levels in soil gas and indoors (1,000 to 5,000 pCi/L and 20 to 100 pCi/L, respectively.)

DEFINITION AND GENERAL PROPERTIES OF SOILS

The term "soil" has many definitions, depending on its usage. To the engineering geologist, soils include all unconsolidated surficial materials above the bedrock. To

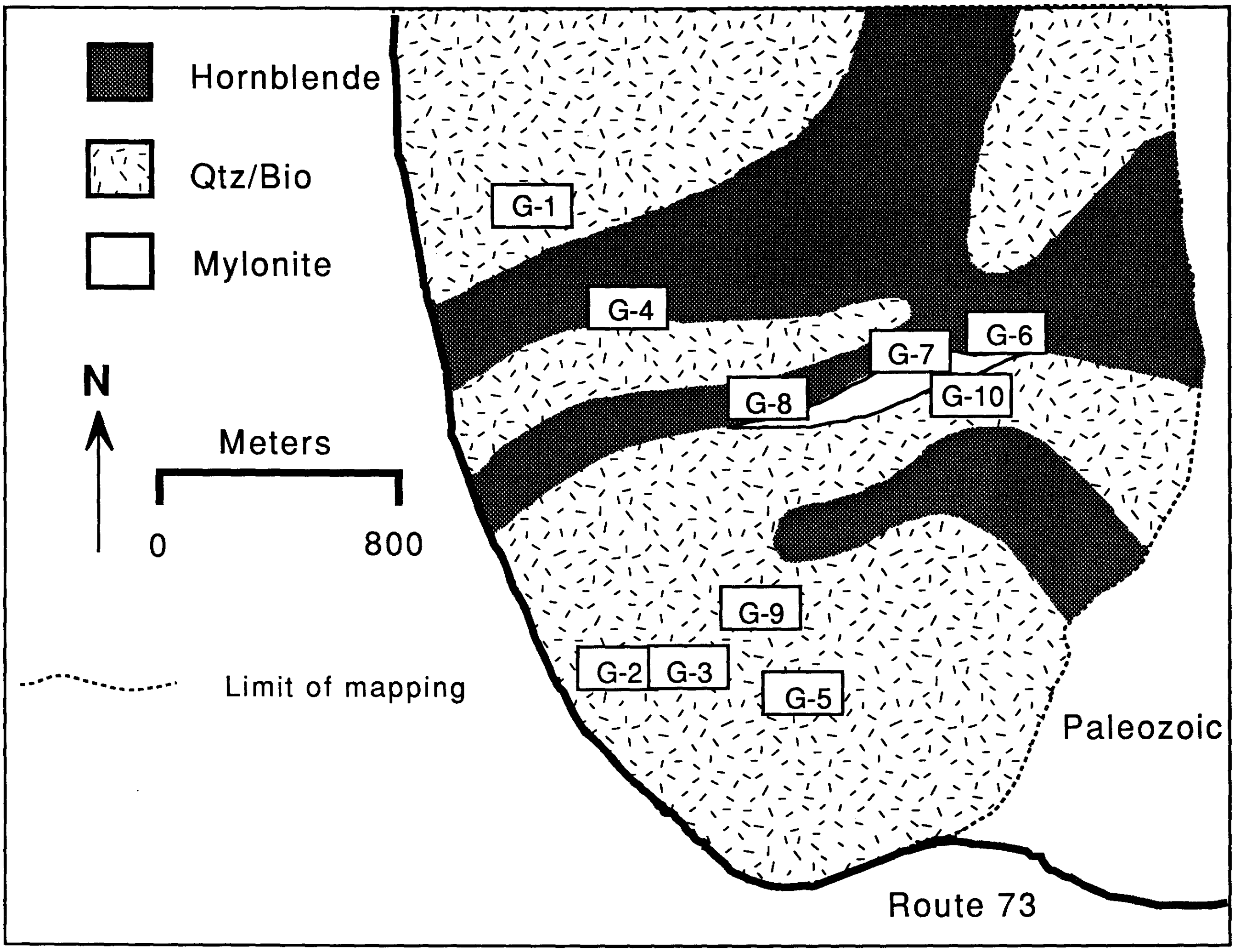


Figure 34. Geology of the Boyertown study area, located in the northeast portion of the Boyertown 7 1/2-minute quadrangle. Labels indicate approximate locations of soil profiles (some of which are listed in table 6). Qtz/Bio, interlayered quartz-feldspar and biotite gneiss.

the soil scientist, a soil is a collection of natural earthy materials containing living matter and capable of supporting plants. A definition by Birkeland (1984, p. 3) will serve to define soil as used in this study: "...a natural body consisting of layers or horizons of mineral and/or organic constituents of variable thickness, which differ from the parent material in their morphological, physical, chemical, and mineralogical properties and their biological characteristics...."

Soils are formed by an interaction of geomorphologic and pedologic processes that result in a distinct vertical arrangement of horizons, called the soil profile. A distinction can be made between a soil profile and a weathering profile in that the soil profile is generally limited to the upper, layered horizons within a much thicker weathering profile. The physical characteristics of the entire weathering profile, including the upper soil horizons, are significant in controlling the availability and migration of radon. This study focused on the upper horizons because (1) most houses are built on or in this zone, in some cases without much disturbance of the profile but in many cases with great disruption; (2) the layered nature of the soil profile produces a marked anisotropy in the physical properties; (3) in much of the previous work on soils and radon, a certain amount of homogeneity for "soil" has been assumed by treating all weathered and unconsolidated material above solid bedrock as a single unit; and (4) many of the in situ tests for radon in soil gas are conducted in these upper soil horizons, and some differences attributed to inherent differences in soils between areas may instead be due to differences between horizons.

The soil profile consists of several master horizons, the properties of which are related to the vertical movement and accumulation of certain materials, such as organic matter, clay, or iron. The A horizon forms at the surface as organic matter accumulates and clays and soluble salts are

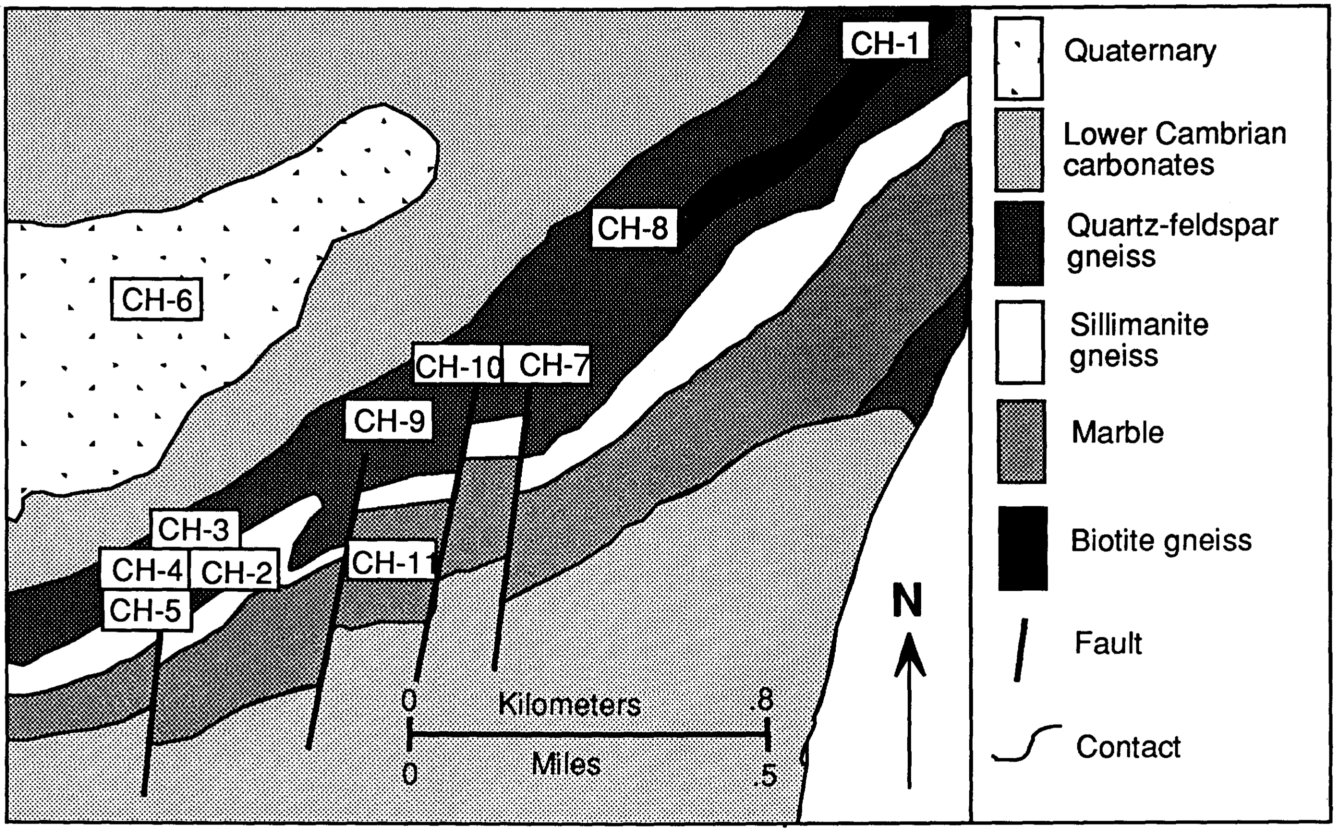


Figure 35. Geology of the Easton study area, located in the northeast portion of the Easton 7 1/2-minute quadrangle. Labels indicate approximate locations of soil profiles (some of which are listed in table 6).

transported to lower horizons. The underlying B horizon is characterized by clay accumulation, red color due to the oxidation of iron, accumulation of iron compounds, and continued leaching of soluble materials. The B horizon develops characteristic structures as individual soil particles become aggregated into peds. The C horizon consists of weathered parent material; clays and iron oxides accumulate here to a much lesser extent than in the B horizon, and structures are less distinct.

Nazaroff and others (1988) summarized some of the major physical properties of soil that control radon availability and migration, specifically grain size, permeability, porosity, and moisture content. The permeability of a soil is a measure of how easily air or water will flow through it and is often cited as one of the most significant factors controlling indoor radon accumulation. Permeability is not a fundamental soil property but is instead controlled by grain size, shape, and distribution; porosity (void ratio); mineralogy; degree of saturation; soil fabric; nature and temperature of fluid; and type of flow. The soil fabric is particularly important because it relates to the natural state of a soil in situ and includes porosity and permeability, which are due to soil structure and root pores that are generally destroyed during most sampling and testing procedures.

Figure 36 shows that the range of permeabilities of common soil types is more than 10 orders of magnitude (Nazaroff and others, 1988). The permeability within a given soil profile can vary by 2 orders of magnitude or more because the textural and structural properties of different horizons vary. Permeability can also vary horizontally and vertically within a particular horizon by up to 2 orders of magnitude. This natural variability increases the problems of understanding and predicting radon migration; construction activities that disturb the natural soils add to these problems. The clayey B horizon, especially when saturated, may act as a cap that inhibits the vertical migration of radon gas. However, once drained, even very clayey B horizons will have enough permeability, because of texture and soil structures, to allow radon migration. Nazaroff and others (1988) noted that the value of the air permeability remains roughly constant and equal to that for dry soil until moisture content exceeds field capacity. (Field capacity is the amount of water that is held on soil particle surfaces against gravitational pull.) For the textures of the soils derived from crystalline bedrock, the associated permeabilities are around 10^{-12} m^2, which is adequate to allow radon to migrate. Therefore, the soil texture and its associated permeability are only limiting factors when the moisture content is well above field capacity.

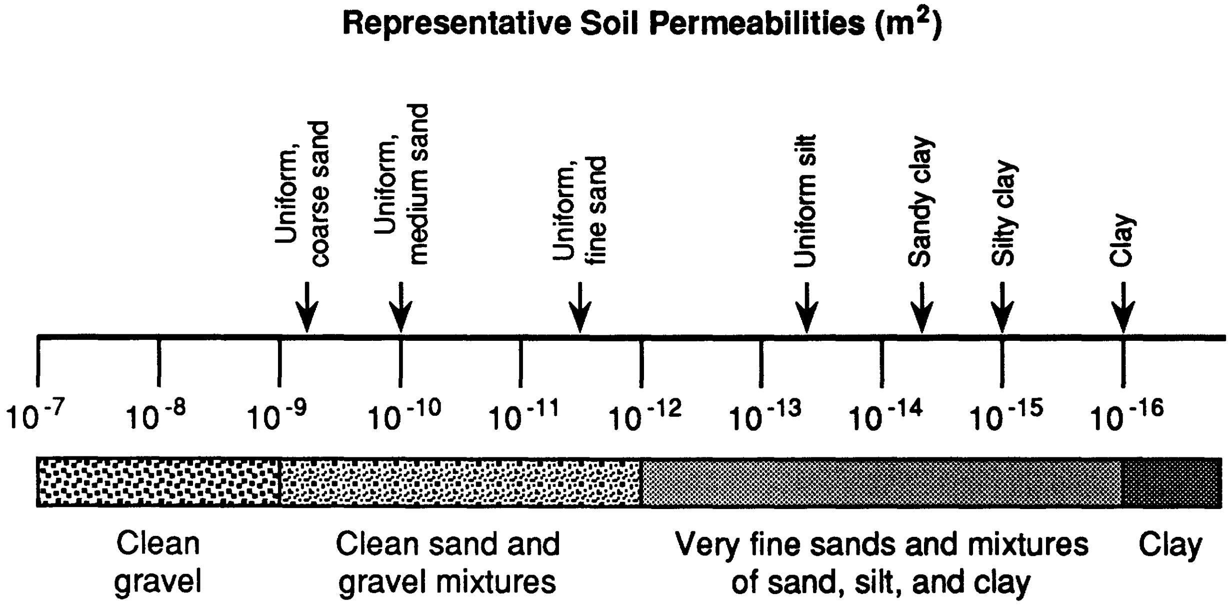


Figure 36. Typical soil permeabilities (modified from Nazaroff and others, 1988, p. 62). Permeability is expressed as specific (or intrinsic) permeability, usually in units of centimeters squared, which is based on the physical properties of the soil medium without regard to the characteristics of the fluid.

SOILS OF BOYERTOWN AND EASTON

Natural and manmade vertical exposures provide the best sites for describing and sampling soils, but in the developed areas of Boyertown and Easton, these were generally not available. In most areas, augering provided adequate general information, although horizon depths, boundaries, and some structural features could not be described accurately. The gravelly nature of some of the soils precluded augering more than 30 to 40 cm, so the nature of the lower B and C horizons could not always be determined. In a few key areas, small pits were excavated by hand to a depth of 75 to 150 cm. After each soil was described, 2 to 5 kg of soil material (the fraction less than about 1 cm) were collected from each horizon for laboratory analysis.

Soils in the Chestnut Hill-Easton area are part of the Conestoga silt loam and Conestoga-Hollinger extremely stony silt loam (Soil Conservation Service, 1974). The sampled soils developed on the quartz-feldspar gneiss, sillimanite gneiss, and pegmatites are generally thinner and sandier than the Soil Conservation Service (SCS) type profile. The generalized composite description that follows is shown schematically in figure 37.

Easton quartz-feldspar gneiss and sillimanite gneiss:

Ap—0 to 15 cm; silt loam to sandy loam; moist color 10YR 3/2 (very dark gray brown); single grain to weak, fine, subangular blocky structure; loose, nonsticky, nonplastic consistence.

BA—15 to 20 cm; sandy loam; moist color 10YR 3/4 (dark brown); weak, fine, subangular blocky structure; very friable, nonsticky, nonplastic consistence.

Bt—20 to 50 cm; sandy clay loam to loamy sand; moist color 7.5YR 5/6 (strong brown); weak to moderate, fine to medium, subangular blocky structure; friable, slightly sticky, slightly plastic consistence; abundant thin clay films.

BC—50 to 70 cm; loamy sand to sand; moist color 10YR 4/4 (dark yellowish brown); weak, fine, subangular blocky structure; very friable, nonsticky, nonplastic consistence.

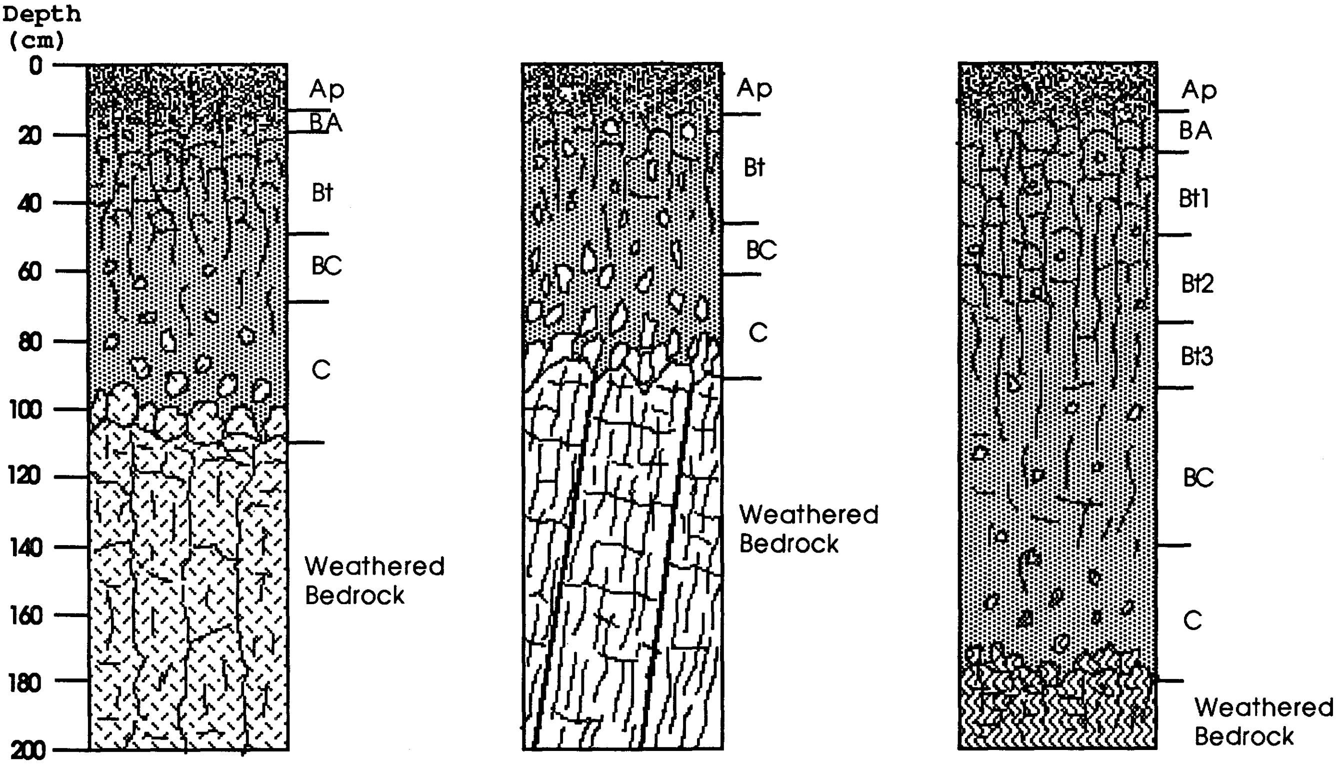
C—70 to 110+cm; loamy sand to sand; moist color 10YR 4/6 (dark yellowish brown); single grain structure; loose, nonsticky, nonplastic consistence.

Soils in the Gablesville-Boyertown area are part of the Chester channery silt loam (SCS, 1970). The soils developed on the biotite gneiss and hornblende gneiss are sandier but similar to the SCS type profile. The soils developed on the quartz-feldspar gneiss, particularly in the mylonitic zone, are much thinner and coarser (50 to 80% coarse fragments). The composite descriptions that follow are shown schematically in figure 37.

Boyertown quartz-feldspar gneiss:

Ap—0 to 15 cm; very gravelly silt loam to sandy loam; moist color 10YR 3/3 (dark brown); single grain to weak, fine, subangular blocky structure; very friable, nonsticky, nonplastic consistence.

Bt—15 to 45 cm; very gravelly sandy loam to loamy sand; moist color 7.5YR 6/6 (reddish yellow) to 10YR 5/6



Easton quartz-feldspar gneiss and sillimanite gneiss

Boyertown quartz-feldspar gneiss

Boyertown biotite gneiss and horneblende gneiss

Figure 37. Representative soil profiles from Boyertown and Easton, Pa. See text for detailed discussion.

(yellowish brown); weak to moderate, fine to medium, subangular blocky structure; friable, slightly sticky, slightly plastic consistence; abundant thin clay films.

BC—45 to 60 cm; very gravelly sandy loam to loamy sand; moist color 7.5YR 6/7 (reddish yellow) to 10YR 6/6 (brownish yellow); weak, fine, subangular blocky structure; very friable, slightly sticky to nonsticky, slightly plastic to nonplastic consistence.

C—60 to 95+ cm; very gravelly loamy sand to sandy loam; moist color 7.5YR 5/6 (strong brown); single grain to weak, fine, subangular blocky structure; loose, nonsticky, nonplastic consistence.

Boyertown biotite gneiss and hornblende gneiss:

Ap—0 to 15 cm; channery silt loam to sandy loam; moist color 10YR 3/3 (dark brown) to 7.5YR 3/4 (dark brown); single grain to weak, fine, subangular blocky structure; loose to very friable, slightly sticky to nonsticky, slightly plastic to nonplastic consistence.

BA—15 to 25 cm; channery to gravelly, loam to sandy clay loam; moist color 10YR 4/3 (dark brown); weak to moderate, very fine to fine, subangular blocky structure; very friable, sticky, plastic consistence; many thin clay films.

Bt1—25 to 50 cm; channery to gravelly, sandy loam to sandy clay loam; moist color 7.5YR 4/6 (strong brown) to 5YR 3/4 (dark reddish brown); moderate to strong, fine to medium, subangular blocky structure; very friable, sticky, plastic consistence; abundant thin to thick clay films.

Bt2—50 to 75 cm; channery to gravelly, sandy loam to sandy clay loam; moist color 7.5YR 5/6 (strong brown) to 5YR 4/6 (yellowish red); moderate to strong, fine to medium, subangular blocky structure; friable, sticky, plastic consistence; abundant thick clay films.

Bt3—75 to 95 cm; channery to gravelly sandy loam; moist color 7.5YR 6/6 (reddish yellow) to 7.5YR 5/8 (strong brown); weak to moderate, fine to medium, subangular blocky structure; friable, slightly sticky to sticky, slightly plastic to plastic consistence; abundant thin to thick clay films.

Table 5. Textural analysis of selected soils

[In weight percent; clay fraction determined by X-ray particle-size analysis; sand and silt fractions determined by both wet and dry sieve analysis]

Sample number/location	Clay	Silt	Sand	Soil texture
Boyertown biotite gneiss				
G–6B(B1)	4.0	28.5	67.5	sandy loam.
G–6B(B21t)	12.2	37.0	50.8	loam.
G–6B(B22t)	13.3	33.8	52.9	sandy loam.
G–6B(B23t)	12.8	31.4	55.9	sandy loam.
G–6B(B3)	9.5	27.7	62.7	sandy loam.
Boyertown quartz-feldspar gneiss				
G–8D(Ap)	5.3	35.5	59.1	sandy loam.
G–8D(Bt)	4.2	42.1	53.8	sandy loam.
G–8D(75)	12.7	29.2	58.1	sandy loam.
G–8DC	9.0	27.3	63.7	sandy loam.
Easton quartz-feldspar gneiss				
CH–9B(B1)	8.8	21.8	69.4	sandy loam.
CH–9B(B21t)	8.0	27.0	65.0	sandy loam.
CH–9B(B22t)	14.0	24.5	61.5	sandy loam.
CH–9B(B3)	6.1	10.8	83.1	loamy sand.
CH–9B(C1)	4.3	10.6	85.1	loamy sand.
CH–9B(C2)	14.1	28.1	57.8	sandy loam.
CH–9B(C3)	5.2	19.3	75.6	loamy sand.

BC—95 to 140 cm; channery to gravelly sandy loam; moist color 7.5YR 6/6 (reddish yellow); weak to moderate, fine to medium, subangular blocky structure; very friable, slightly sticky to nonsticky, slightly plastic to nonplastic; many thin clay films.

C—140 to 180+ cm; channery to gravelly, loamy sand to sandy loam; moist color 7.5YR 5/7 (strong brown); weak, fine, subangular blocky to weak platy; very friable, nonsticky, nonplastic consistence.

Particle-size analyses of the less-than-2-mm fraction have been completed on three representative soils, one from each major rock type. The sand and silt fractions were determined by both wet and dry sieve analysis; the clay fraction was determined by use of an X-ray particle-size analyzer (table 5). The analyses allow the soil texture to be classified, as shown on figure 38. All the soils tested from Boyertown and Easton fall into the range of loam to loamy sand. Clay contents range from 4 to 14 weight percent, with the highest amounts in the B horizons.

The Boyertown biotite and hornblende gneisses produce the finest textured soils of those sampled. They weather to fine sand, silt, and clay; small, angular rock fragments occur mainly at depth. These soils are also the thickest and have more recognizable horizons, but these observations may be biased by the better exposures available for the biotite gneiss (natural cuts and pits instead of auger holes). The Boyertown quartz-feldspar gneiss weathers to coarse sand and larger, angular, platy rock fragments that are present throughout the profile (although more concentrated at depth). The Easton quartz-feldspar gneiss and pegmatite weather by granular disintegration to coarse sand and small, equidimensional rock fragments (present mainly in the lower horizons).

The Easton soils are generally more permeable than the Boyertown soils because the Easton soils are sandier. Soils on the Boyertown mylonitic gneiss are also relatively permeable because the bedrock is highly fractured and the soil is coarse overall. The finer textured soils on the biotite gneiss are slightly less permeable. All these soils, however, are well-graded (poorly sorted), gravelly, sandy loams or loamy sands, the permeabilities of which probably vary no more than 1 order of magnitude (fig. 36).

RADON EMANATION OF SOILS

As a check on in situ soil-gas measurements, an experiment to measure the radon emanation of soil samples was conducted. The samples collected in the field represent a bulk sample of the finer (generally the less-than-1-cm) fraction of the soil (although larger fragments were excluded from the sample, these would contribute proportionally less radon than the finer fraction because these larger fragments have smaller surface area to volume ratios). For the emanation experiment, a sample of about 100 g (grams) of soil was split from the bulk sample by use of a coarse sample splitter; a second sample of about 250 g was sent to an outside laboratory for analysis of uranium and radium content. Because the initial sampling and subsequent splitting disaggregated the soil peds, the experiment does not represent field conditions. It does, however, give an indication of radon availability as related to soil texture (particle sizes).

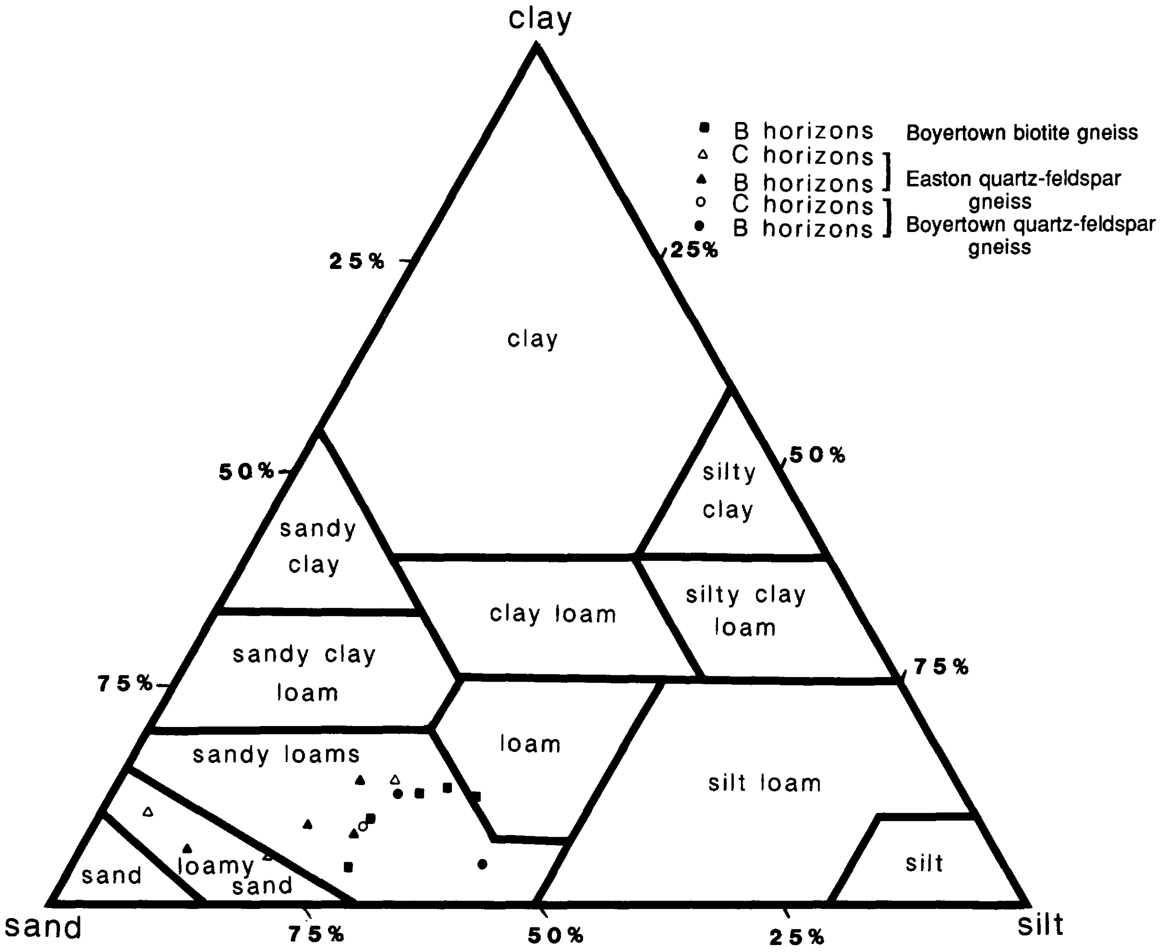


Figure 38. The distribution of soil textures for B and C horizons from selected soils (SCS classification system).

The soil samples were weighed, air dried in the lab for 24 to 36 hours, and weighed again. The dried soil samples were placed in 250-mL (milliliter) Erlenmeyer flasks and sealed with rubber stoppers fitted with septa. The volume of each stoppered flask was estimated by comparing the level of the stopper in the flask with an identical flask whose volume had been measured and marked. The volume of the soil was calculated from the weight of the dry soil by assuming a specific gravity of 2.65 for the mineral solids. It was assumed in this calculation that all the voids in the soil were filled with air and that all the voids were connected to the air in the flask (and could therefore contribute radon to the air in the flask). The samples were allowed to sit for at least 28 days so that the radon and radon daughters in each flask could come to equilibrium with the radium in the soil.

The flask was then injected with 50 cm^3 (cubic centimeters of room air, which was mixed by pumping the syringe 5 to 10 times. A 50-cm^3 sample was extracted from the flask and immediately injected into the detector. The sample was measured for 5 minutes and recorded as the raw count, from which the background count was subtracted. Total and specific activities calculated from these values are shown in table 6, along with laboratory data for uranium and radium contents.

Possible sources of error in the experiment include (1) leakage from the flask through the septa or around the rubber stoppers and (2) inaccurate estimates of flask volume, air volume, and soil volume. When measurements were repeated after another 30 days, 75 percent of the samples produced values within 20 percent of the first measurement.

DISCUSSION

Gundersen and others (1988) noted a strong correlation between soil-gas radon, indoor radon, and the concentration of uranium in the underlying bedrock. In the Boyertown area, radon activities were highest in the mylonite zone and lowest over the hornblende gneiss. In the Easton area, even higher radon activities were found in the

Table 6. Lab data for uranium (U), radium (Ra), and radon (Rn) for Pennsylvania soils

[G prefix, Gablesville-Boyertown soil; Ch prefix, Chestnut Hill-Easton soil; cm, centimeters; ppm, parts per million; pCi/g, picocuries per gram; pCi/L, picocuries per liter; %, percent; chem U, chemically determined uranium; eU, equivalent uranium; eTh, equivalent thorium; K%, weight percent potassium; —, no data]

Soil sample	Depth (cm)	Chem U (ppm)	U (pCi/g)	eU (ppm)	eTh (ppm)	K %	Ra-226 (pCi/g)	Ra-228 (pCi/g)	Total activity (pCi/L)	Specific activity (pCi/g)	In situ soil Rn (pCi/L)
CH-2(8)	8	210	70.1	120	210	2.3	40.0	23.0	23,378	205	—
CH-2(60)	60	230	76.8	130	340	5.7	43.0	37.0	31,823	287	—
CH-3B(40)	40	35	11.7	30	44	4.7	10.0	4.8	3,228	35	—
CH-3B(75)	75	26	8.7	27	22	4.0	9.1	2.4	2,838	31	8,000
CH-3B(180)	180	20	6.7	22	45	4.0	7.4	5.0	2,569	29	—
CH-7(20)	20	31	10.4	28	28	3.1	9.3	3.0	5,471	49	—
CH-7(35)	35	32	10.7	30	39	3.5	9.9	4.3	5,446	48	—
CH-7(50)	50	30	10.0	27	19	2.2	8.9	2.1	4,825	47	—
CH-7(65)	65	25	8.4	24	12	1.3	8.0	1.4	5,336	49	—
CH-7(85)	85	27	9.0	25	15	1.3	8.4	1.7	9,165	87	—
CH-9A(Ap)	10	44	14.7	47	75	4.2	16.0	8.3	2,355	25	—
CH-9A(B21t)	30	39	13.0	35	60	4.1	12.0	6.6	5,193	54	—
CH-9A(B22t)	50	40	13.4	45	64	3.6	15.0	7.0	4,162	44	22,750
CH-9B(B1)	8	22	7.3	21	23	5.0	6.9	2.5	5,231	48	—
CH-9B(B21t)	25	29	9.7	31	20	4.7	10.0	2.2	3,639	35	—
CH-9B(B22t)	40	19	6.3	22	17	4.6	7.3	1.9	4,890	46	—
CH-9B(B3)	55	19	6.3	17	36	4.5	5.5	4.0	3,629	33	—
CH-9B(C1)	68	16	5.3	16	22	5.3	5.4	2.4	2,539	23	—
CH-9B(C2)	78	25	8.4	34	34	3.5	11.0	3.7	5,813	58	—
CH-9B(C3)	88	57	19.0	53	66	4.2	18.0	7.2	8,194	78	—
CH-10A(B3)	33	48	16.0	52	41	5.7	17.0	4.5	6,702	68	—
CH-10A(C)	45	46	15.4	54	19	5.9	18.0	2.1	5,730	57	11,000
CH-10C(20)	20	60	20.0	61	52	4.5	20.0	5.7	9,768	103	12,000
CH-11	5	18	6.0	29	31	1.4	9.8	3.4	2,009	22	8,500
G-4B(Ap)	14	6	2.0	5	20	3.9	1.8	2.2	1,486	15	—
G-4B(B21t)	33	9	3.0	8	21	4.5	2.6	2.3	1,878	19	—
G-4B(B22t)	50	12	4.0	5	19	4.6	1.8	2.1	2,004	19	—
G-4B(B3)	75	13	4.3	10	19	4.7	3.2	2.1	2,912	27	2,400
G-6B(A)	8	9	3.0	8	22	4.1	2.7	2.4	1,331	14	—
G-68(B1)	20	9	3.0	9	27	3.9	3.1	3.0	157	2	—
G-6B(B21t)	34	7	2.3	6	12	2.8	2.0	1.3	847	9	—
G-6B(B22t)	56	5	1.7	3	10	1.9	.9	1.1	639	7	—
G-6B(B23t)	88	5	1.7	4	4	1.3	1.5	.5	836	10	—
G-6B(B3)	125	3	1.0	2	7	.7	.8	.8	598	7	500
G-8D(Ap)	24	12	4.0	11	56	4.7	3.8	6.2	2,162	24	—
G-8D(Bt)	43	10	3.3	10	57	4.7	3.4	6.3	2,073	22	—
G-8D(75)	75	25	8.4	26	81	5.2	8.6	8.9	4,516	46	5,000
G-8D(C)	80	16	5.3	18	77	5.5	6.0	8.5	1,080	11	—
G-8D(Cr)	95	13	4.3	12	39	5.4	3.9	4.3	3,088	28	—
G-9A(B1)	33	1	.3	2	5	1.2	.8	.6	250	3	—
G-9A(B22t)	85	2	.7	2	9	1.4	.6	1.0	373	4	—
G-9A(B32)	165	2	.7	5	2	3.5	1.6	.2	307	3	—
G-9A(C)	250	10	3.3	9	2	2.9	3.1	.3	2,421	29	1,250
G-10(B)	5	35	11.7	42	89	5.4	14.0	9.8	7,479	78	7,250

quartz-feldspar gneiss. The results from the radon emanation experiment and laboratory analysis support these general conclusions.

Figure 39 shows the relation between uranium and radium. Except for the outlier values, uranium and radium in the soils are near secular equilibrium, as suggested by the high degree of correlation shown by the regression line on figure 39. This correlation indicates that no geochemical enrichment or depletion of uranium and radium has occurred in these soils. The two outlier values come from a

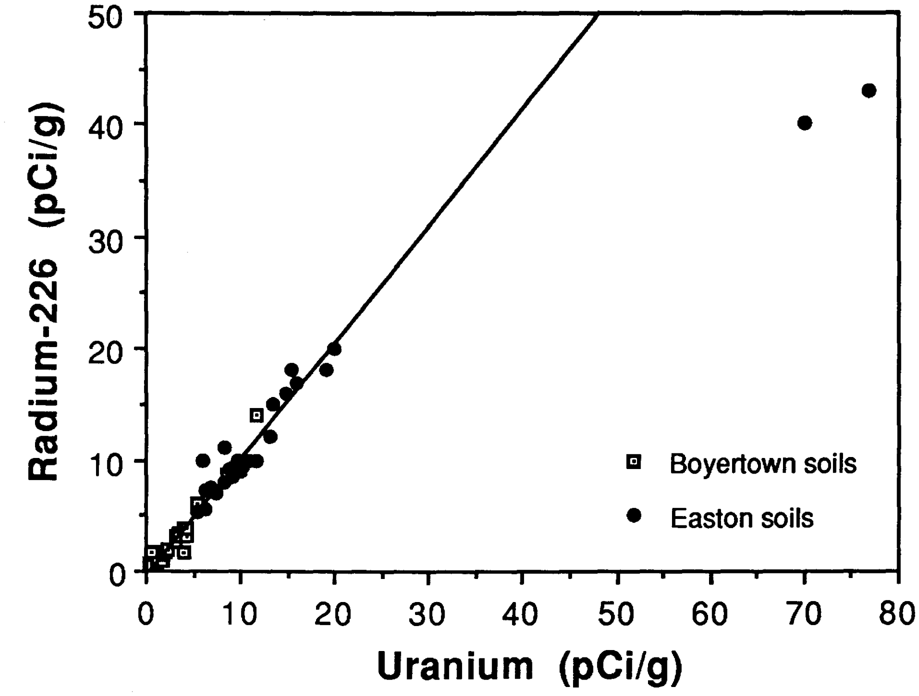


Figure 39. The relationship between uranium and radium for soils from Boyertown and Easton, Pa. The regression line for this set of data, excluding the anomalous outliers, shows that uranium and radium are in or near secular equilibrium. R^2, the correlation coefficient, is 0.953. pCi/g, picocuries per gram.

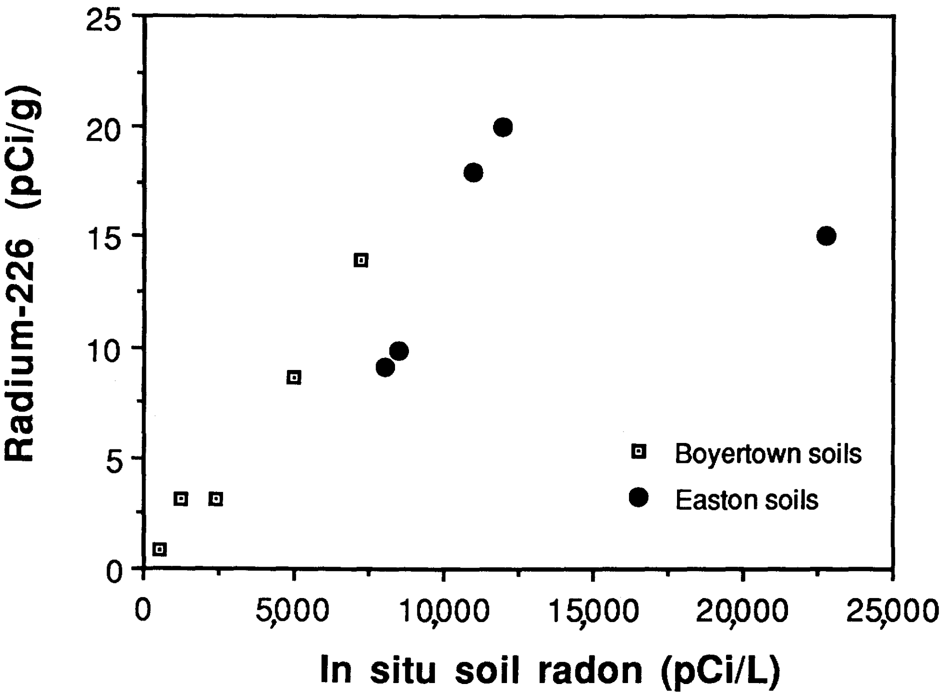


Figure 40. The relationship between radium and in situ radon activity measured at 75 cm in depth in selected soils. pCi/g, picocuries per gram.

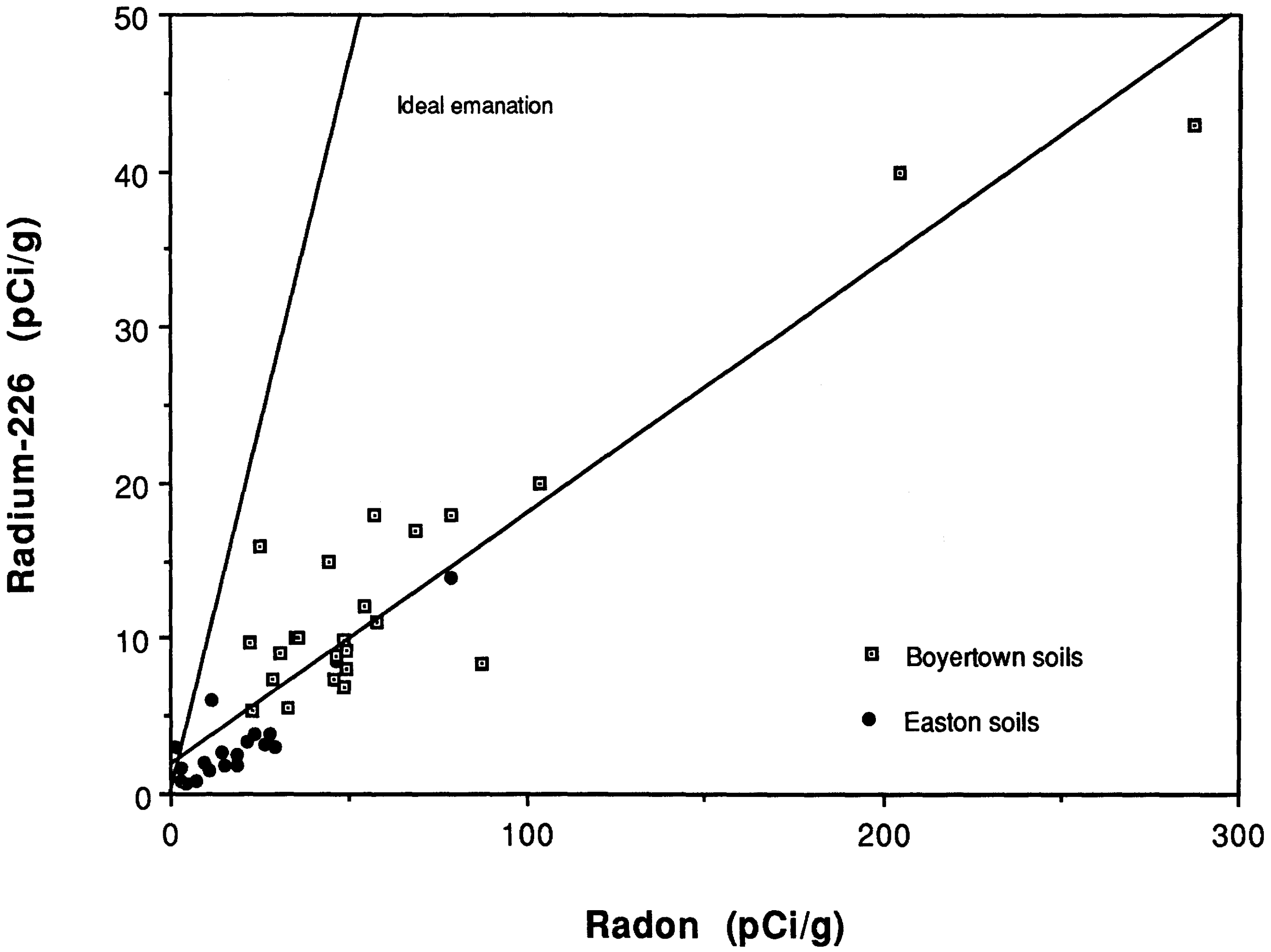


Figure 41. The relationship between radium and radon, as measured from soil emanation experiments. The ideal emanation line assumes that all radon produced is available to the soil air. R^2, the correlation coefficient, is 0.863. pCi/g, picocuries per gram.

site that had been scraped down to very permeable weathered bedrock; apparently some of the radium has been leached from this site. Note that the soils from Easton generally have more uranium and radium than those from Boyertown.

Figure 40 shows a positive correlation between radium concentrations and in situ radon activity measured at 75 cm in depth in the soils. The overall positive correlation reflects the fact that only the soils are producing radon and that transient radon from other sources is not a factor. The anomalous point can be accounted for readily by higher permeability, probably related to the extensive joint system present in the Easton rocks. Figure 41 shows the relationship between radium concentrations and radon as measured in the emanation experiment. Although the soils differ texturally, the emanation of the dry soils was similar. Again, the values were higher for Easton than for Boyertown. These results indicate that, in saprolitic metamorphic terrains exposed below the glacial limit, radon in soil gas directly reflects the uranium content of the rocks. Permeability does not differ by more than an order of magnitude, so emanation experiments, such as the one discussed here, can be used to measure radon production of a soil in lieu of field measurements of radon. These measurements of radon in soil, both in situ and laboratory, can be compared favorably to the indoor radon values and thus used predictively. Figure 42 shows radon in soil gas and total activity of radon measured in the emanation experiment plotted against indoor radon for seven of the home sites. The anomalous data point at 22,750 pCi/L should theoretically have an indoor radon value much higher than that measured. Here, housing construction has probably played an important role.

SUMMARY AND CONCLUSIONS

The most significant overall controlling factor of the availability of radon is the uranium and radium content of

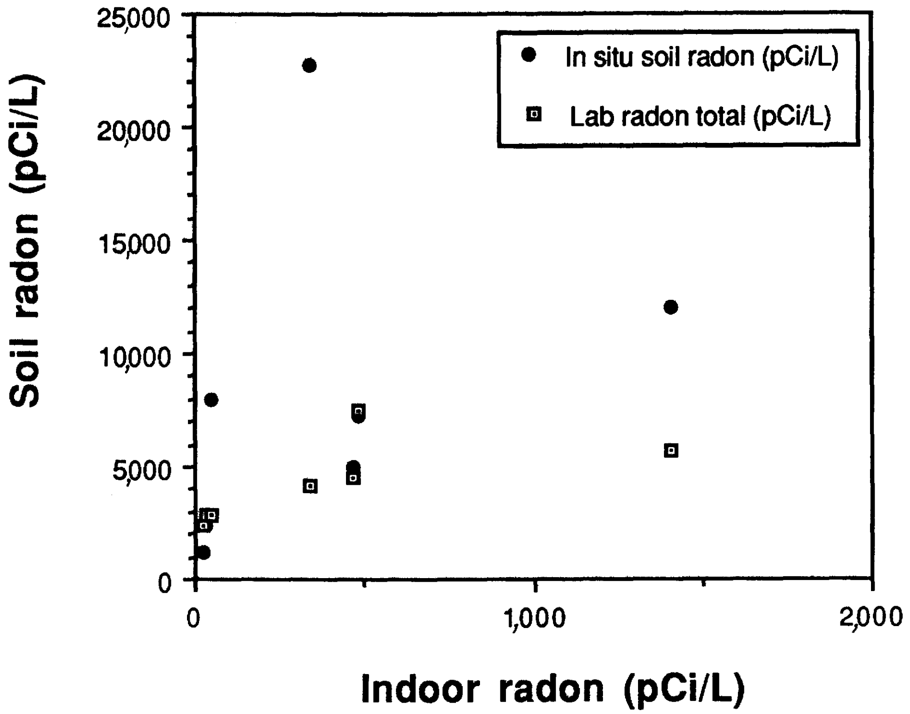


Figure 42. Radon in soil and total activity versus indoor radon. pCi/L, picocuries per liter.

the bedrock and soil materials. Radon migration is controlled by many factors but mainly by permeability. In this study, the permeabilities of the subsurface materials appear to be more than sufficient to allow radon to migrate freely. The soil environment into which a house is built is highly variable and therefore difficult to characterize. As stated previously, many of the in situ tests for radon in soil gas are drawn from the B and C horizons, and differences attributed to inherent differences in soils between *areas* may in some cases be due to differences between *horizons*. This observation could account for the variability seen within geologic units and some of the variability between geologic units. However, the geology and geochemistry of the soils generally provide a stronger overprint than the difference between soil horizons within rock units, so the prediction of *ranges* of soil-gas and indoor radon for a particular geologic unit is possible (similar results were found by Schumann and Owen, 1988). Distinctions can be made only where magnitude differences in geochemistry exist. For example, in Boyertown, the difference in radon concentrations, both indoors and in soils, is striking between the hornblende gneiss and the mylonite. The two rock types within the QFB, however, are not statistically distinguishable. In Easton, the difference between the highest readings in the biotite gneiss and the quartz-feldspar gneiss is an order of magnitude; however, the sillimanite gneiss and the marble are statistically indistinguishable because of the geological variability within each unit. Careful examination of the physical and chemical properties of the soil must be made in order for values of radon in soil gas to be used effectively.

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RADON IN SOIL GAS AND GAMMA-RAY ACTIVITY OF ROCKS AND SOILS AT THE MULLIGAN QUARRY, CLINTON, NEW JERSEY

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Abstract

Total-count gamma-ray data, gamma-ray spectrometer data, and soil-gas radon measurements were made at the Mulligan Quarry, Clinton, N.J. Although the entire area shows relatively high values of radioactivity, some areas within the quarry show radioactivity levels that are greater than twice background with all three measuring techniques used. Radon in soil-gas measurements generally shows high values in locations where gamma-ray activity is high, but this is not always the case. One area shows a high radon soil-gas value that corresponds to a relatively low equivalent uranium value, and another area shows a high equivalent uranium value that corresponds to a relatively low radon value.

High gamma activity and high radon in soil-gas concentrations appear to be related to mineralization that occurs in a shear zone at the southern end of the quarry and may be related to fracture-filling material throughout the entire quarry. Soil development on top of the quarry prevents direct tracing of the fractures seen on the working face and may broaden any radon and (or) gamma-ray highs associated with these concealed fractures.

INTRODUCTION

Recognition of the health hazards that may result from inhalation of radon or its daughter products has prompted many recent studies of factors related to the concentration of radon inside homes and other buildings. The Environmental Protection Agency (EPA) has established 4 pCi/L (picocuries per liter) as the maximum acceptable level for indoor radon (EPA, 1986).

Details of the formation of radon from the decay of uranium and the health hazards associated with breathing it and its particulate decay products may be found elsewhere in this volume and are not repeated here. It is sufficient, for the purpose of this paper, to state that radon-222 is a naturally occurring, radioactive, noble gas formed in the uranium-238-radium-226 decay series and that inhalation of radon-222 may cause thousands of deaths from lung cancer each year (EPA, 1986).

Several areas in the United States are known where the radon concentration in homes is many times this maximum level. One such area is in Clinton, N.J.

Mulligan Quarry in Clinton, N.J., offers a unique opportunity to study radon concentrations in both soil and bedrock in a region where private homes have elevated levels of radon. Located just east of the homes, the inactive quarry has a face of exposed rock, and at the upper level of the quarry the same type of soil exists as is found in the housing development (Patrick, 1920).

The purpose of this study was to investigate the distribution of radioactive material in the rock and soil and of radon in the soil gas in this area. Information from this study should add to the general knowledge of indoor radon problems and their relation to geology. These data should have application in the evaluation of future building sites in similar areas or in the formulation of predictive models to identify potential problem areas.

GEOLOGIC SETTING

The Reading Prong province lies between the Valley and Ridge and the Piedmont provinces (fig. 43). Ridges of Middle Proterozoic mixed gneisses and granitoid bodies separated by intermontane belts of Cambrian and Ordovician sedimentary rocks compose the Reading Prong province of New Jersey (Drake, 1970).

Cambrian and Ordovician carbonates of the Kittatinny Supergroup contain a basal arkose unit that unconformably overlies the Middle Proterozoic rocks. The arkose grades upward into a sequence of platform carbonates containing predominantly dolomite with lesser amounts of limestone and chert. Dolomite of the upper part of the Kittatinny Supergroup was the rock worked at the quarry. Here the dolomite is thin to medium bedded, fine to medium crystalline, with minor discontinous quartz sand lenses, bedded chert, and black dolomitic shale.

Stratigraphically and structurally above these carbonates is a Middle Ordovician sequence that contains coarse granular sandstone, black, red, and green shales, and minor interbedded limestone (Markewicz, 1967). This sequence has been described as both autochthonous flysch deposits (Kummel, 1940; Markewicz, 1967) and far-traveled Jutland klippe related to the Taconic deformational period (Drake, 1969; Rogers, 1971; Zen, 1972; Perissoratis and others, 1979).

Unconformably overlying all older rocks is the Newark Supergroup of Late Triassic and Early Jurassic age (Van Houten, 1969). Locally, only the Passaic Formation of Late Triassic age is present. This formation consists of fanglomerate and red shale that blanketed the entire study area but were eroded to their present location south of Mulligan Quarry (fig. 43).

Three major deformation periods juxtaposed the geologic units into their present positions. Generally, the

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[2] New Jersey Geological Survey, Newark, N.J.

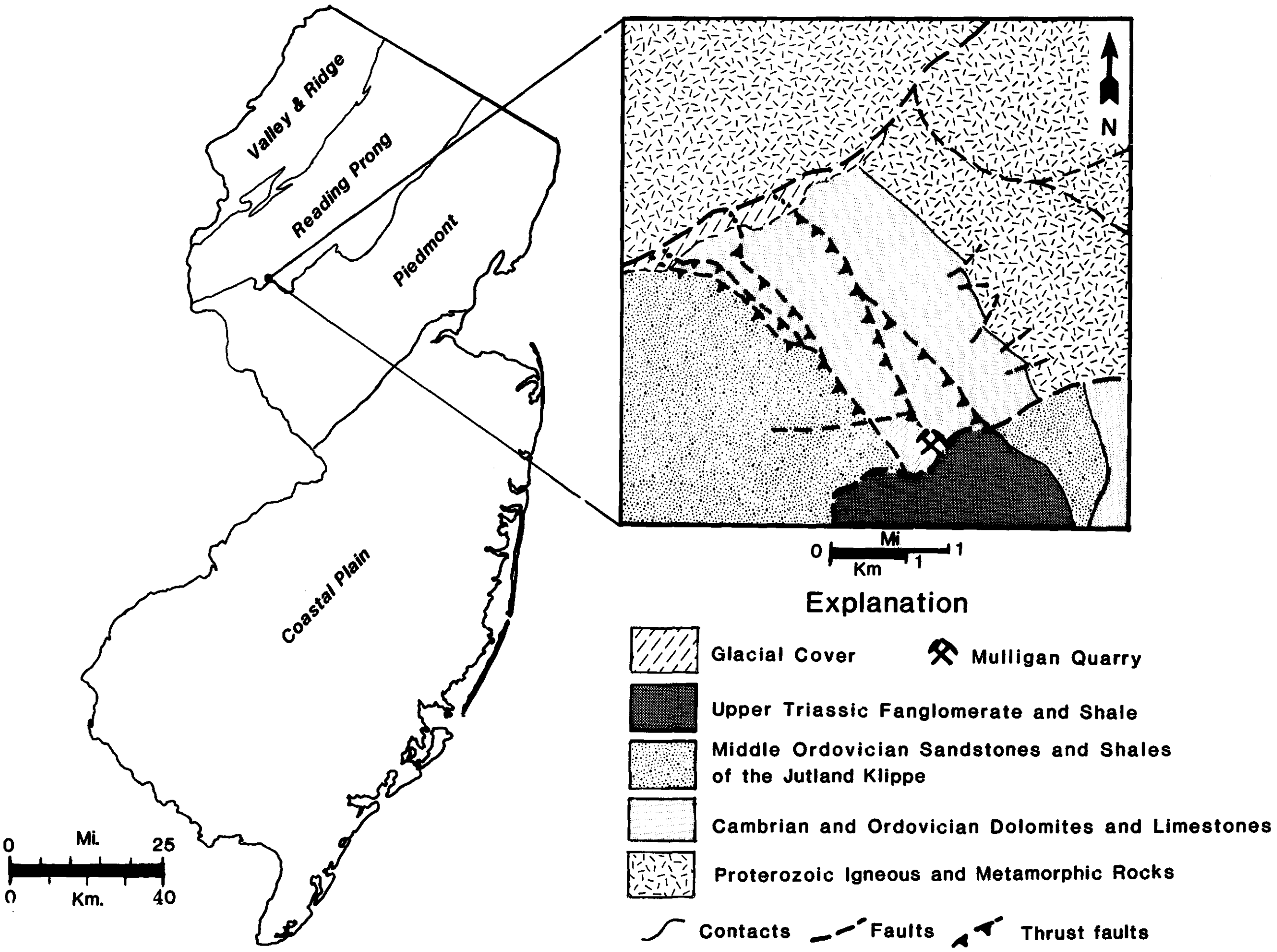

Figure 43. Location of study area, Mulligan Quarry, Clinton, N.J.

Proterozoic and Paleozoic units were folded and thrust faulted during the Taconic and Alleghenian orogenies. In the study area, the thrusts and beds generally strike northwest and dip to the southwest. Regionally, northeast to east-northeast thrust faults occur, which separate the Proterozoic and Paleozoic rocks.

In the Mesozoic rifting, initiation of the breakup of Gondwanaland reactivated some east-northeast-striking regional thrust faults as listric normal faults (Ratcliffe, 1980; Ratcliffe and others, 1986). Associated with this extensional period, east-west-striking, high-angle normal and reverse faults formed, further complicating the structure. Mesozoic rocks strike east-northeast and dip moderately northwest. Structurally, all three deformational periods affected the rocks of Mulligan Quarry, but high-angle normal and reverse faults predominate.

GENERAL URANIUM DISTRIBUTION

Uranium concentrations are of primary concern for this study because it is this element that generally determines the maximum amount of radon-222 that can be produced from a given material. The average crustal abundances of uranium and thorium in continental sialic rocks are 3 and 10 ppm (parts per million), respectively (Polanski, 1965). Uranium occurs naturally in a variety of minerals and rock types. It tends to occur in those igneous rocks that are richest in silica and alkaline components. A primary ore of uranium is pitchblend, and it is often concentrated in minerals such as monazite, zircon, and apatite (Polanski, 1965). Uranium also occurs in a wide variety of secondary minerals such as carnotite, autunite, tyuyamunite, and uranophane. Areas that contain uranium-bearing rocks or minerals in the shallow subsurface, or are covered by natural soils derived from these materials, have the potential to be radon problem areas.

Carbonate rocks, particularly limestone and dolomite, normally contain low concentrations of uranium. Very pure calcium carbonate minerals normally contain about 1 ppm uranium; however, some carbonate rocks may contain greater than 10 ppm uranium that is associated with noncarbonate (heavy-metal resistates, sedimentary or marine apatite, fluorite, or some types of organic matter) constituents (Bell, 1963).

Carbonate rocks exposed at the Mulligan Quarry, near homes that have elevated indoor radon values, were studied by McKeown and Klemic (1953). These writers described a 15-ft-wide shear zone near the southern end of the quarry which, according to their report, consisted of a cemented limestone breccia. Within the major shear zone they noted a structurally younger, 2- to 3-ft-wide shear zone that consisted of crushed and fractured limestone. The shear zone displayed elevated radioactivity (no specific values were given) as far as it could be traced, but the radioactivity decreased to the southwest. They also reported analyses that showed uranium values from material presumably within the shear zone and from a black limestone about 500 ft north of the shear zone of 680 and 460 ppm, respectively. Uranium-bearing apatite was identified in this area by the authors.

Bell (1963) also briefly discussed the Mulligan Quarry. He described the rock exposed at the quarry as dolomite, or at least of the same chemical composition as dolomite, of the Kittatinny Limestone, now named the Kittatinny Supergroup. He also noted the presence of an unidentified yellow powdery uranium mineral, apatite, and phosphatic rocks at the quarry. Bell reported data from two samples taken from unspecified locations at the quarry that show uranium concentrations of 6 to 20 ppm.

Popper and Blauvelt (1980) determined that the radioactive minerals present at Mulligan Quarry were apatite and hydroxyapatite and that their distribution appeared to be controlled by bedding surfaces and fractures. C. Bell (unpub. data, 1987) also suggested that uranium mineralization is structurally controlled.

METHODS OF STUDY

Three methods were used to measure radioactivity from rock and soil and the concentration of radon from soil-gas samples. Each method is briefly described below.

Total-count gamma-ray measurements were made with a Geometrics Gamma Ray Scintillometer, Model GR101A. Data from this instrument are given in counts per second (CPS) and show relative variations in total gamma-ray intensity only. Total-count scintillometer data contain information from several radionuclides collected simultaneously and do not distinguish among the individual elements. Because of the reports in the literature (McKeown and Klemic, 1953; Bell, 1963) of the association between elevated radioactivity values and uranium concentration, we used the scintillometer to make a preliminary survey of the area.

The gamma-ray spectrometer partitions counts, based upon gamma-ray energy, and allows one to determine the energy and the amounts of potassium-40, bismuth-214, and thallium-208 present at each location. The concentrations of the two latter nuclides are proportional, respectively, to the concentrations of uranium-238 and thorium-232 present if secular equilibrium is assumed. During each measurement, the instrument was placed on the ground and counts were collected for 300 seconds. The counts that were recorded from the three channels were converted to a percentage of potassium-40 and an equivalent parts per million value for uranium-238 (eU) and thorium-232 (eTh).

Soil-gas samples were collected by driving a pencil-sized hollow steel probe into the soil to a depth of about 75 cm (Reimer and Bowles, 1979) and withdrawing a 10 cm^3 (cubic centimeter) sample of the soil gas into a syringe for radon analysis. The soil gas was then injected into a portable scintillometer that measured the decay events of the sample resulting from radon-222. A minimum of 10 minutes was allowed before analysis of each sample, to eliminate the contribution by other short-lived radon isotopes. The maximum time between collection and analysis of each sample was about 5 hours. Radon-222, with a half-life of about 3.8 days, would suffer a decay loss of less than 5 percent of the original number of atoms collected during this time. Radioactivity of these samples was measured for a period of 1 minute and is given in picocuries per liter (pCi/L). Two instruments were used for these measurements, one manufactured by Bonder Kleg, the other by EDA (EDA RDA–200). Duplicate samples measured on each instrument agreed to within 10 percent of one another.

A small area near the southern end of the quarry between the creek and the base of the cliff was used to establish an arbitrary background level for the measuring techniques used. Background for the total-count scintillometer was determined to be about 60 CPS, background for the gamma-ray spectrometer was determined to be about 24 ppm eU, and the background value used for radon in soil gas was 2,600 pCi/L. It is acknowledged that this entire area is high in terms of radioactivity and could be considered one large anomaly; however, in order to study the distribution of uranium in soil and rock, the distribution of radon in soil gas, the relation between the two, and their relation to the geology, we adopted the term "anomaly" to represent an area that was at least twice the background value for that particular variable.

RESULTS AND DISCUSSION

Gamma-Ray Scintillometer Survey

The location of each traverse of this survey is shown in figure 44. Traverse 1 (stations 10 to 28) was made near the creek bank, traverse 2 (stations 29 to 49) was made near the base of the quarry face, and traverse 3 (stations 50 to 70)

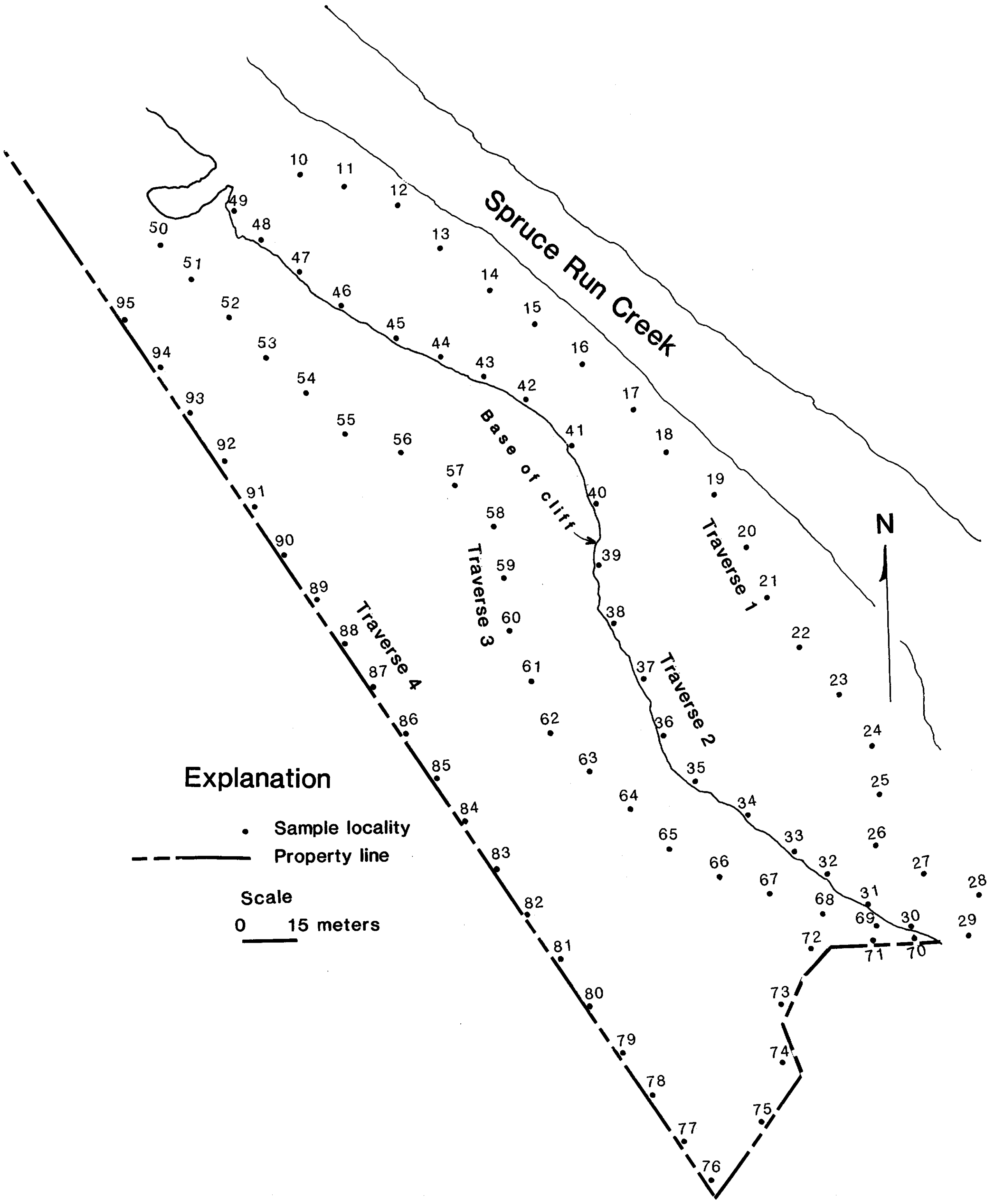

Figure 44. Locations of stations along traverses from a survey made with a gamma-ray scintillometer, Mulligan Quarry, Clinton, N.J.

and traverse 4 (stations 71 to 95) were made on top of the quarry, which stands 15 m or more above the creek.

Total-count gamma-ray scintillometer data collected on traverse 1 show values (mean value = 77 CPS) generally the same as background (background = 60 CPS) and show little variability (range = 40–110 CPS). This result may be related to relatively homogeneous alluvial deposits blanketing the bedrock near the creek bank.

Traverse 2, made along the base of the quarry face, shows a greater range in gamma-ray intensity (25–180 CPS) but similar average of all measurements (mean value = 72 CPS) as compared to traverse 1. Low gamma-ray readings here are probably related to the contribution of dolomite in the quarry face (which is generally not highly radioactive except where fractured) to the total signal. Zones of elevated gamma-ray intensities are associated with open fractures or what appear to be recemented breccia zones.

Traverse 3 was made on top of the quarry. Gamma-ray intensity along this traverse shows the greatest range of the four traverses (40–350 CPS), with the highest value measured, and a mean value (131 CPS) of about twice the background value.

Traverse 4 of the survey also was made on top of the quarry along the western property boundary of the Clinton Historical Museum. These gamma-ray measurements have a mean value of 148 CPS, greater than two times background, and a range in values of 50–250 CPS, intermediate between the ranges for traverses 2 and 3.

Four areas of elevated gamma-ray intensities (greater than two times background) are shown in figure 45 The two areas at the southern end of the quarry (A and B) are near the large radioactive shear zone mentioned previously.

Area C is located along the projected strike of two prominent fracture sets, neither of which shows elevated gamma-ray intensity at the quarry face. This area may be related to the non-uniform distribution of uranium along fractures projected into the quarry face. Area D is located near the north end of the quarry and lies along the projected trace of a prominent fracture set that displays elevated gamma-ray intensity at the quarry face.

Gamma-Ray Spectrometer Survey

Data were collected with a gamma-ray spectrometer along two traverses (5 and 6) on top of the quarry (fig. 46). Traverse 5 was made between traverses 3 and 4, and traverse 6 was made along traverse 4.

The calculated uranium concentration (eU) along the two traverses is shown in figure 47. These values range from about 5 to over 70 ppm eU and have a mean value of about 28 ppm eU. If the same criterion for "anomalous values" is used with this data set as that used for the total-count gamma-ray data set (readings two times background), three such areas are identified in figure 47 (areas E, F, and G). All are located within the areas defined as anomalous by total gamma-ray scintillometer data but are smaller.

Radon Soil-Gas Survey

Results of the radon soil-gas measurements and the gamma-ray spectrometer data are listed in table 7 and are shown in figure 48. Radon values range from 300 to 20,700 pCi/L and have a mean value of about 5,600 pCi/L. The background radon value was about 2,600 pCi/L. Areas where radon activities were greater than two times background are also shown in figure 48. Three of the four anomalous areas (areas H, I, and J) overlap the anomalous areas identified from gamma-ray spectrometer data (eU) and total-count gamma-ray data.

A crude correlation between soil-gas radon values and equivalent uranium exists (fig. 49); however, some sample locations show high equivalent uranium values and low radon values. Also, some locations show low equivalent uranium values and high radon values. The lack of a better correlation between these two parameters may be due to variables that affect gamma-ray measurements, the particle size of the uranium-bearing minerals, or physical properties of the soil.

Two possibilities can explain the lack of correlation between equivalent uranium and radon values measured near the surface. Thicker soil over a localized uranium source would result in lower equivalent uranium values caused by gamma-ray attenuation and could cause lower radon values because of the increased migration path length. More moisture in the soil results in lower equivalent uranium values and lower radon values because wetter soil attenuates gamma-rays and retards radon mobility.

Data regarding soil thickness and moisture content were not collected, and effects from these variables cannot be determined. However, variability in gamma-ray data taken from exposed bedrock along the base of the quarry face indicates a non-uniform distribution of radioactive material in the host rock. This fact, plus the proximity of the gamma-ray and radon anomalies at the southern end of the quarry to the shear zone, suggests that the distribution of these anomalies is controlled, in large part, by the distribution of radioactive materials in the host rock.

SUMMARY

Results of this study show that generalizations resulting from averages of large classes of rock types (carbonates) can be misleading when details of specific locations are considered. With regard to radon, the evaluation of an area for future development does not appear to be a simple matter. These data show that significant variations of radon in soil gas can occur over relatively short distances and that

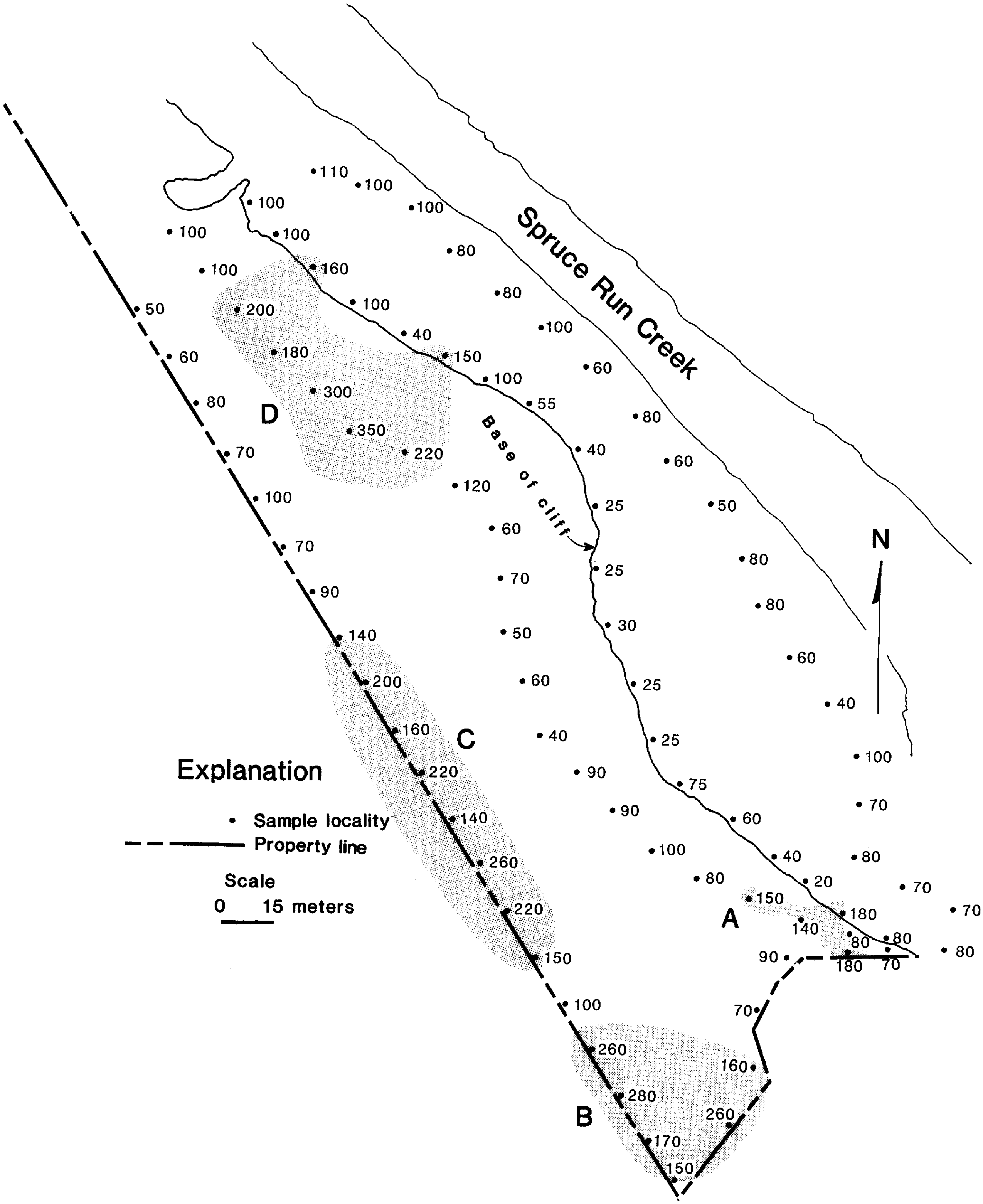

Figure 45. Gamma-ray scintillometer data, Mulligan Quarry, Clinton, N.J. Numbers shown indicate total gamma-ray intensity at that locality in counts per second. Stippled areas (A–D) indicate areas that are at least twice background level.

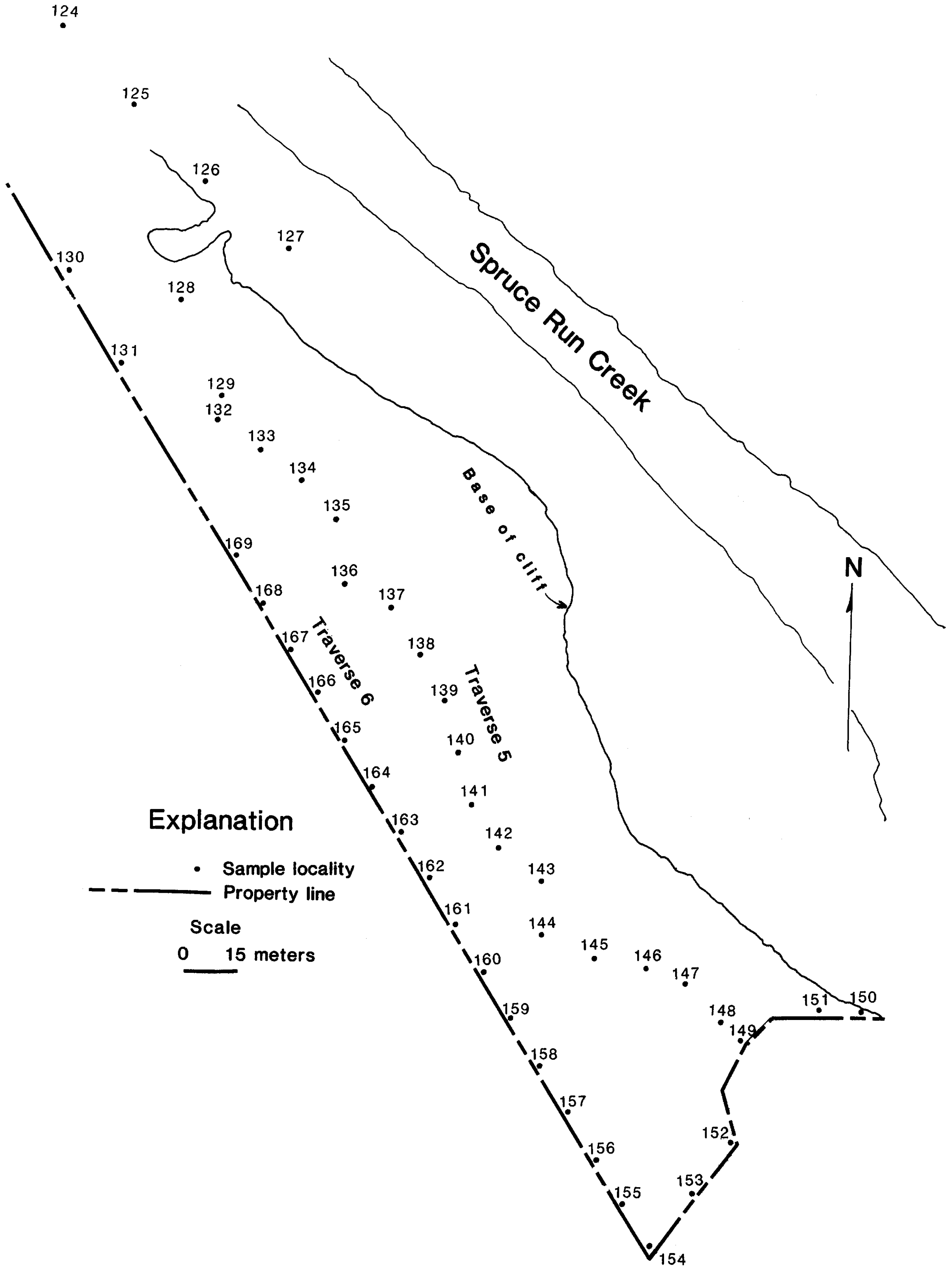

Figure 46. Locations of stations along traverses from a survey made with a gamma-ray spectrometer and soil-gas measuring equipment, Mulligan Quarry, Clinton, N.J.

Table 7. Gamma-ray spectrometer data and calculated concentrations of potassium, uranium, and thorium, Mulligan Quarry, Clinton, N.J.

[pCi/L, picocuries per liter; CPS, counts per second; %, percent; ppm, parts per million]

Sample	Radon (pCi/L)	Potassium (CPS)	Uranium (CPS)	Thorium (CPS)	Potassium (%)	Equivalent uranium (ppm)	Equivalent thorium (ppm)
124	8,850	3,177	1,687	726	1.8	13.6	20.3
125	4,200	2,727	902	336	2.1	7.6	9.0
126	2,500	3,127	2,704	550	.7	26.9	15.2
127	1,800	4,101	3,315	698	1.2	32.8	19.5
128	2,350	3,825	2,740	772	1.4	25.5	21.6
129	1,550	6,273	5,246	1,298	1.6	50.5	36.9
130	2,150	4,321	877	495	4.0	5.9	13.6
131	2,300	3,201	1,011	442	2.6	7.9	12.1
132	1,400	3,199	1,270	471	2.3	10.7	12.9
133	2,500	4,066	3,104	921	1.3	28.5	26.0
135	4,050	3,957	2,227	732	2.2	19.8	20.5
137	5,100	2,613	1,525	537	1.3	13.2	14.8
139	5,250	2,660	1,854	537	1.0	17.0	14.8
141	16,850	5,563	4,194	1,104	1.9	39.8	31.3
142	2,500	3,741	2,671	640	1.4	25.8	17.8
143	2,250	3,520	2,531	684	1.3	23.8	19.1
144	7,250	5,983	5,598	1,606	.9	52.1	45.8
145	5,300	3,091	2,270	815	1.1	19.6	22.9
146	4,000	2,972	1,912	882	1.3	14.9	24.8
147	2,200	1,807	1,228	559	.7	9.5	15.4
149	12,100	3,356	2,601	698	1.1	24.5	19.5
150	850	2,676	1,767	675	1.2	14.9	18.8
151	8,450	7,530	7,096	1,139	1.1	73.5	32.3
152	8,800	4,387	3,849	809	.9	38.2	22.7
154	12,900	3,504	3,058	813	.7	28.9	22.8
156	4,100	3,019	2,566	922	.7	22.2	26.0
157	8,000	3,893	3,380	1,042	.8	30.8	29.5
158	1,750	2,912	2,568	862	.6	22.7	24.2
159	4,350	2,976	2,579	705	.6	24.1	19.7
160	8,500	4,888	4,555	1,196	.7	43.3	33.9
161	20,700	8,573	8,214	2,342	1.1	76.7	67.2
162	6,900	7,198	7,127	2,189	.7	65.2	62.8
163	4,750	6,247	5,959	1,301	.8	58.9	37.0
164	900	7,506	7,092	1,449	1.1	70.9	41.3
165	11,900	4,665	3,942	997	1.1	37.7	28.2
166	350	3,227	2,264	739	1.3	20.2	20.7
167	650	2,246	884	446	1.6	6.4	12.2
168	1,700	3,370	717	344	3.1	5.3	9.2
169	300	2,912	1,089	429	2.2	8.9	11.7

they do not always occur in a predictable manner. A simple hand-held scintillometer might be used as a first-order survey tool if it is understood that excavation into the earth might expose problems not detected at the surface. Gamma-ray spectrometer data would serve to distinguish between areas of high uranium or high potassium concentrations. Soil-gas radon measurements are the most direct of the techniques used in this study to evaluate potential radon problems arising from construction of buildings in areas having elevated amounts of uranium in the soil or bedrock.

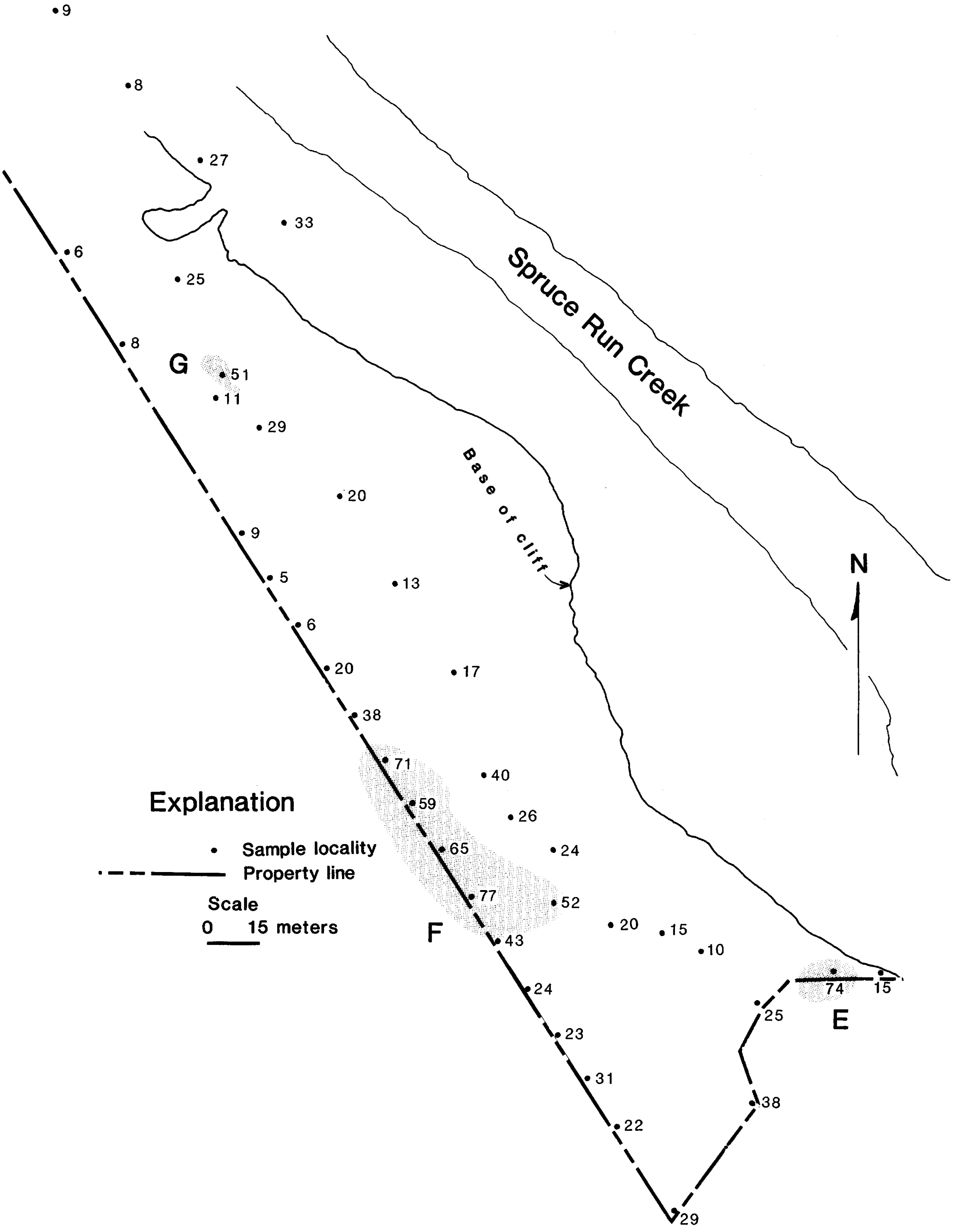

Figure 47. Equivalent uranium concentrations (eU) in parts per million, calculated from gamma-ray spectometer data, Mulligan Quarry, Clinton, N.J. Stippled areas (E–G) indicate areas that are at least twice background level.

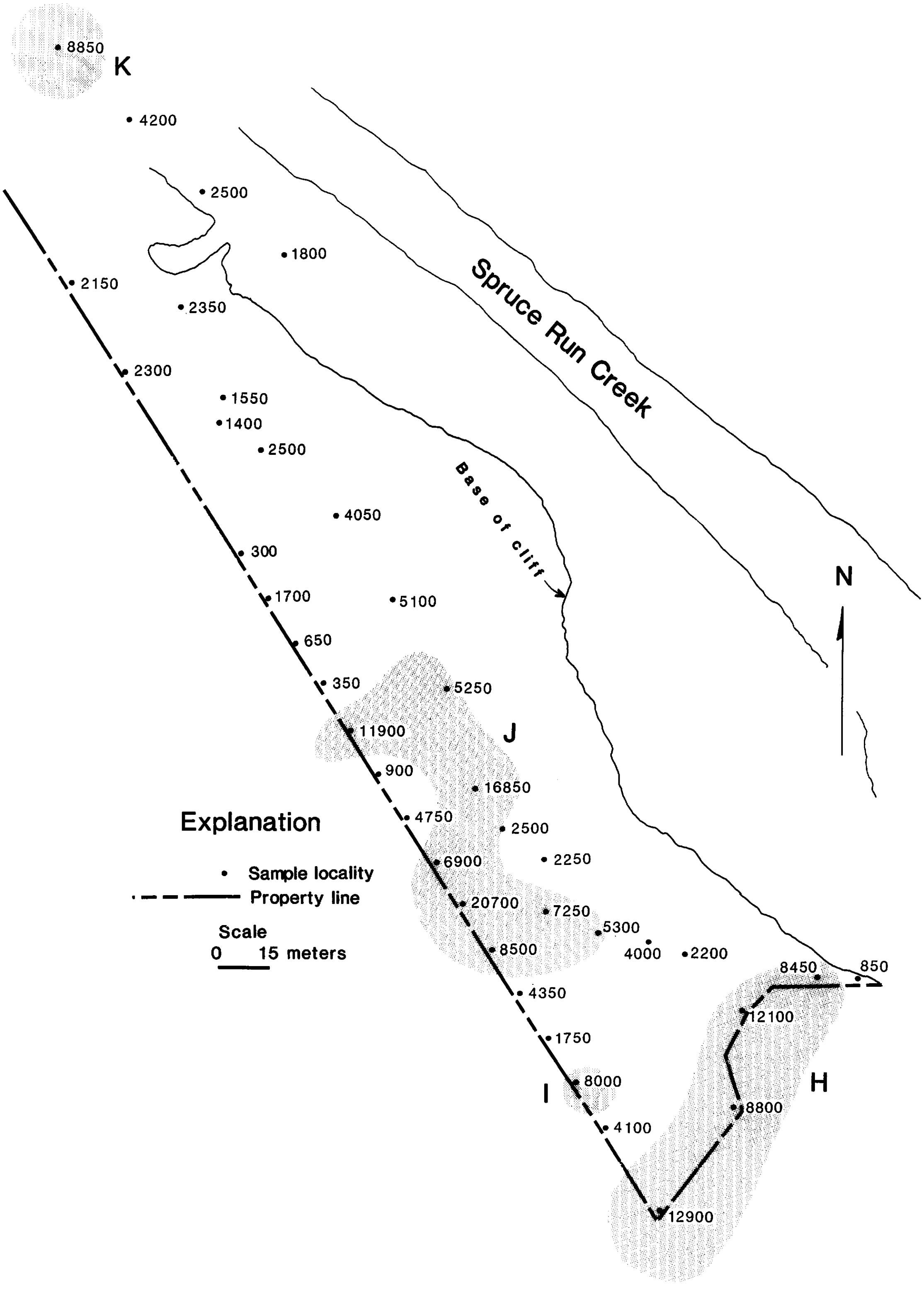

Figure 48. Radon-222 activity from soil-gas samples given in picocuries per liter, Mulligan Quarry, Clinton, N.J. Stippled areas (H–J) indicate areas that are at least twice background level.

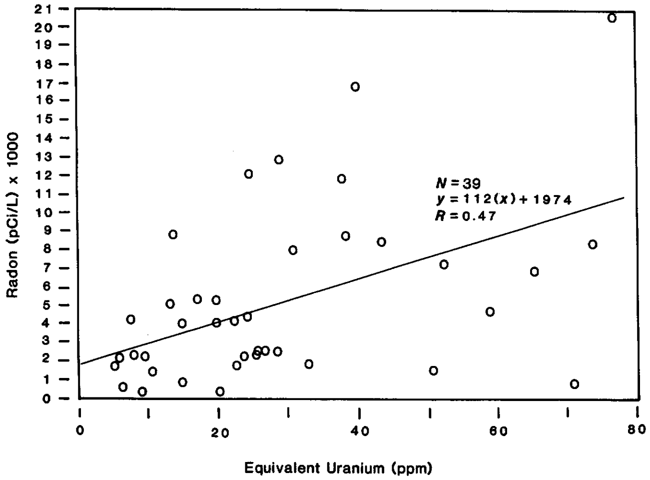

Figure 49. Cross plot of soil-gas radon-222 activities versus equivalent uranium values, Mulligan Quarry, Clinton, N.J. ppm, parts per million; pCi/L, picocuries per liter.

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RADON IN SOIL GAS ALONG ACTIVE FAULTS IN CENTRAL CALIFORNIA

Chi-Yu King, Calvin Walkingstick, and David Basler

Abstract

Radon concentration in subsurface soil gas was monitored by the Track-Etch method during the decade 1975 to 1985 at 60 sites along a 380-kilometer segment of the San Andreas fault system between Santa Rosa and Cholame in central California. The recorded radon concentrations show large spatial differences not attributable to differences in uranium content of local soil. The radon data also show large temporal variations, part of which are seasonal in appearance. The nonseasonal components of the radon variations may be correlated with occurrences of larger local earthquakes.

INTRODUCTION

Radon content of soil gas was monitored from 1975 to 1985 at a network of 60 stations along several active faults in central California in search of possible precursors to earthquakes. Some of the earlier results have been discussed by King (1978, 1980). In this section, we present the entire set of radon data and compare them with some local seismic and meteorological data but without detailed discussion on their correlation. This data set may be useful in helping to understand the variability of radon concentration in a near-surface environment, a problem pertinent to indoor radon pollution.

MONITORING SITES

The radon monitoring network consisted initially of 20 stations along the Calaveras and the San Andreas faults between Hollister and San Benito (site 19, fig. 50). It was later expanded in stages to include 60 stations that covered several major traces of a 380-km (kilometer) segment of the San Andreas fault system between Santa Rosa and Cholame (site 30, fig. 50). The station location and installation dates are shown in table 8 and figure 50. A detailed description of the site conditions may be found in Gaman and King (1979). Stations were also installed in the Geysers geothermal area and in southern California; these data sets will be reported elsewhere.

MONITORING TECHNIQUE

Radon concentration was monitored by the Track-Etch method developed by the Terradex Corporation. A small chip of cellulose nitrate, which is sensitive to alpha radiation, was attached to the inside bottom of a plastic cup 9 cm (centimeters) high with a 7-cm aperture. The cup was put upside down at the bottom of a shallow hole about 10 cm in diameter and 70 cm in depth to expose the chip to soil gas. The hole was supported by a capped plastic pipe about 80 cm long, which was also used to reduce the effect of rainfall, wind, and temperature variation on the radon measurement. After a period of a week to several weeks or longer, the cup was replaced by another, and the retrieved cup was sent to Terradex, where the chip was chemically etched and the alpha-particle tracks in the chip were counted under a microscope. The track density within an area of about 6 mm^2 (square millimeters) was then used as a measure of the average concentration of radon isotopes in the soil gas during the period of exposure.

The recorded radon time series at the 60 stations are shown in figure 51 in units of number of tracks per 100 'fields of view' per week, each unit being equivalent to 2.7 pCi/L (picocuries per liter). Longer exposure periods were used during latter years in order to cut cost. Three stations (Nos. 10, 11, and 31) changed locations, and only data recorded at the new locations are shown. Data gaps are caused by a variety of incidents such as vandalism. For a period of about 2 years beginning in mid-1982, chips of higher sensitivity (by a factor of 5) were used; the difference in sensitivity is corrected in the plotted time series.

The radon data can be compared with earthquake data in various ways. Because earlier data appeared to show correlation with larger earthquakes (King, 1978, 1980), we show in figure 51 the occurrences of earthquakes of Richter magnitude ≥ 4.0 within 50 km of each station on the same time scale.

To compare with meteorological data, we show in figure 52 the amounts of daily rainfall and maximum/minimum temperatures recorded at Gilroy (21 km northwest of Hollister), which is near the center of the radon-monitoring network. Most rainfall in the study area occurs in the winter months.

As reported by King (1980), the long-term average radon concentrations do not appear to depend strongly on the amounts of uranium or its "emanating" part in the local soils, as determined by gamma-ray spectroscopy with soil samples collected from the bottom of some monitoring holes (fig. 53).

DISCUSSION

As shown in figure 53, the soil-gas radon concentrations recorded at different stations were greatly different, and the difference amounted to an order of magnitude for sites of equal uranium content in the local soil. This spatial variation suggests that radon generation and migration in the ground are significantly affected by several other factors, such as permeability, porosity, water content, and topography.

The radon concentrations recorded at many sites also show large temporal, especially seasonal, variations (fig. 51). However, because the seasonal variations recorded at different sites are not always in phase with one another, it is suggested that variability of rain-water movement in the soil may be the main cause. Rain water may fill pores and reduce permeabiliy in a relatively thin layer at the surface above the measurement depth (0.7 meter) at some sites and thereby produce a cap that may increase radon content at the measurement level, or it may seep to a deeper-than-detector level and prevent radon generated below from reaching the detector at other sites (for example, site 46). Other meteorological parameters, such as barometric pressure and wind speed, probably have not significantly affected the radon data because of the integrated nature of our measurement over relatively long periods.

Some temporal variations are not seasonal in nature. For example, the maximum radon values recorded at site 6 occurred during winters of 1976–79 but during springs or summers subsequently. Because the episodic nonseasonal components of the temporal variations are commonly spatially coherent over long segments of the same fault but not necessarily along another fault nearby, it is suggested that they may be tectonic in nature. (For example, an episodic increase was recorded in the summer of 1978 at sites 1 and 2 on the Calaveras fault, where the radon concentration was extremely low in the summers of 1975 and 1976. Contemporary increases were recorded at several other sites (36–40) farther north along the same fault but not at the nearby sites 4–7 on the San Andreas fault.) Larger earthquakes appear to have a tendency of occurring during episodes of higher radon concentrations at least at some sites.

ACKNOWLEDGMENTS

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Table 8. Soil-gas radon monitoring sites, central California

Site	Name	Latitude (°N)	Longitude (°W)	Elevation (in meters)	Installation date
01	Bolsa Road, on Calaveras fault	36 53	121 25.4	70	5/7/75
02	Wright Road, on Calaveras fault	36 52	121 24.8	81	5/7/75
03	Nyland, on San Andreas fault	36 51	121 32.7	54	5/7/75
04	Artesian Well, on San Andreas fault	36 50	121 31.6	61	5/7/75
05	San Juan Bautista, on San Andreas fault	36 50	121 31.2	65	5/7/75
06	St. Francis Retreat, 25 m northeast of San Andreas fault	36 49	121 29.9	131	5/7/75
07	Harris East, on San Andreas fault	36 46	121 25.8	320	5/7/75
08	Harris West, 300 m southwest of San Andreas fault	36 46.6	121 26.0	274	5/7/75
09	Cienega Well, 70 m southwest of San Andreas fault	36 45.1	121 23.2	344	5/7/75
10	Thompson Creek, a few meters southwest of San Andreas fault	36 42.1	121 19.1	244	1/18/78
11	Pescadero Creek, on San Andreas fault	36 41.4	121 18.1	232	1/18/78
12	Frank Lewis, near San Andreas fault	36 39.9	121 16.3	229	5/7/75
13	Stone Canyon North, on San Andreas fault	36 38.2	121 14.3	247	5/7/75
14	Stone Canyon Hill, on San Andreas fault	36 38.1	121 14.1	287	5/7/75
15	Willow Creek, on San Andreas fault	36 35.7	121 11.4	317	5/7/75
16	Melendy Corral, on San Andreas fault	36 35.5	121 11.1	308	5/7/75
17	Melendy River, on San Andreas fault	36 35.3	121 10.9	311	5/7/75
18	Melendy Ranch, on San Andreas fault	36 35.0	121 10.6	317	5/7/75
19	San Benito, 30 m southwest of San Andreas fault	36 30.6	121 05.9	476	5/7/75
20	Dry Lake, on San Andreas fault	36 29.5	121 04.7	486	5/7/75
21	Little Rabbit Valley, a few meters southwest of San Andreas fault	36 25.6	121 00.8	549	3/30/76
22	Bitterwater Valley, on San Andreas fault	36 24.4	120 59.6	535	3/30/76
23	Hwy. 198, Mustang Ridge, on San Andreas fault	36 11.6	120 46.2	771	3/30/76
24	Parkfield Junction, 21 km northeast of San Andreas fault	36 04.9	120 28.6	363	3/30/76
25	Little Cholame Creek, 1.6 km northwest of San Andreas fault	35 57.2	120 28.5	546	3/30/76
26	Parkfield Bridge, on San Andreas fault	35 53.7	120 26.0	460	3/30/76
27	Turkey Flat, on San Andreas fault	35 52.7	120 24.9	445	3/30/76
28	Gold Hill, on San Andreas fault	35 49.4	120 21.0	381	3/30/76
29	Water tank, on San Andreas fault	35 45.4	120 18.4	349	3/30/76
30	Hwy. 46, 250 m southwest of San Andreas fault	35 44.0	120 17.4	348	3/30/76
31	Soda Lake, on San Andreas fault	35 54.3	121 36.6	43	5/16/78
32	Watsonville, on San Andreas fault	35 59.6	121 44.1	128	4/6/76
33	Laurel, 180 m northeast of San Andreas fault	37 06.3	121 54.0	567	4/6/76
34	Los Gatos, 400 m southwest of San Andreas fault	37 09.8	121 59.2	305	4/6/76
35	San Jose, 1.9 km southwest of Calaveras fault	37 22.6	121 47.2	311	4/6/76
36	Coyote, 3 km southwest of Calaveras fault	37 14.1	121 40.9	253	4/6/76
37	Anderson Lake, on Calaveras fault	37 09.1	121 34.9	213	4/6/76
38	North of San Felipe Lake, on Calaveras fault	36 59.7	121 28.5	162	4/6/76
39	Shore Road, on Calaveras fault	36 56.6	121 26.7	49	4/6/76
40	Pacheco, 60 m southwest of Calaveras fault	36 55.6	121 26.3	53	4/6/76
41	Spring Down Farm, on San Andreas fault	37 22.9	122 13.6	137	7/15/76
42	Pulgas Water Temple, 250 m northeast of San Andreas fault	37 29.0	122 18.9	98	10/19/76
43	San Andreas, 240 m southwest of San Andreas fault	37 34.6	122 24.5	114	10/19/76
44	Daly City, 1 km southwest of San Andreas fault	37 40.9	122 29.5	146	7/15/76
45	Richmond, on Hayward fault	37 59.4	122 22.3	6	12/15/76
46	Holy Redeemer College, on Hayward fault	37 45.5	122 09.4	46	8/26/76
47	Martinez, 3.6 km southwest of Concord fault	37 58.1	122 06.0	67	8/26/76
48	Dublin, near Calaveras fault	37 41.2	121 55.7	127	9/9/76
49	Calaveras Reservoir, on Calaveras fault	37 30.2	121 49.9	280	10/19/76
50	Fremont, on Hayward fault	37 32.2	121 57.4	18	9/9/76
51	Bolinas, on San Andreas fault	37 56.1	121 41.9	2	2/18/77, 6/8/77
52	Mile 22, on San Andreas fault	37 59.3	122 44.8	108	3/2/77
53	Pt. Reyes Ranger Station, on San Andreas fault	38 02.7	122 47.9	14	2/18/77
54	Bodega Head, 1.1 km southwest of San Andreas fault	38 19.6	123 03.4	15	2/18/77
55	Santa Rosa, 650 m northeast of Rodgers Creek-Healdsburg fault	38 28.2	122 41.0	107	2/18/77
56	Jct. Crane Canyon and Grange Roads, near Rodgers Creek fault	38 22.5	122 38.3	222	4/13/77
57	Willie Bird Turkey Farm, 300 m southwest of Rodgers Creek fault	38 14.2	122 30.7	88	4/13/77
58	Sears Point, 200 m northeast of Rodgers Creek fault	38 10.6	122 27.0	12	2/22/77
59	Cordelia, on Green Valley fault	38 12.2	122 09.2	34	3/2/77
60	Bencia, on Green Valley fault	38 06.0	122 06.3	18	2/22/77

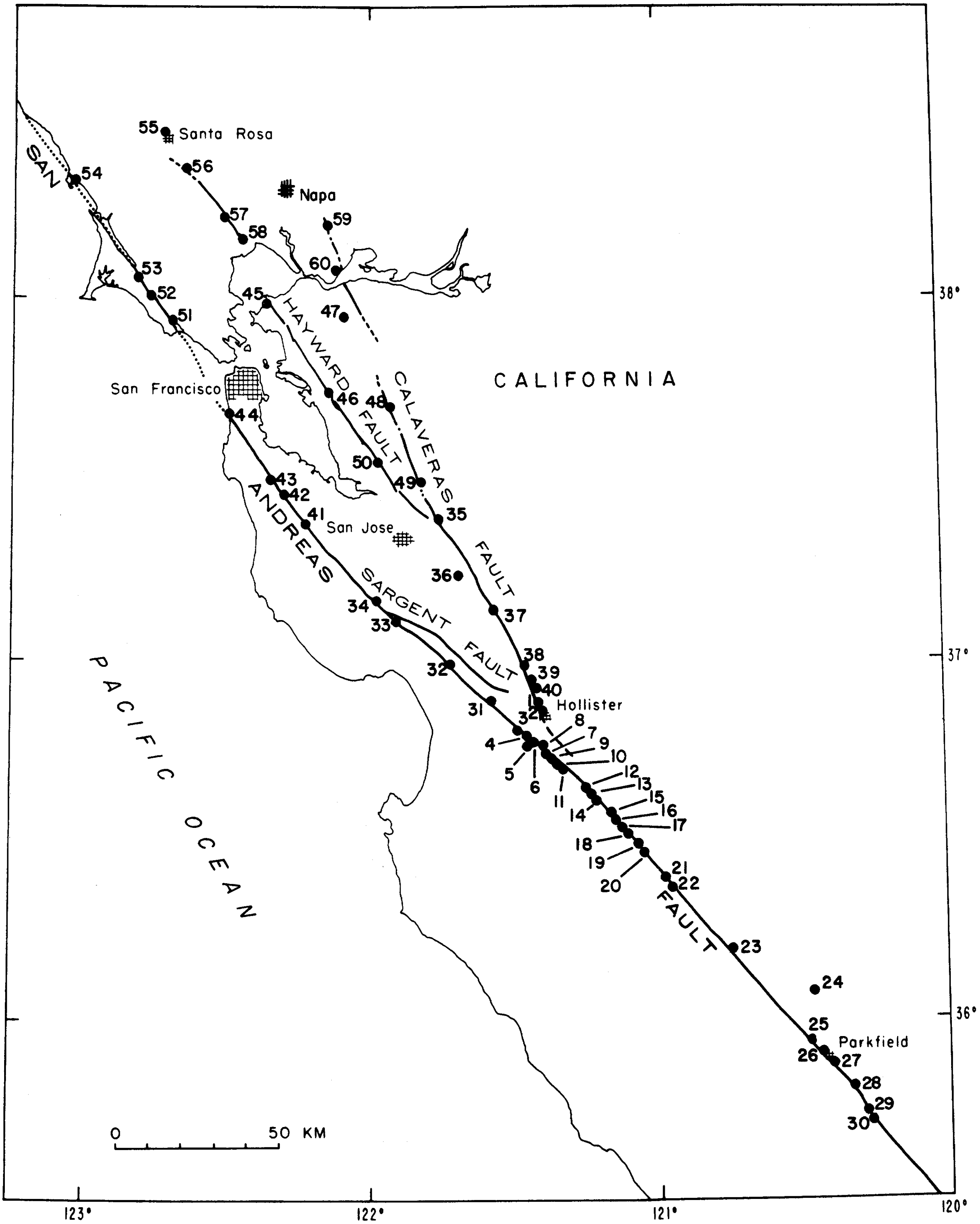


Figure 50. Location of soil-gas radon monitoring stations in central California (solid dots; identification numbers are in chronological order).

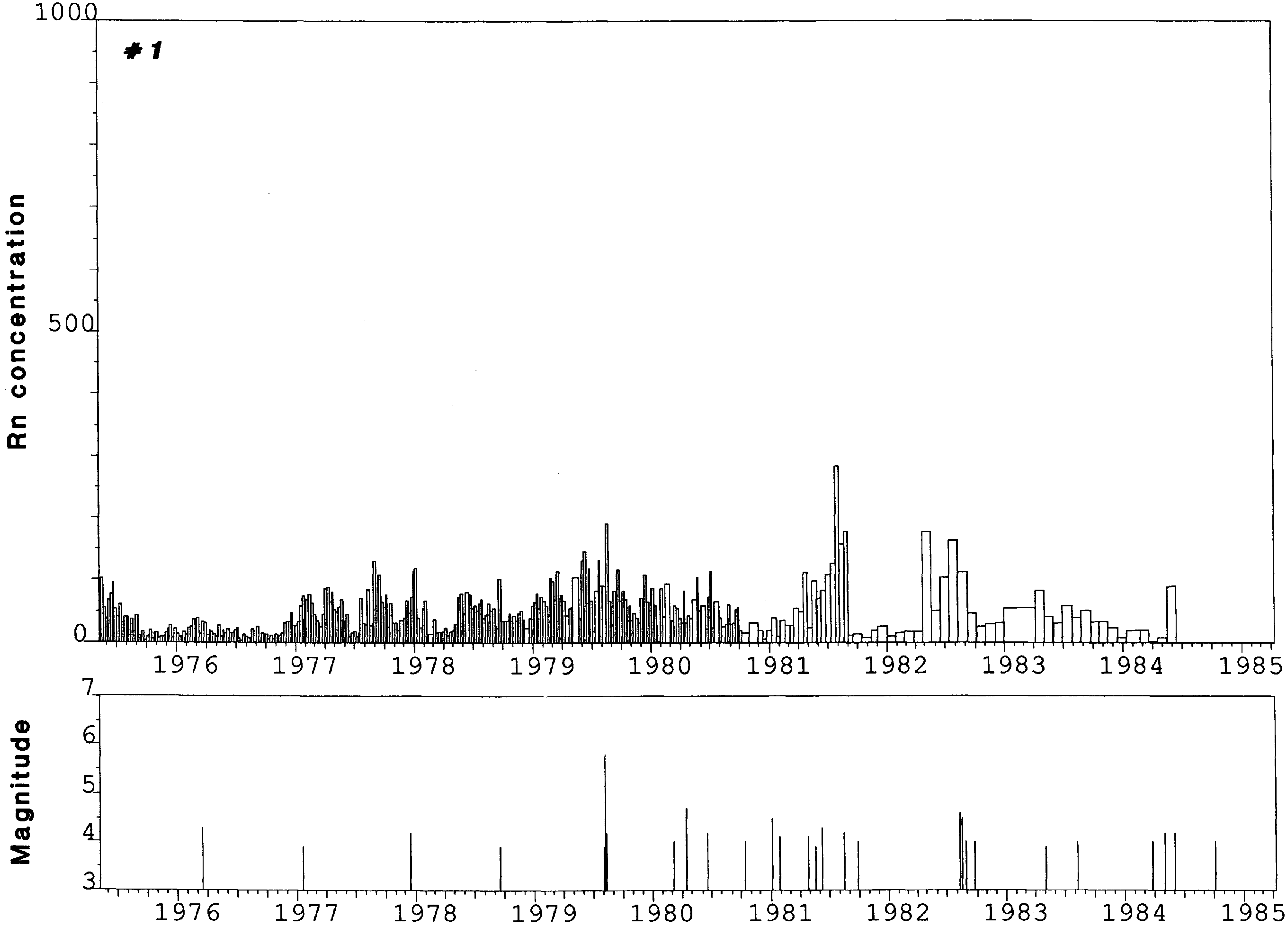
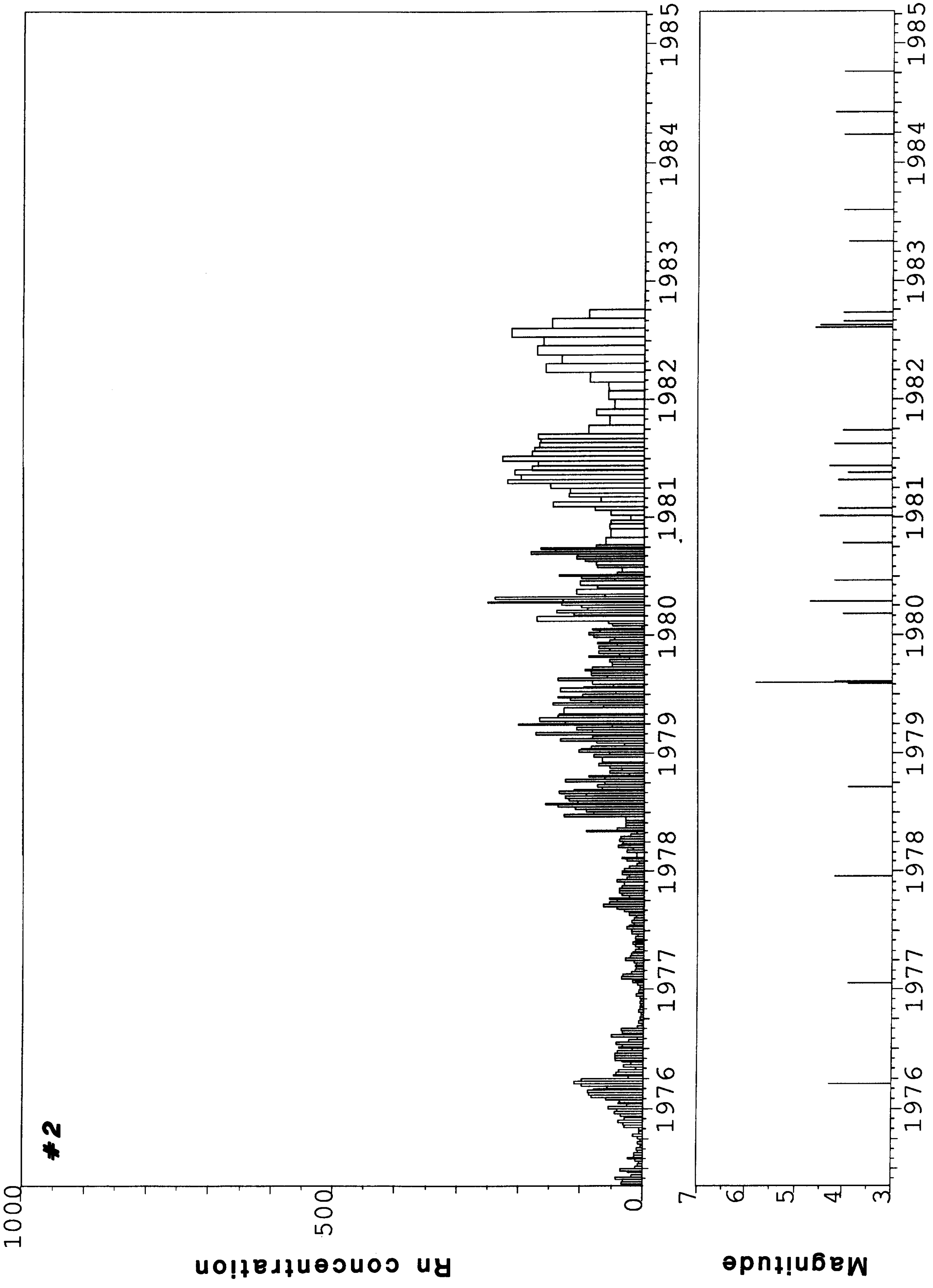


Figure 51. Time series of radon (Rn) concentration recorded at 60 stations along several active faults in central California. The concentrations are in units of number of tracks per 100 'fields of view' per week, each unit being equivalent to 2.7 picocuries per liter. The occurrence of earthquakes of Richter magnitude≥4.0 within 50 kilometers of each section is also shown on the same time scale.



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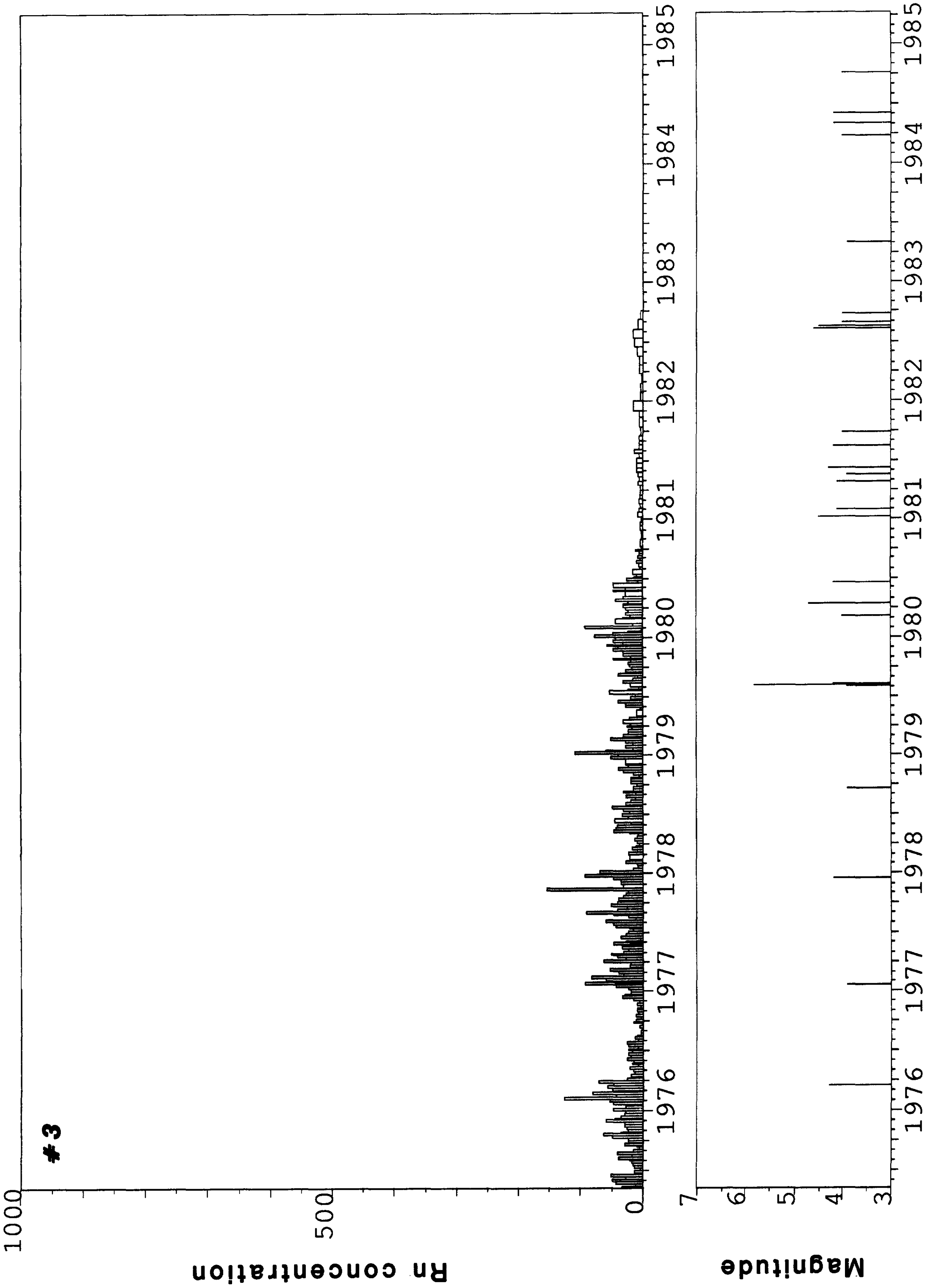

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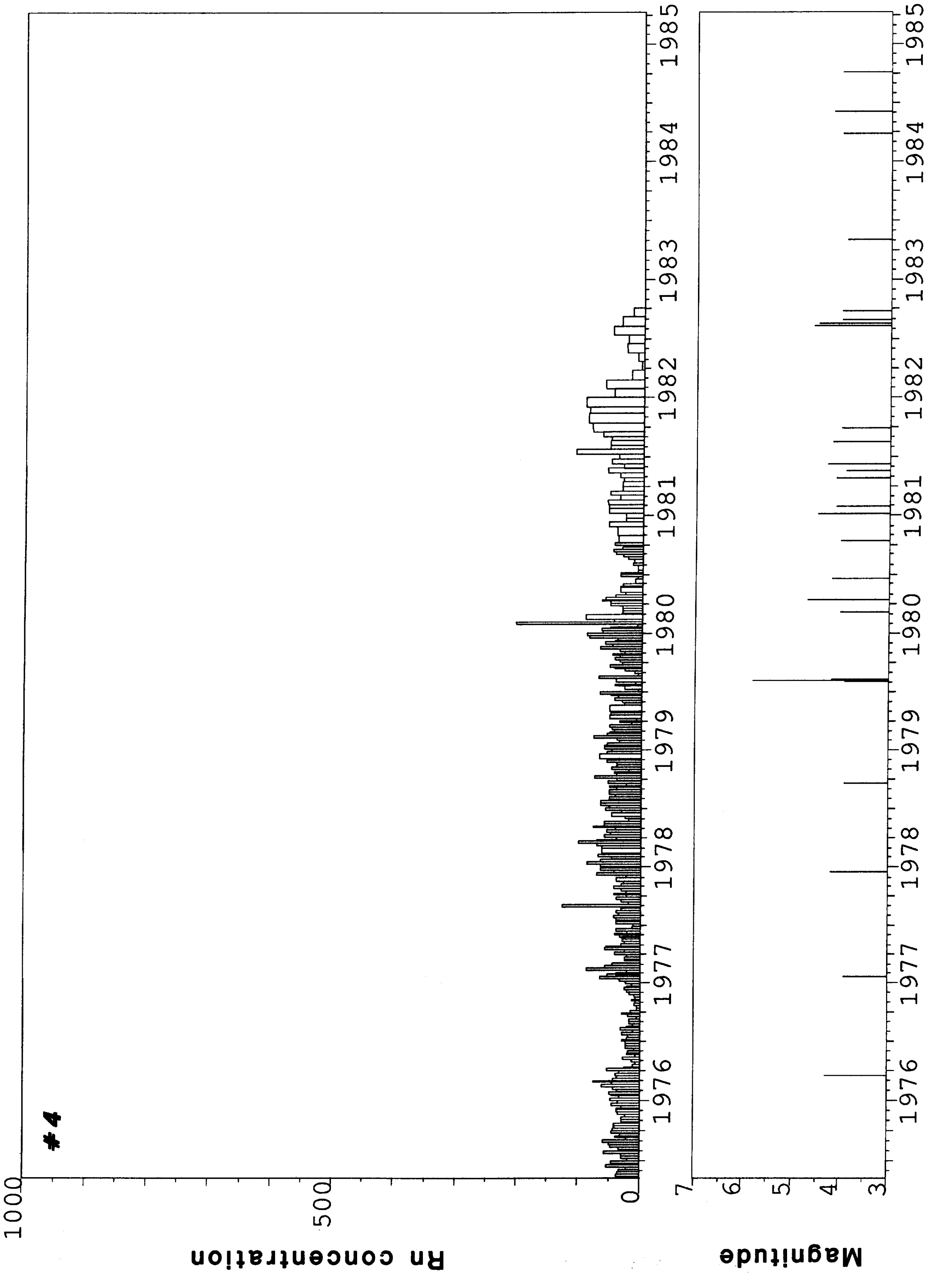

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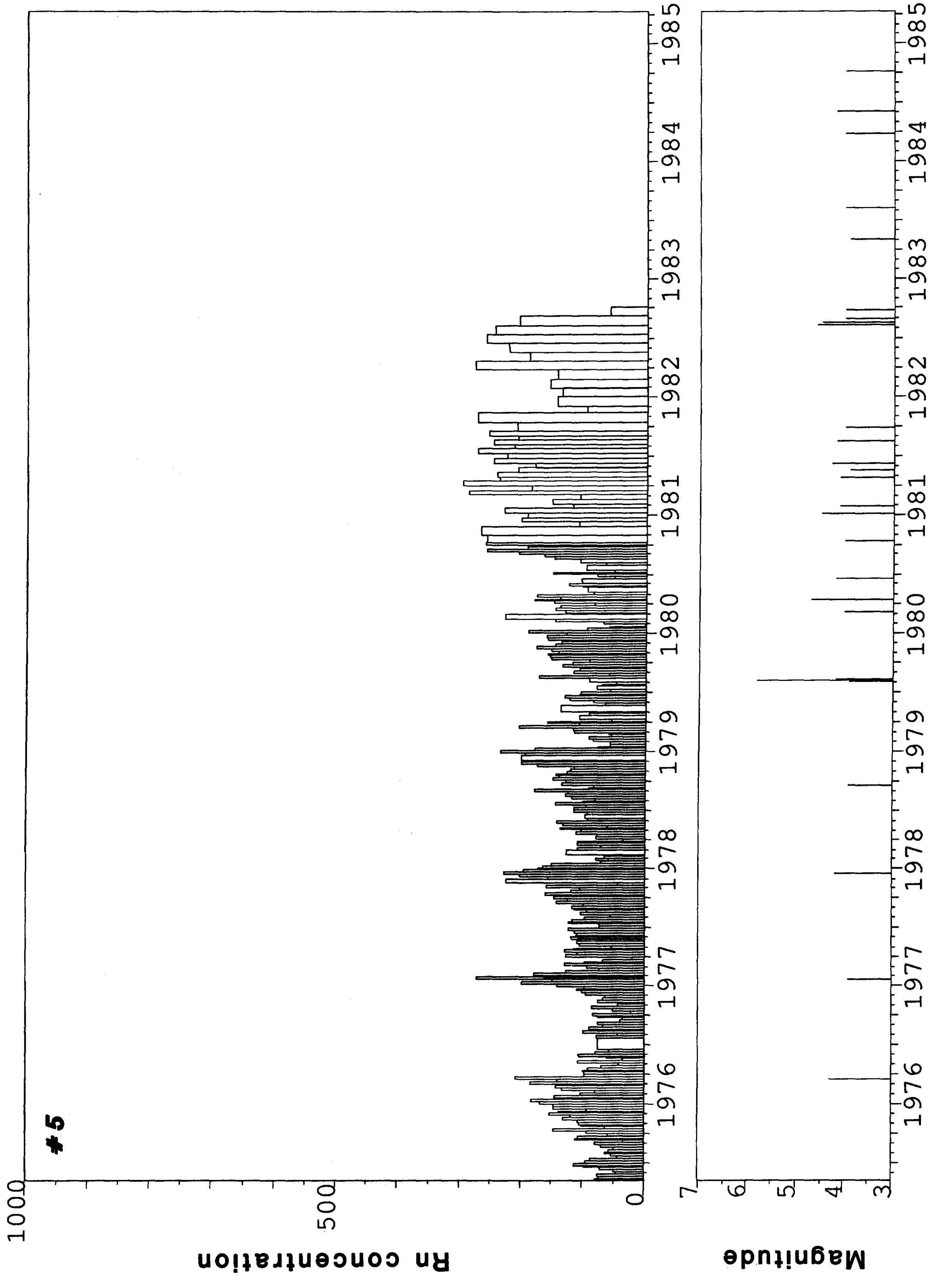

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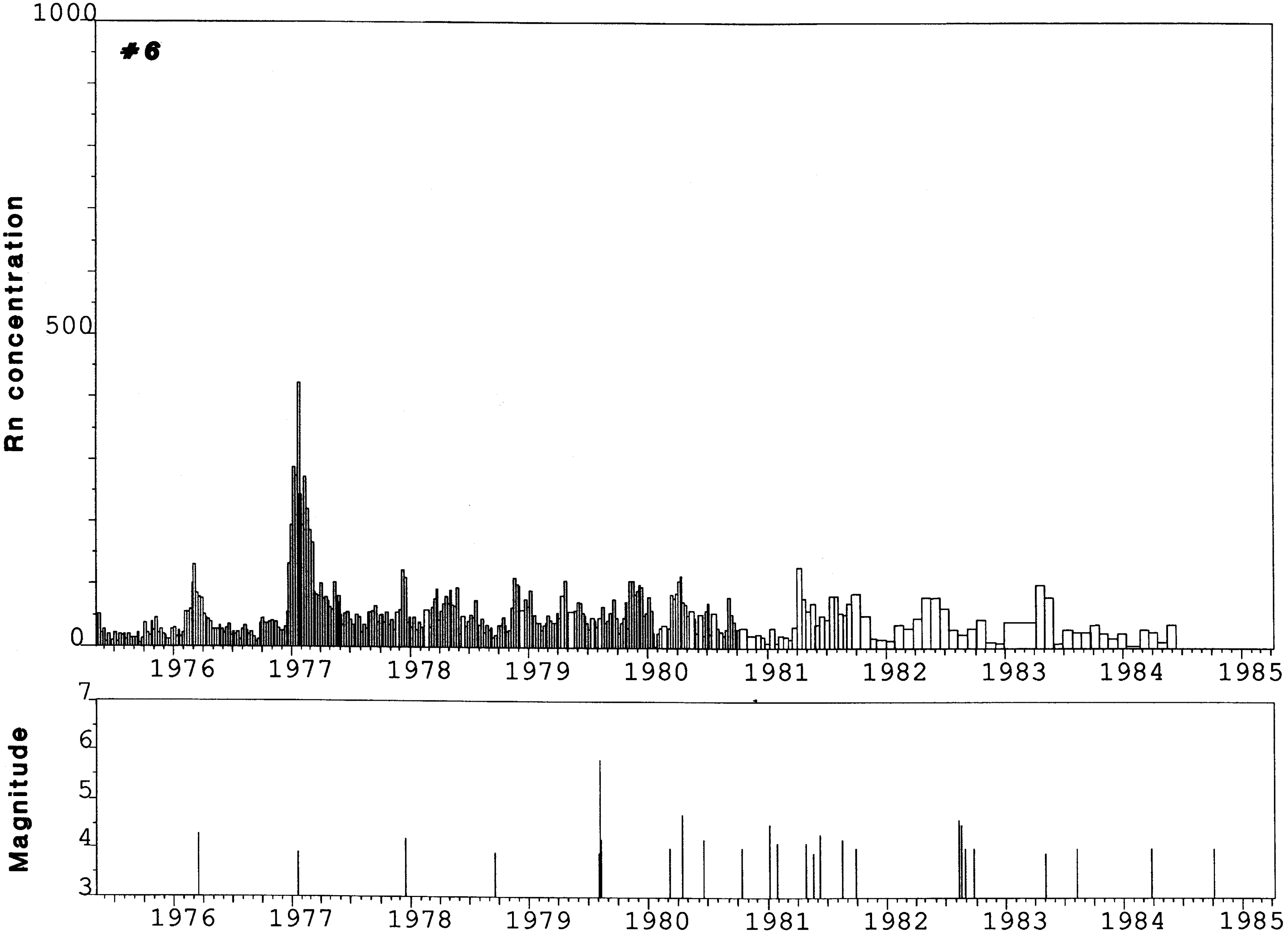

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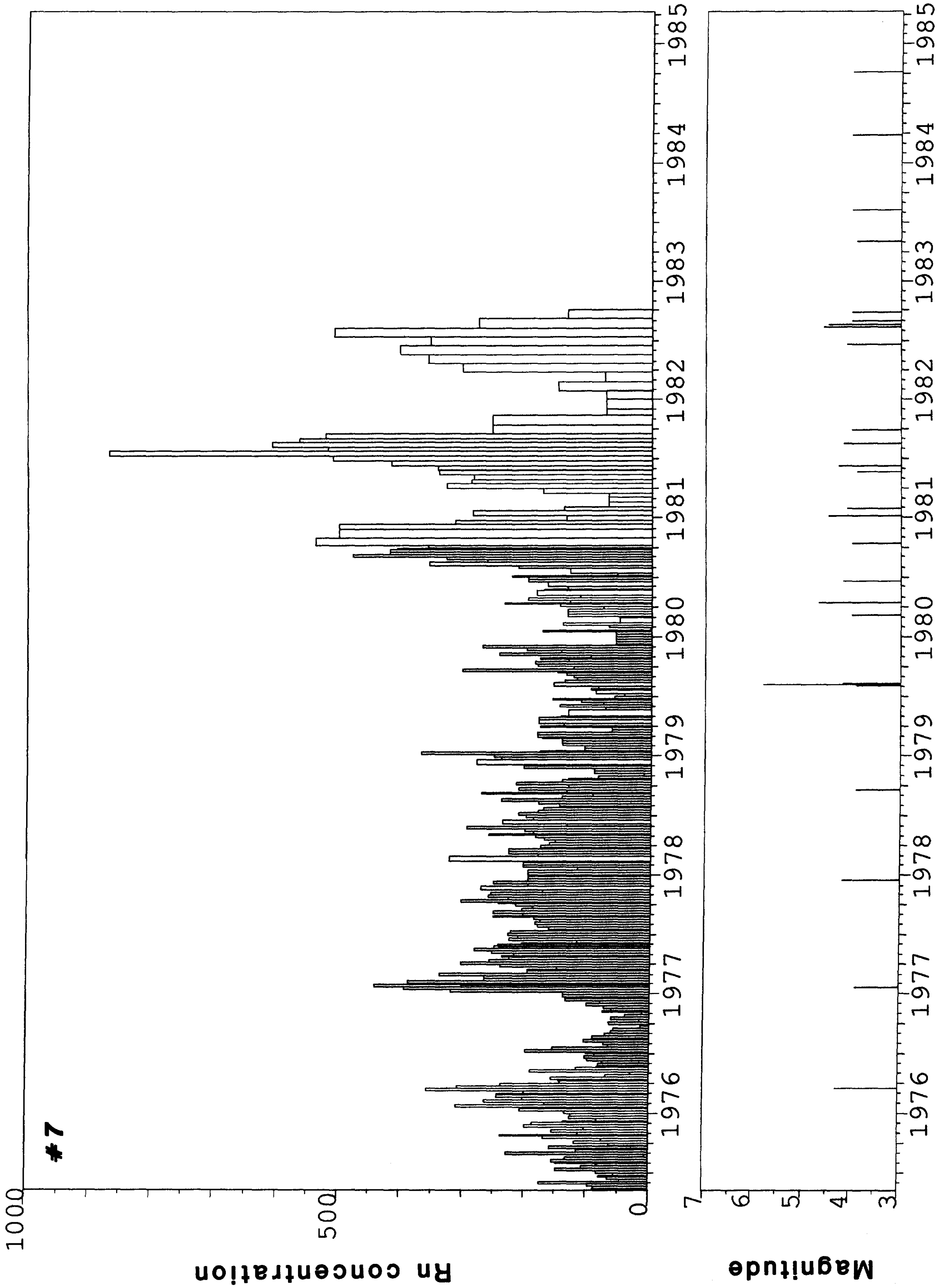

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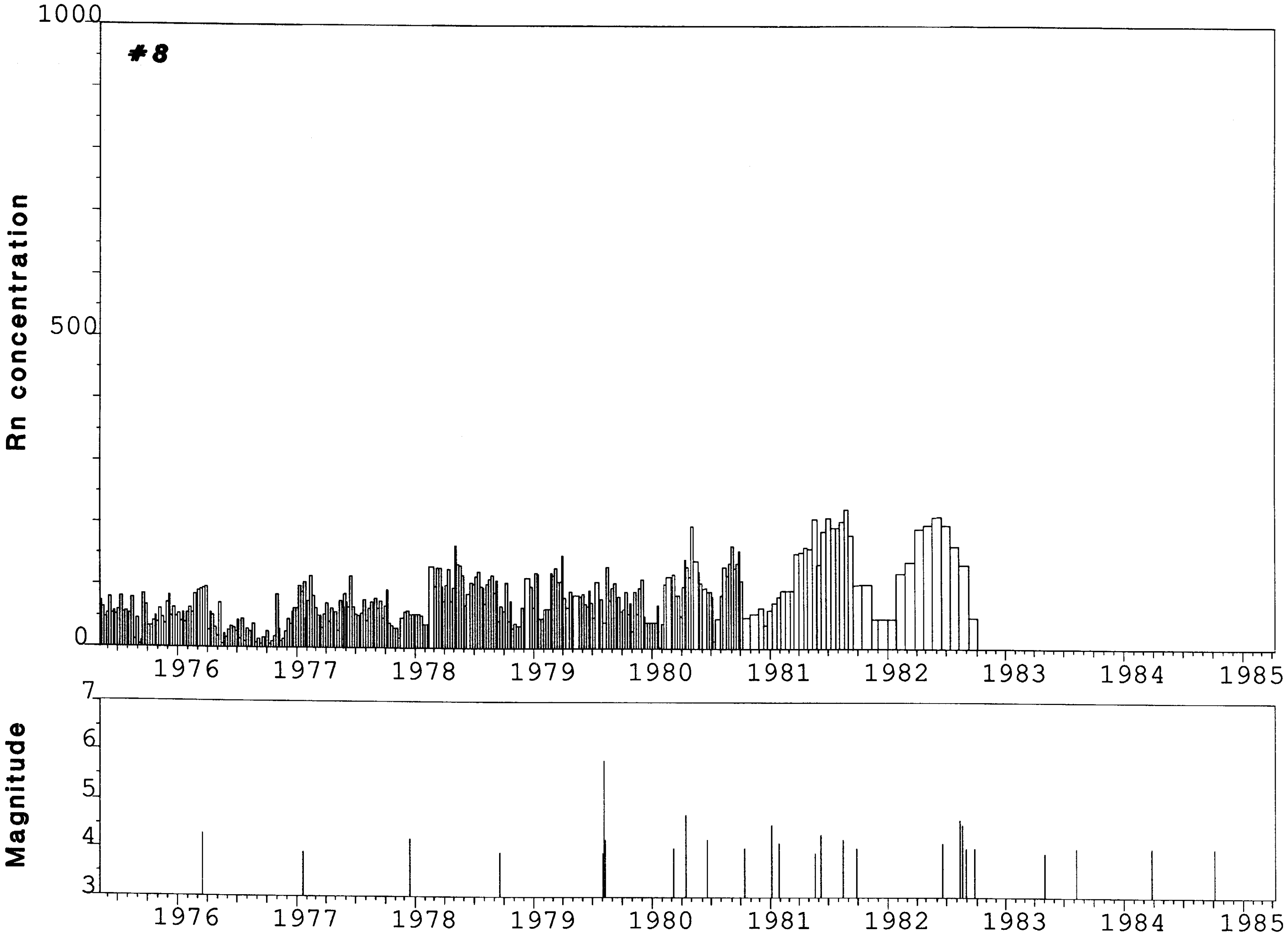

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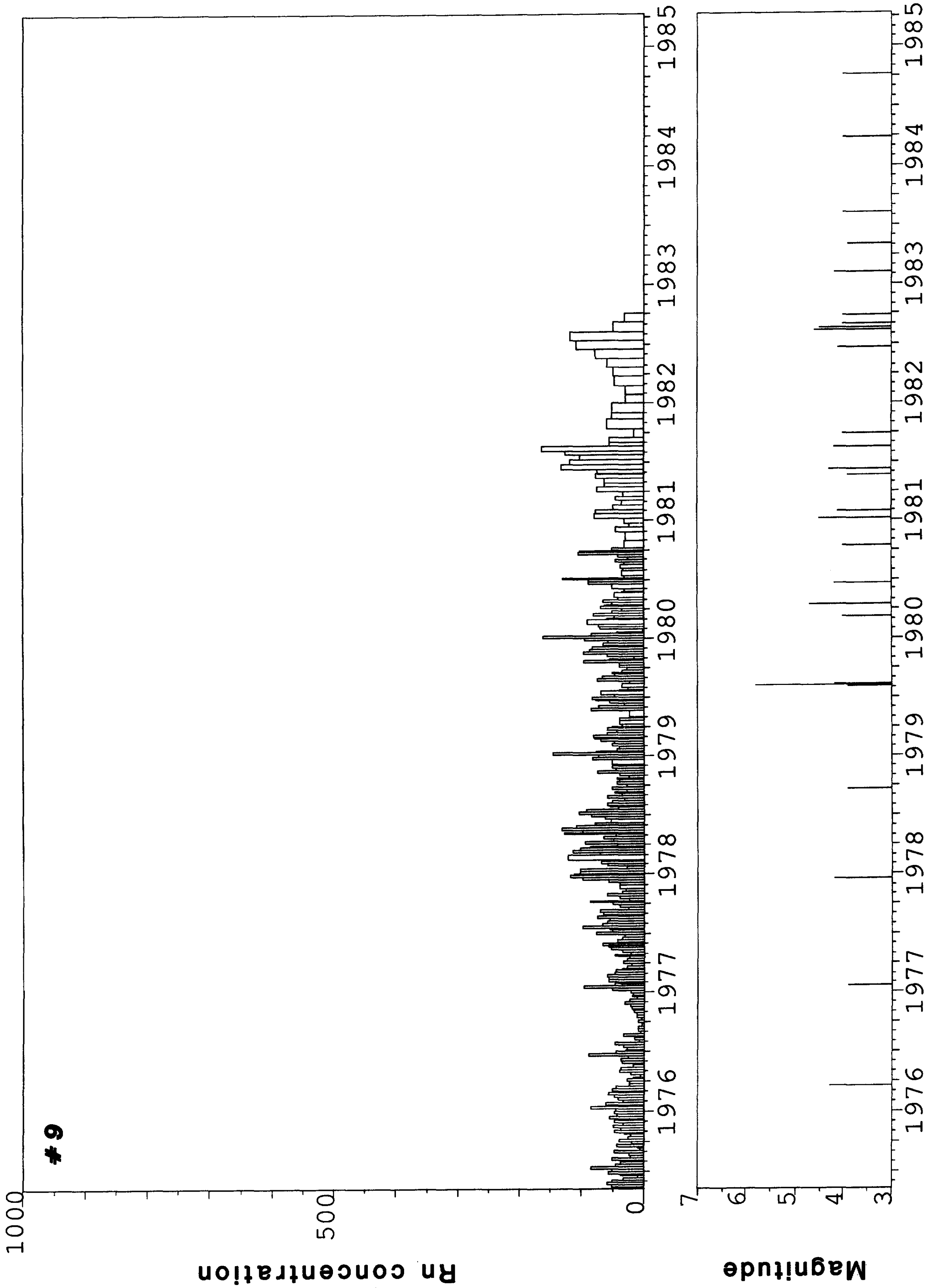

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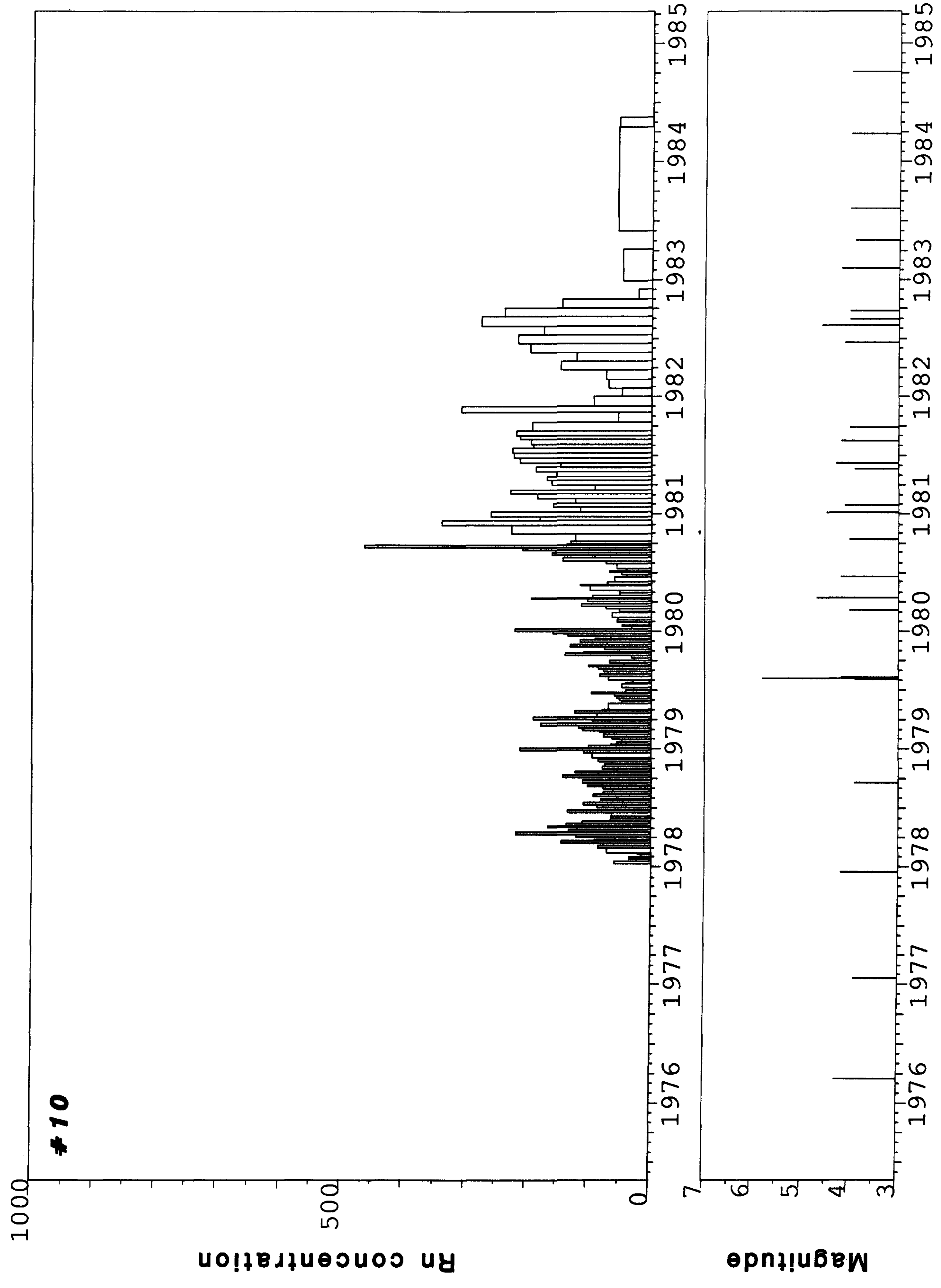

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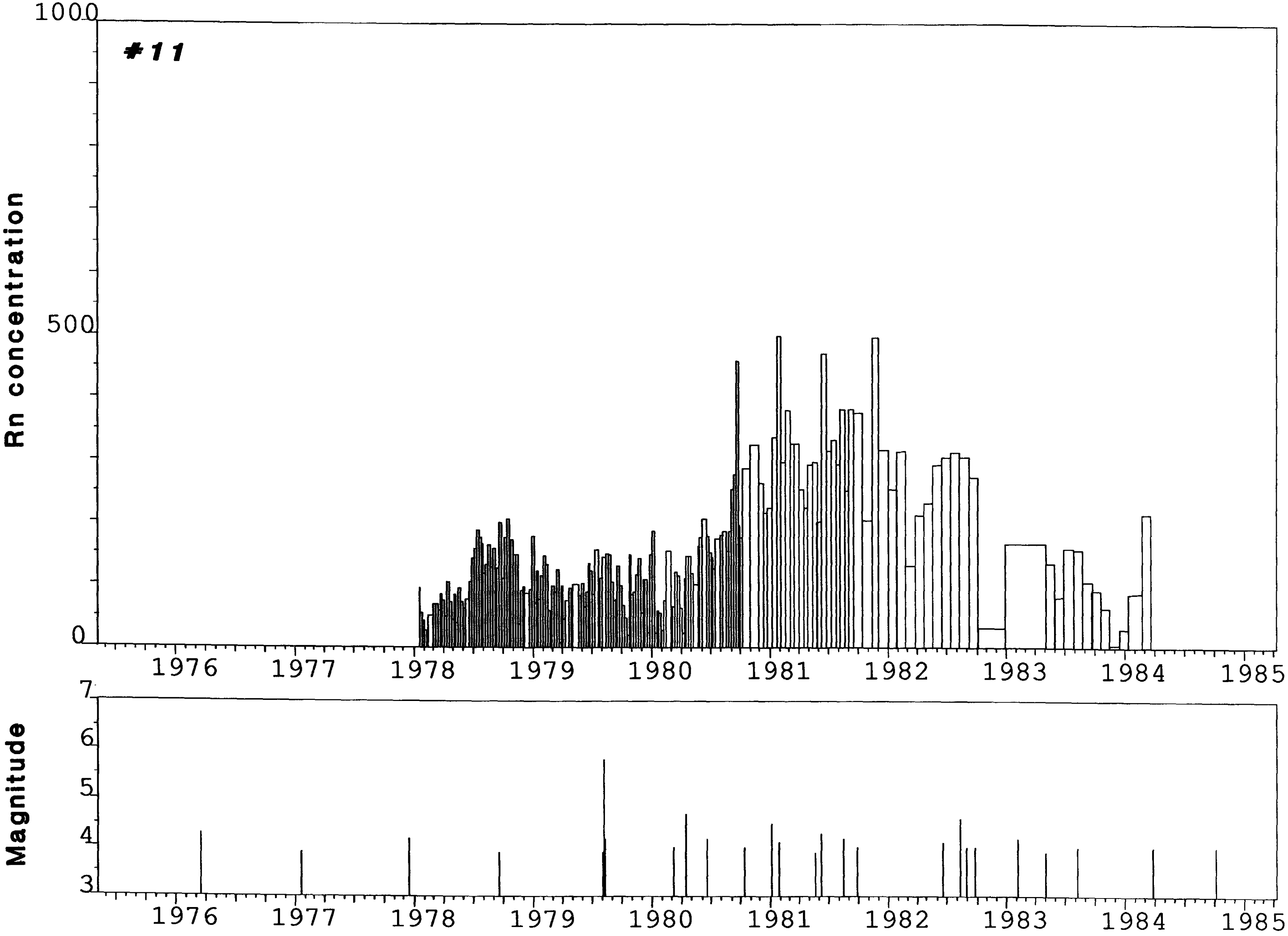

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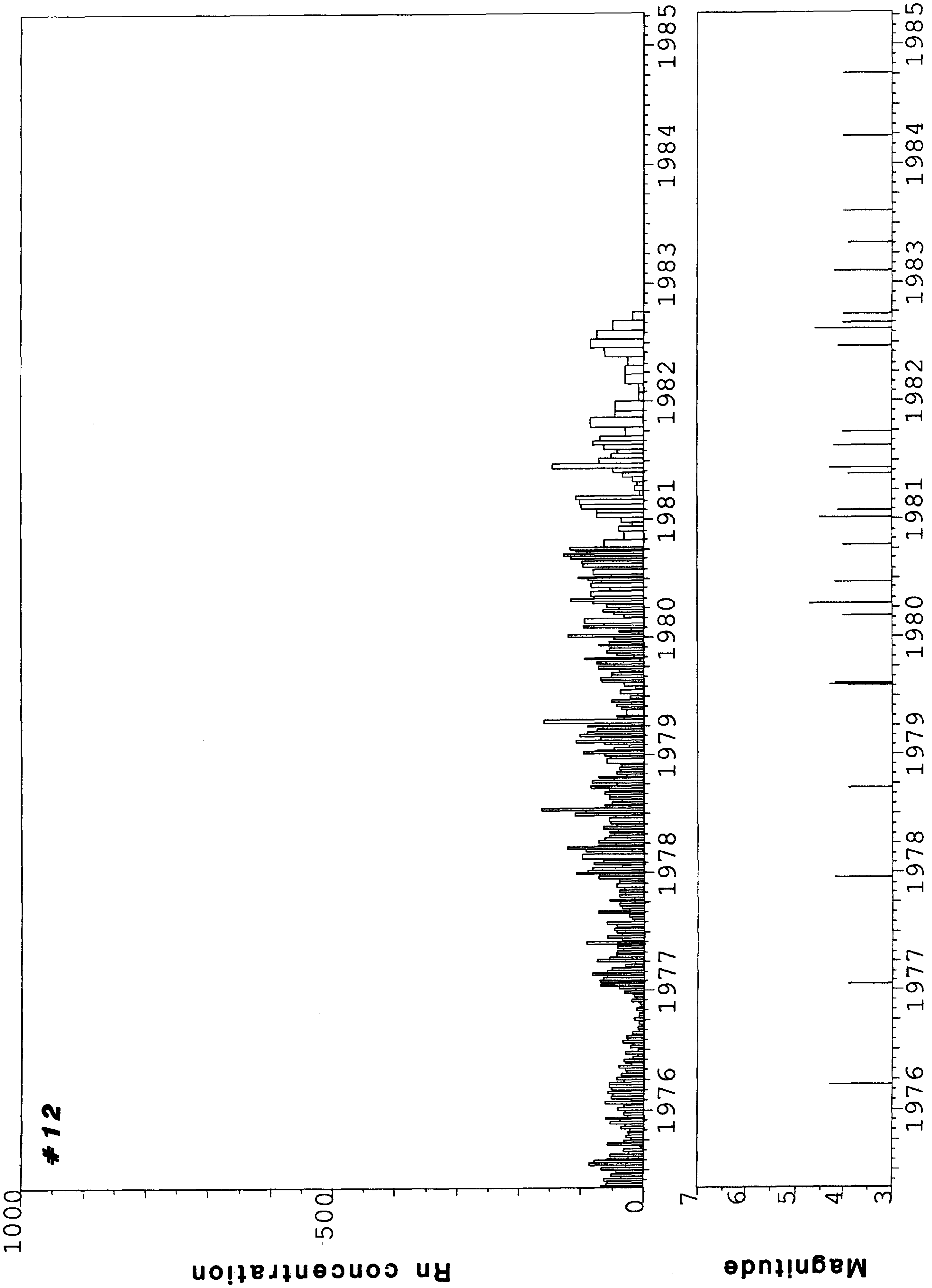

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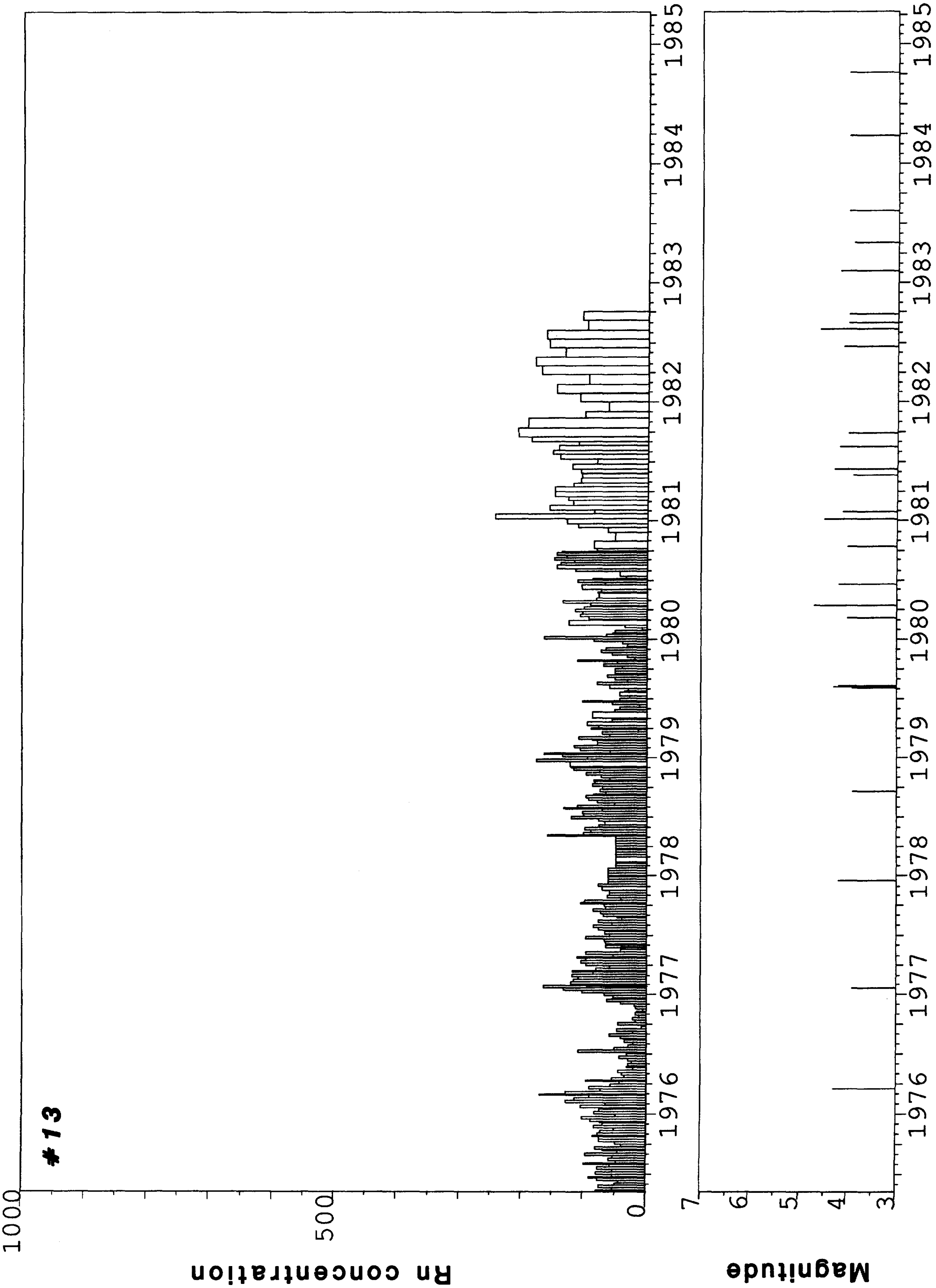

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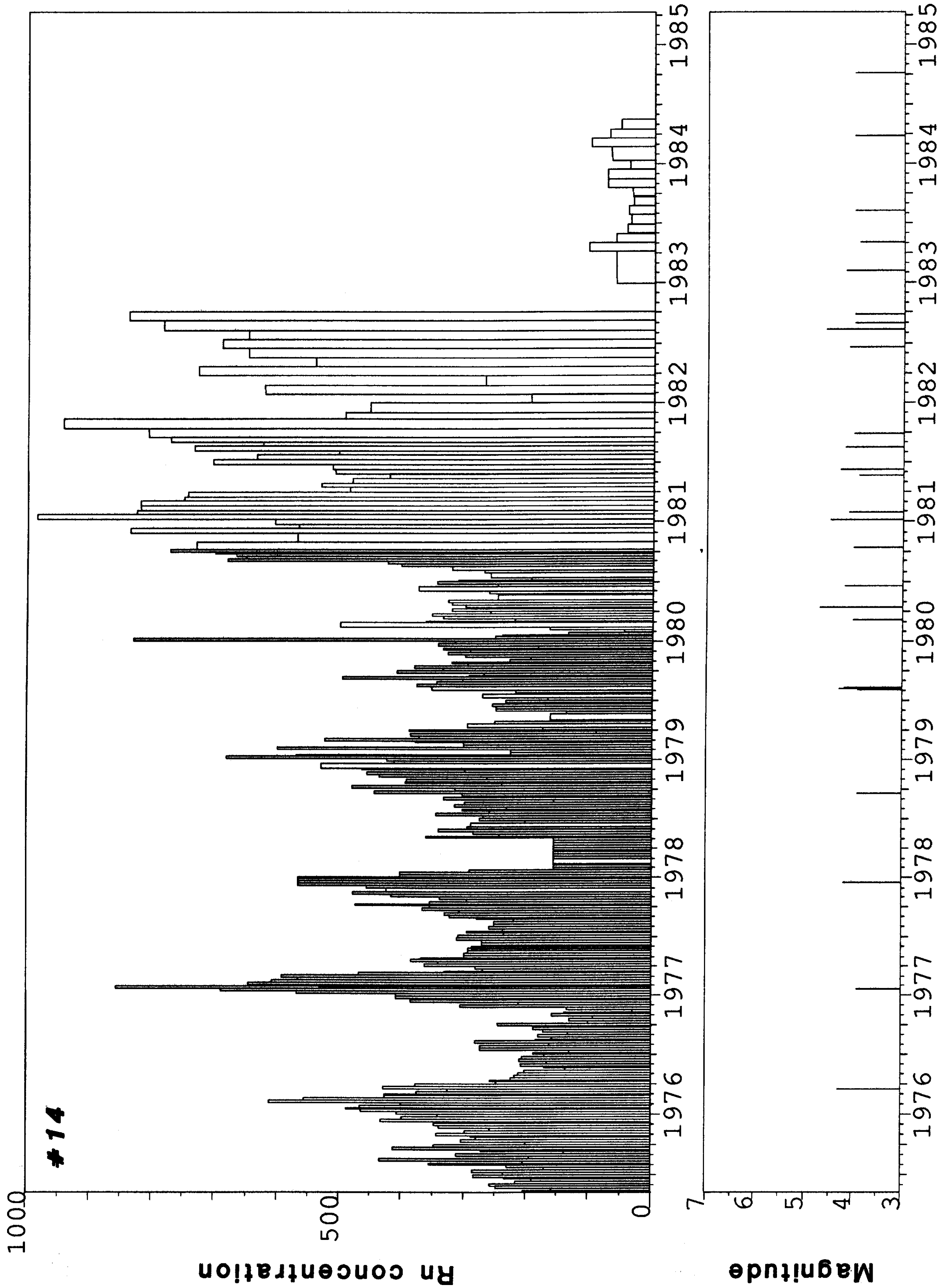

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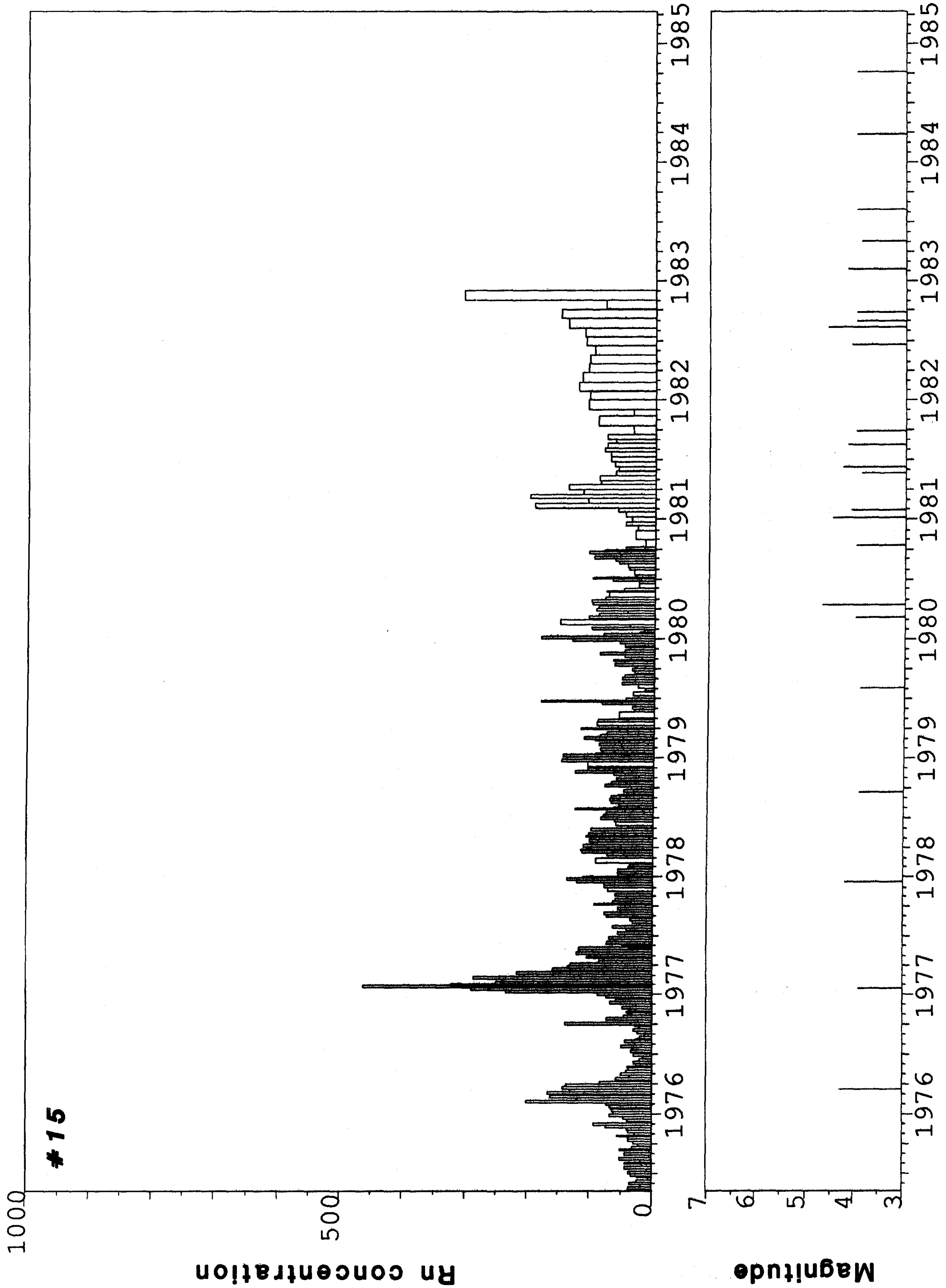

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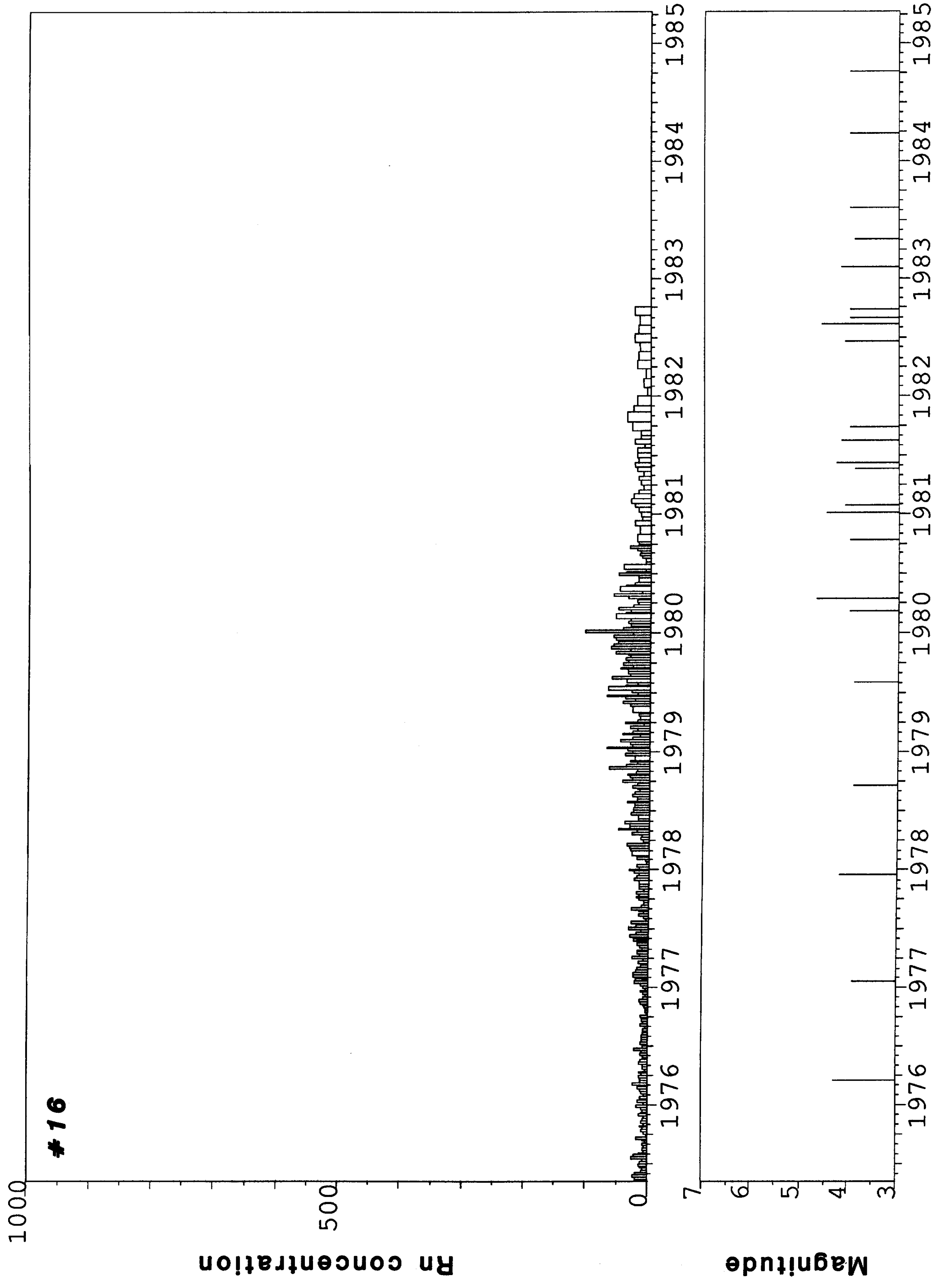

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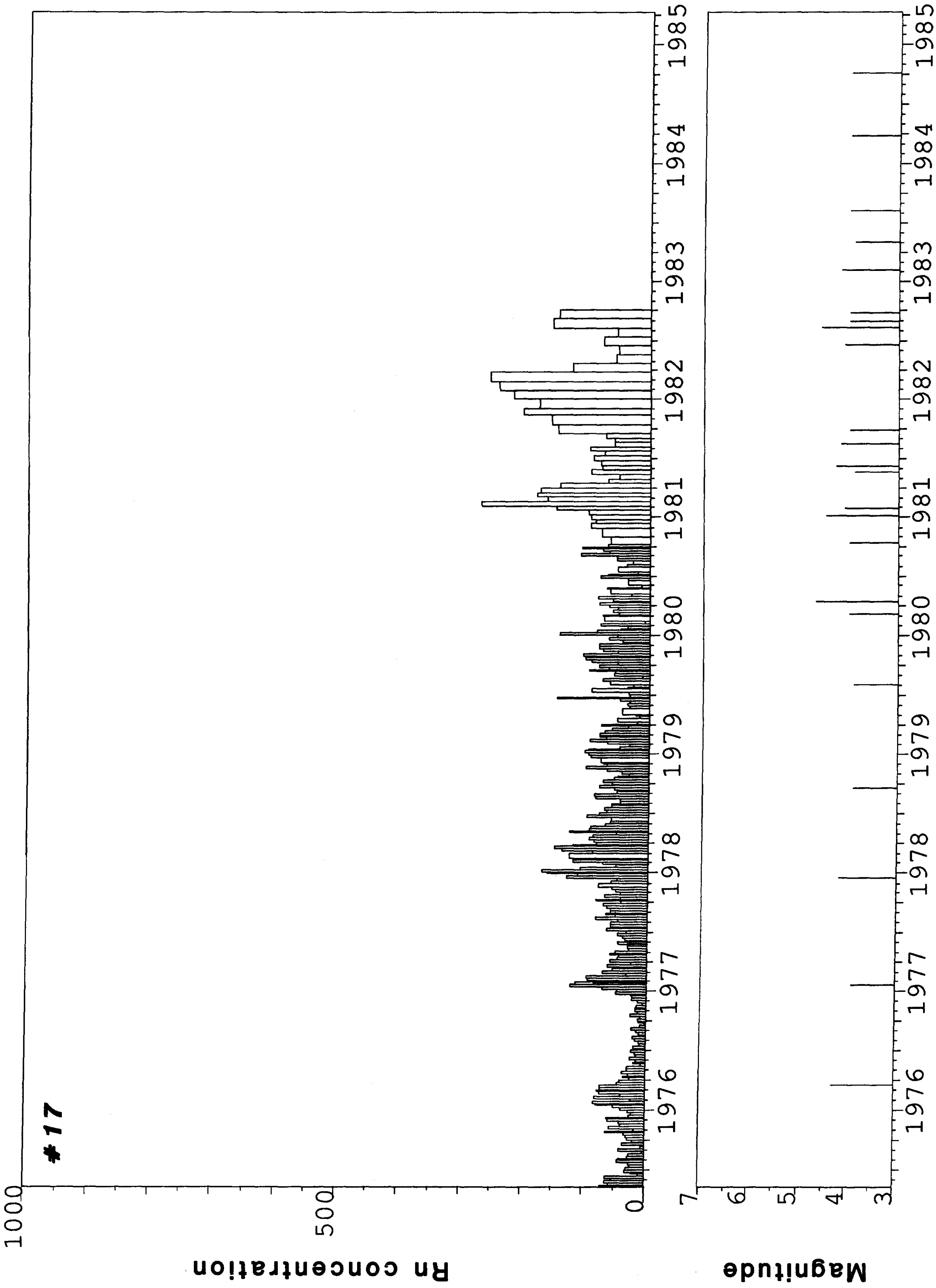

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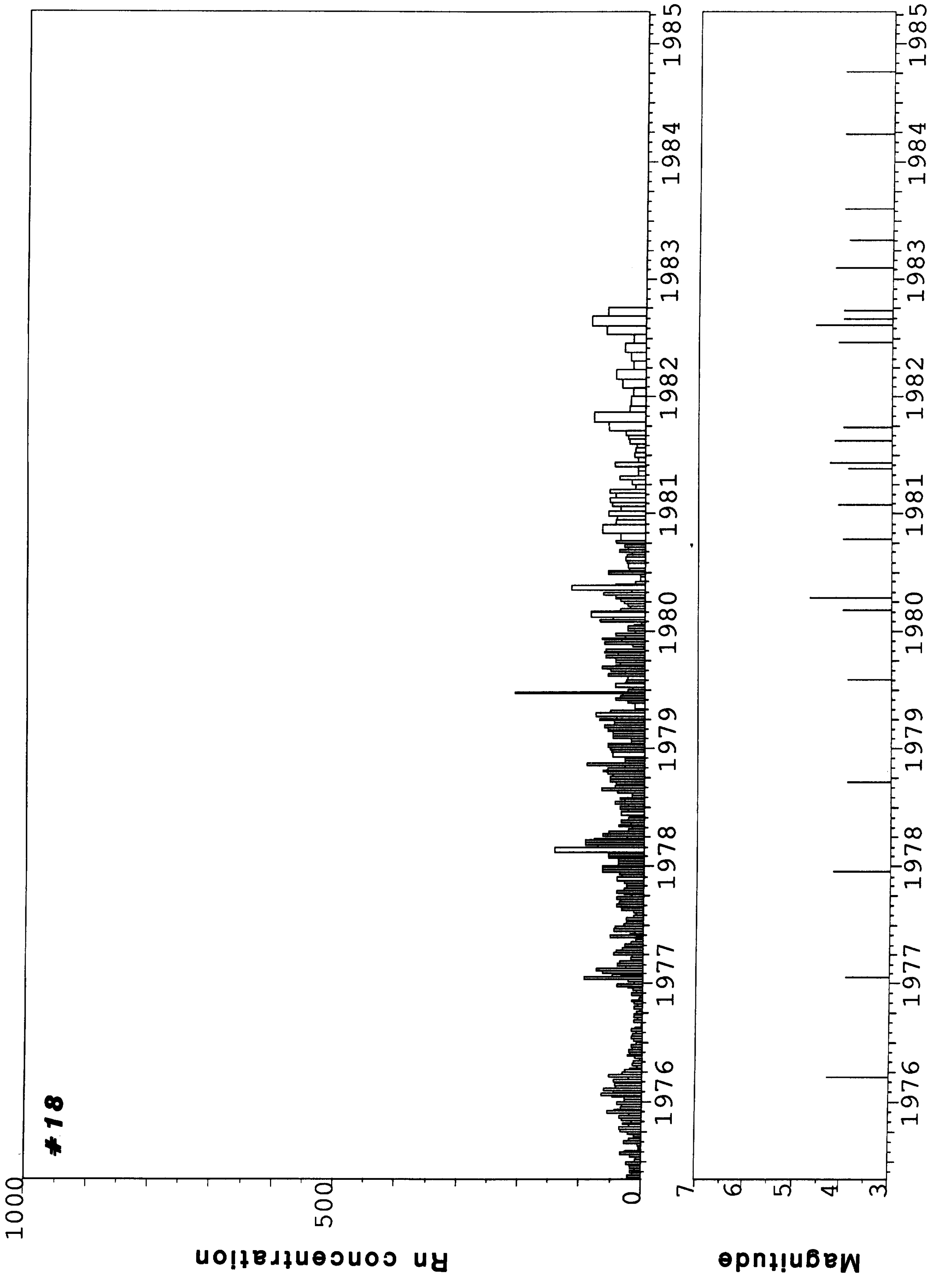

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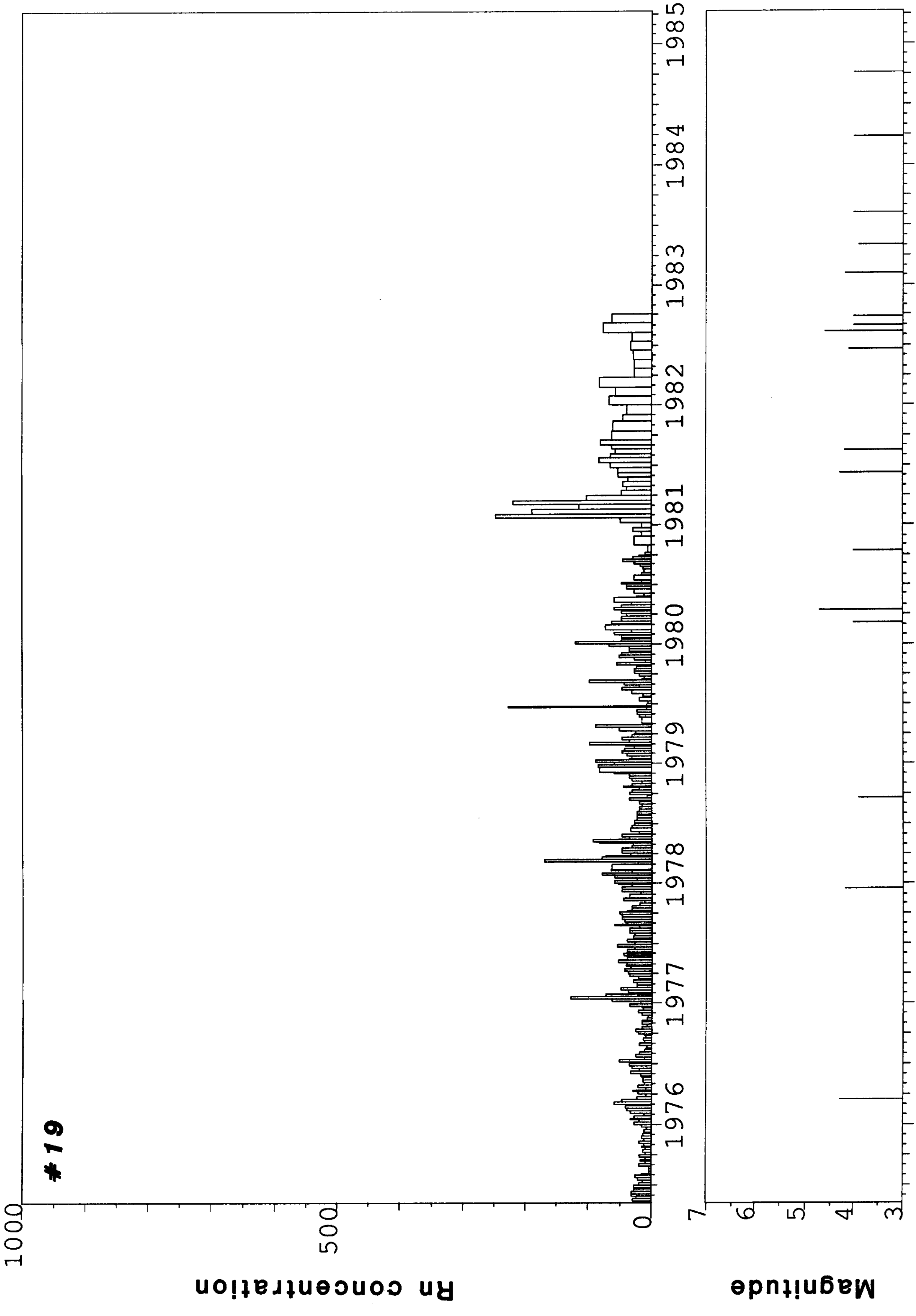

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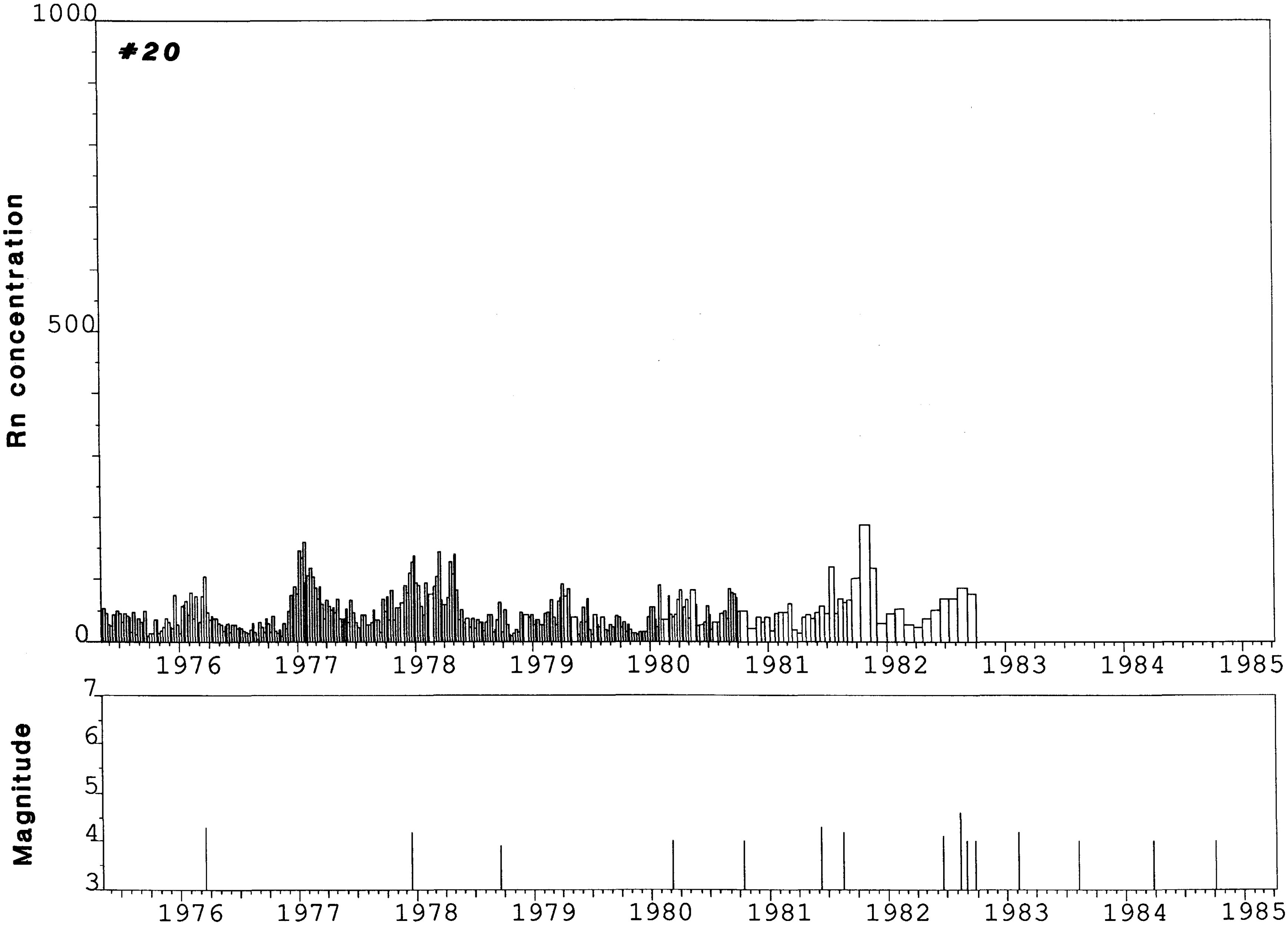

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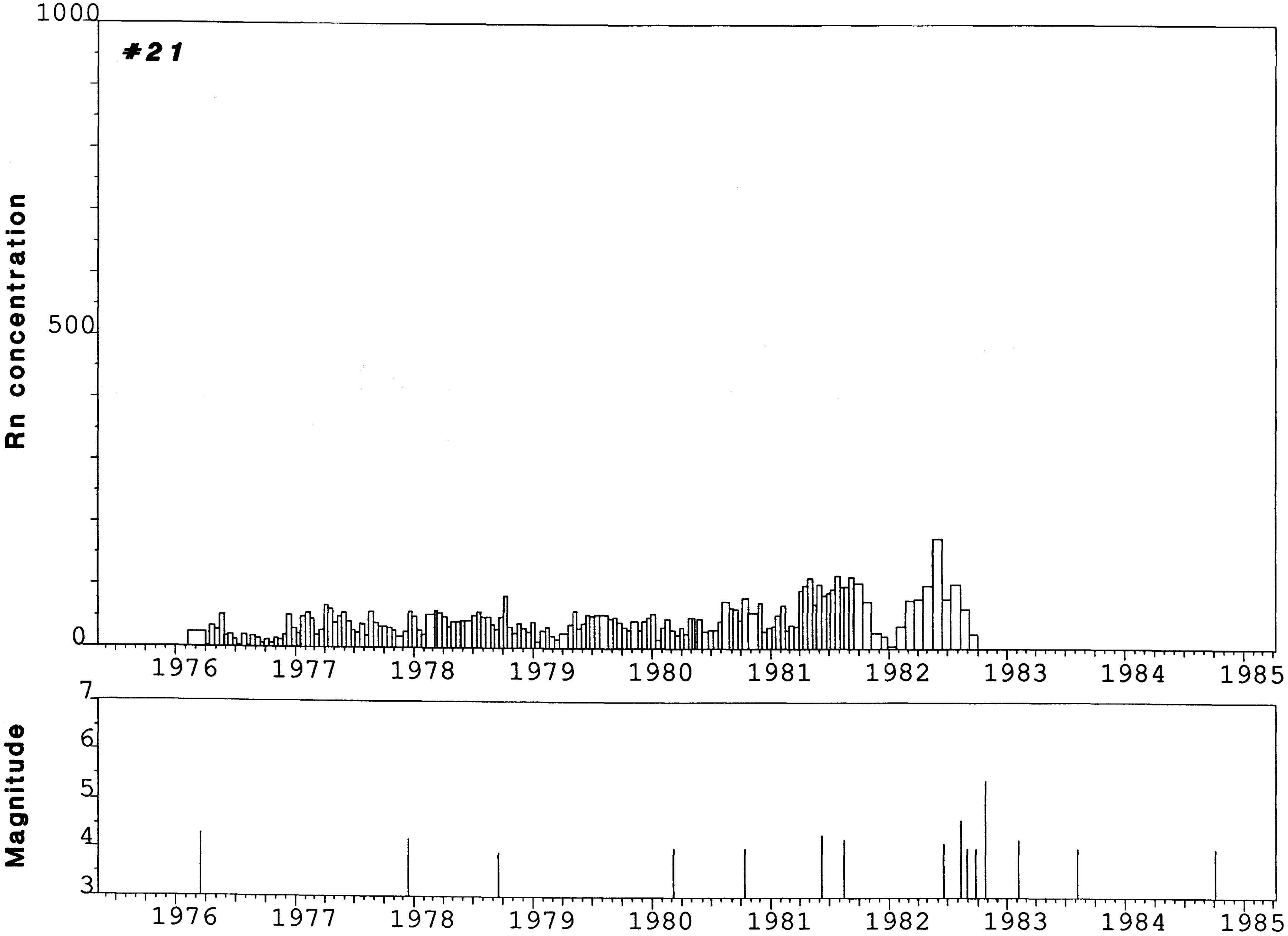

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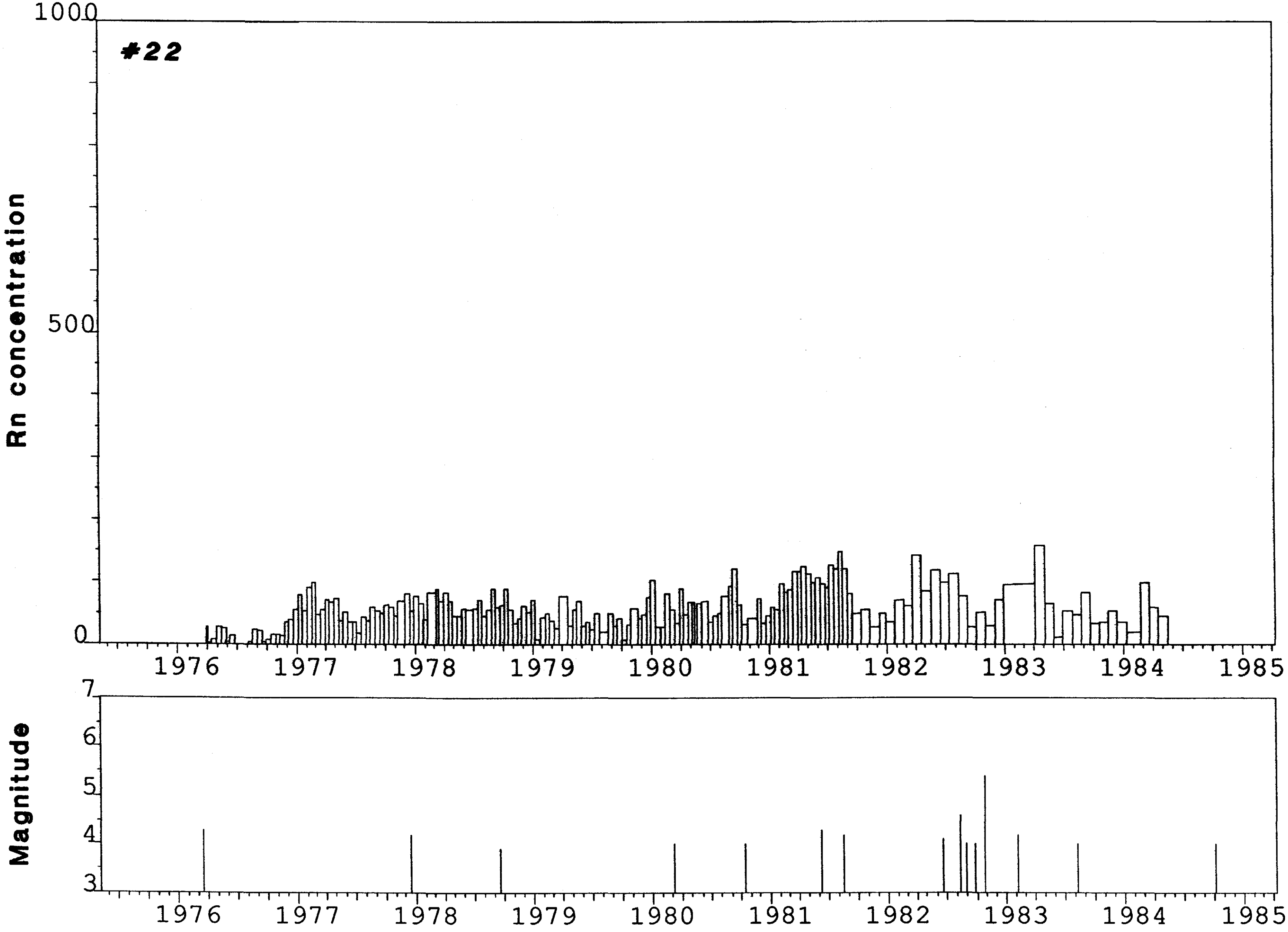

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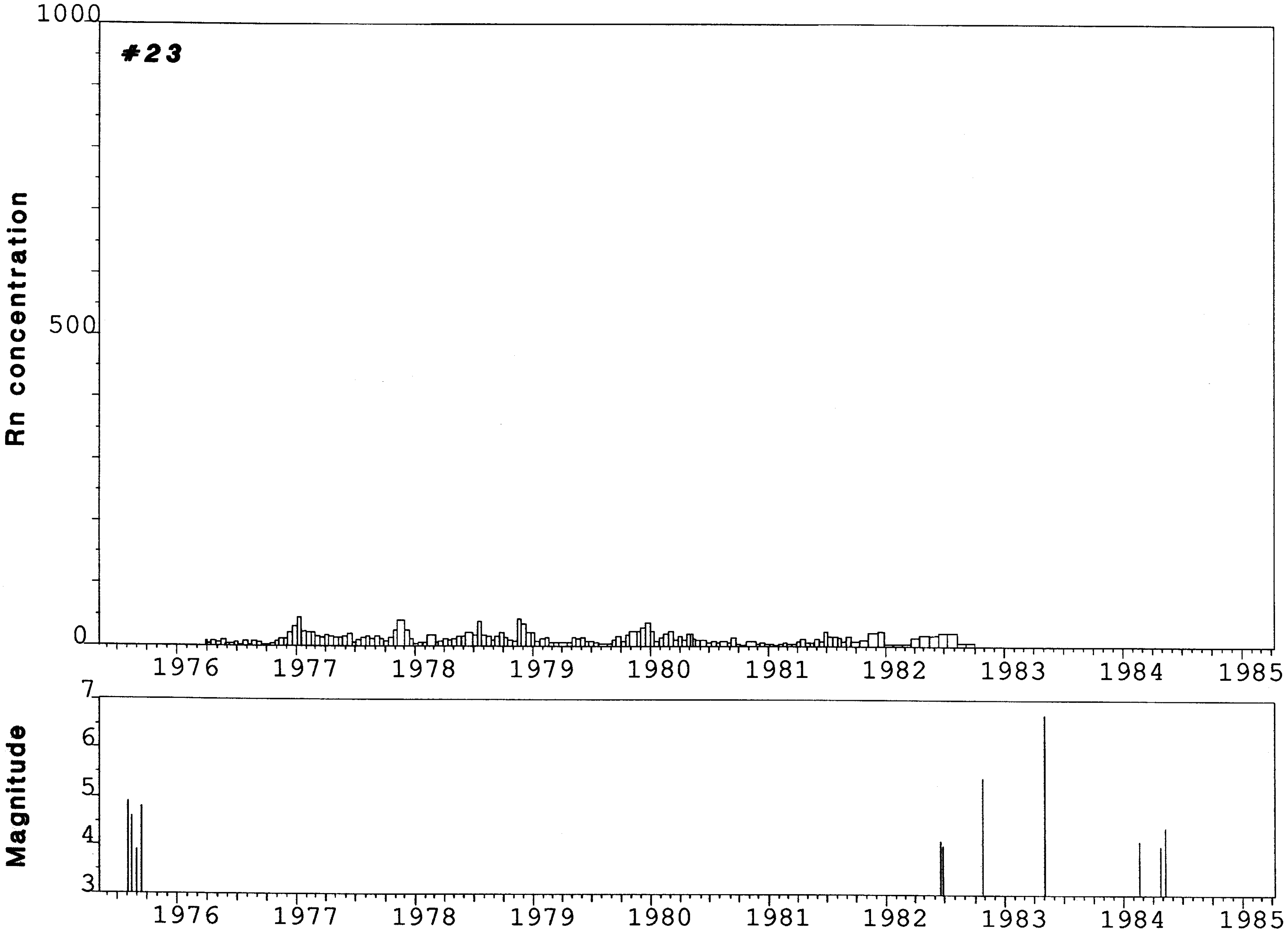

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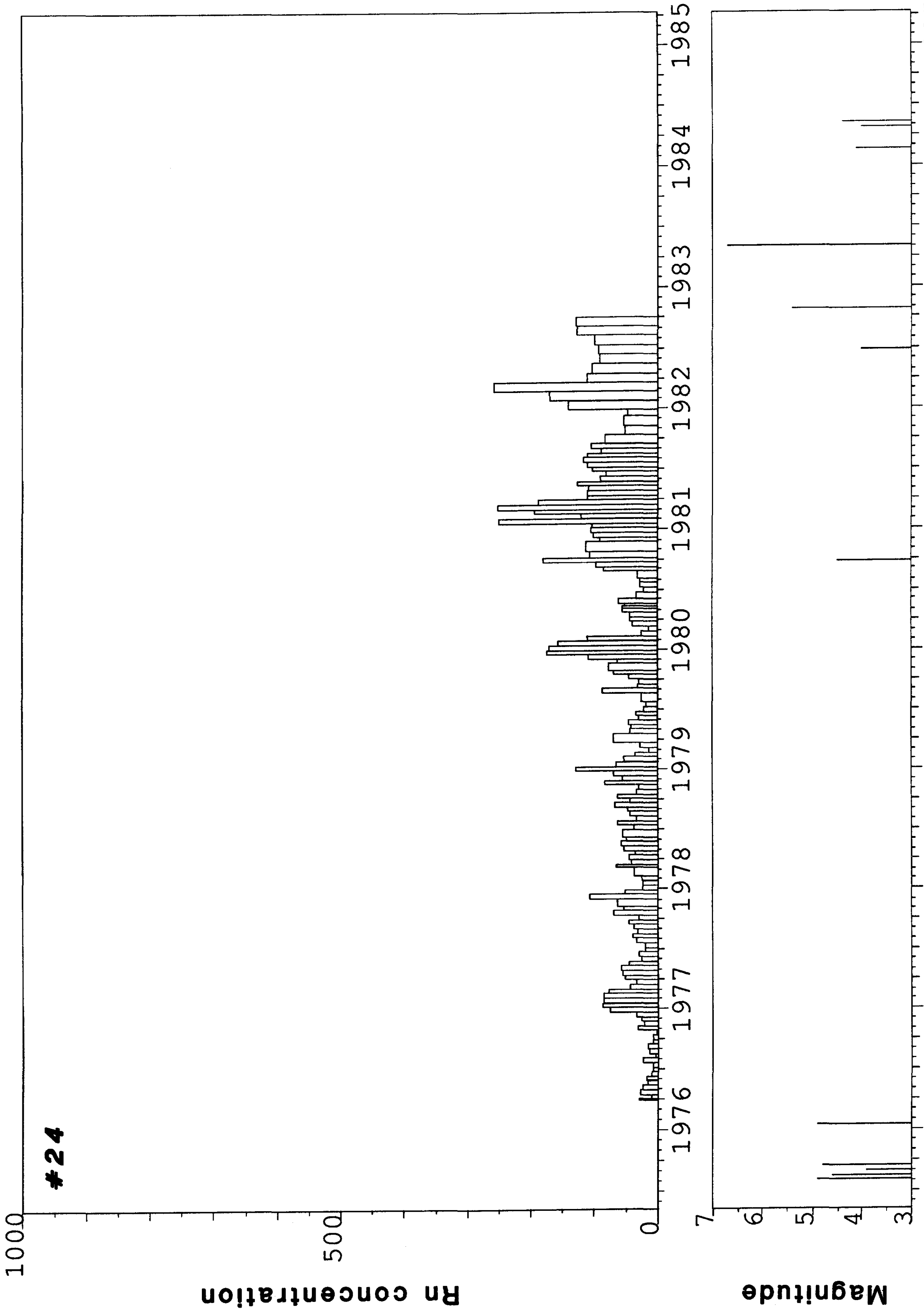

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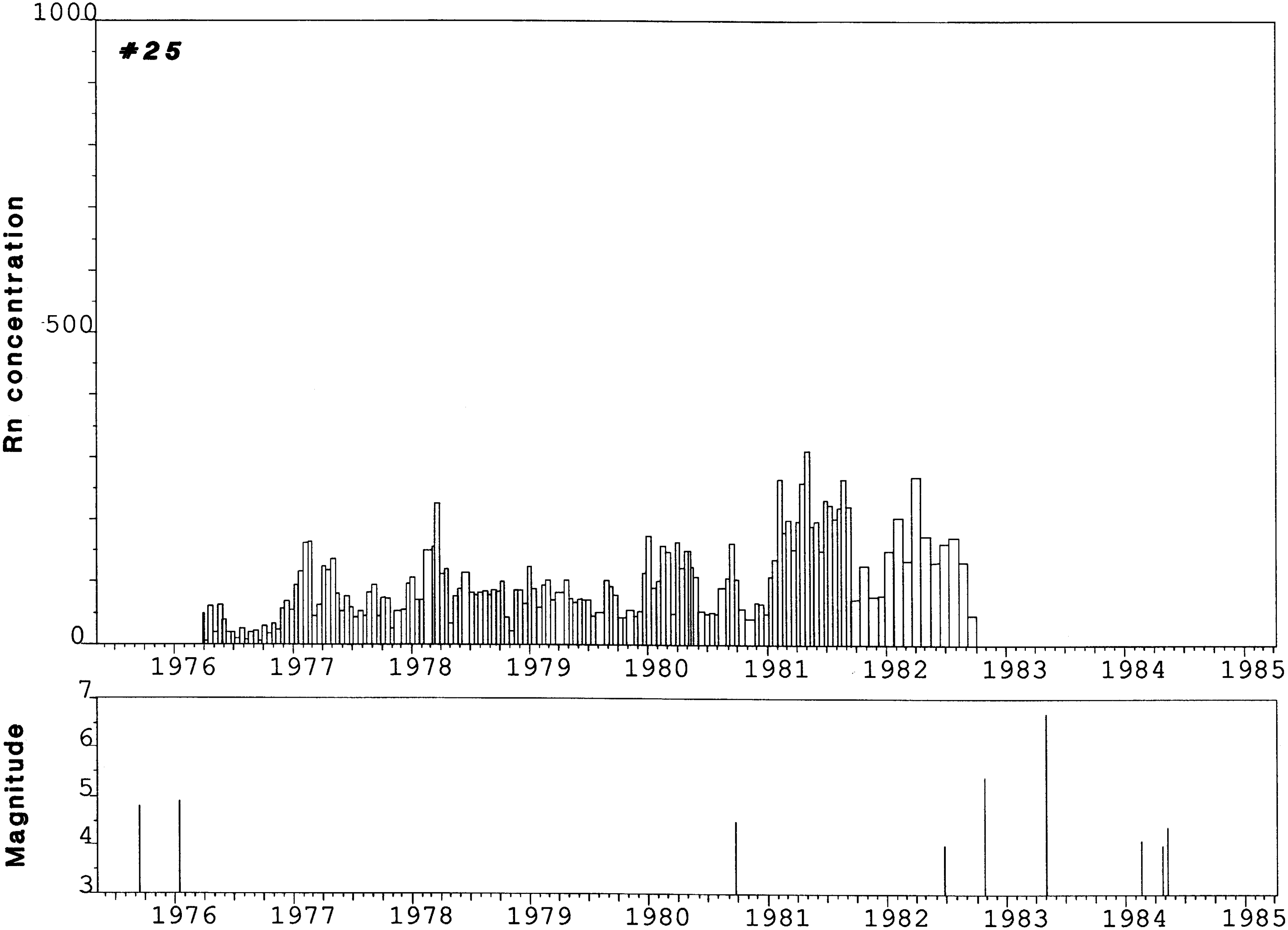

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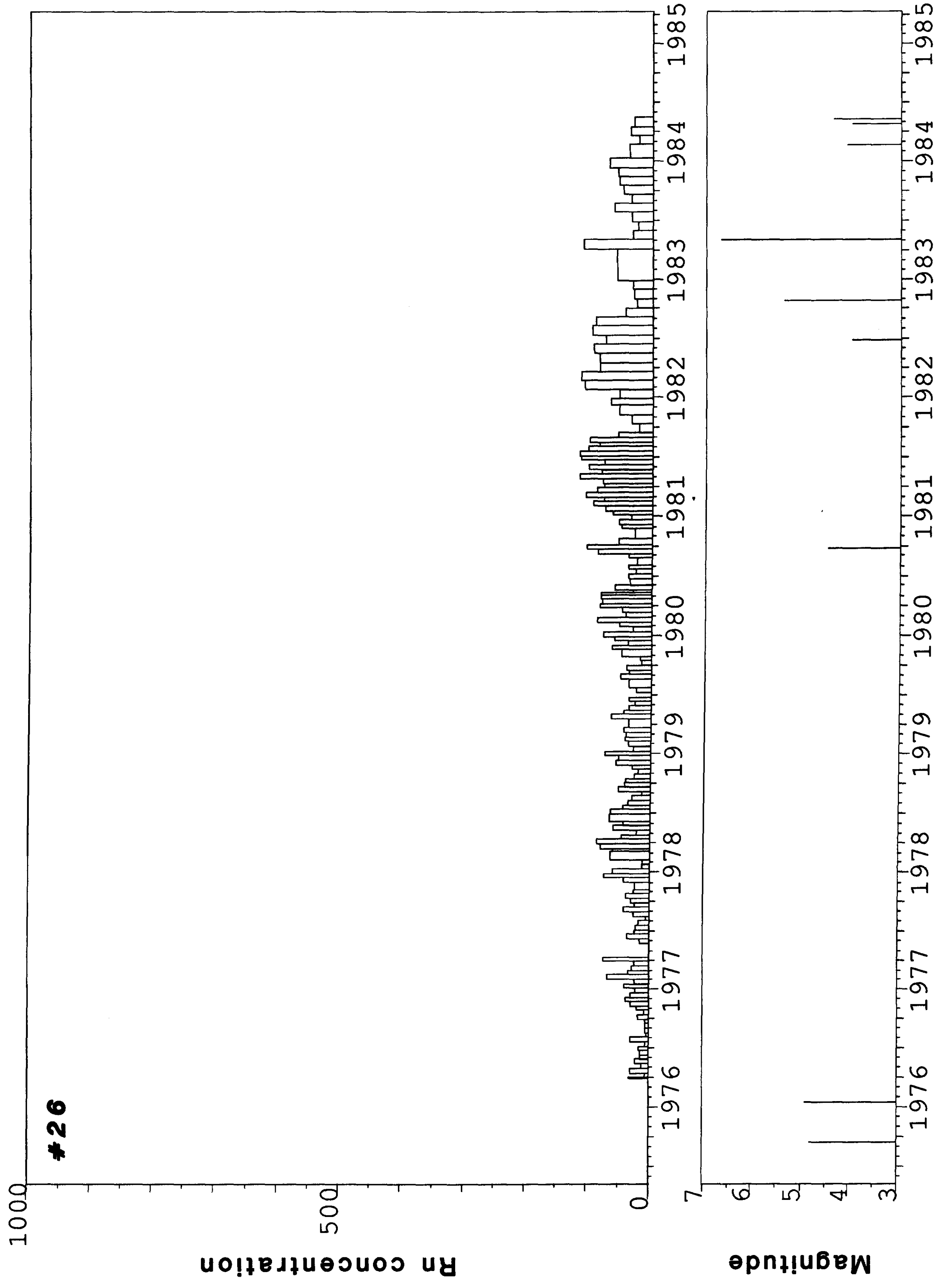

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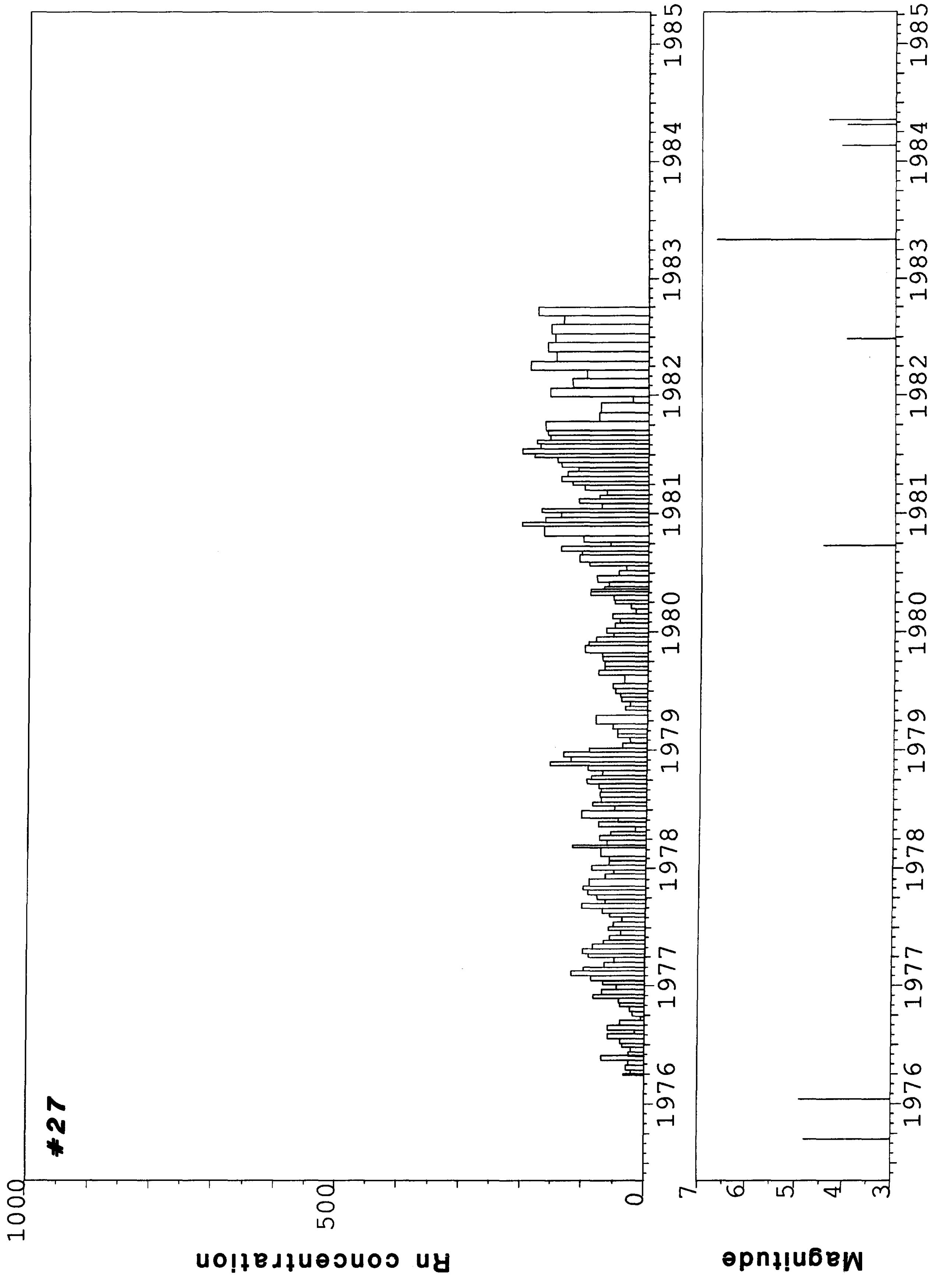

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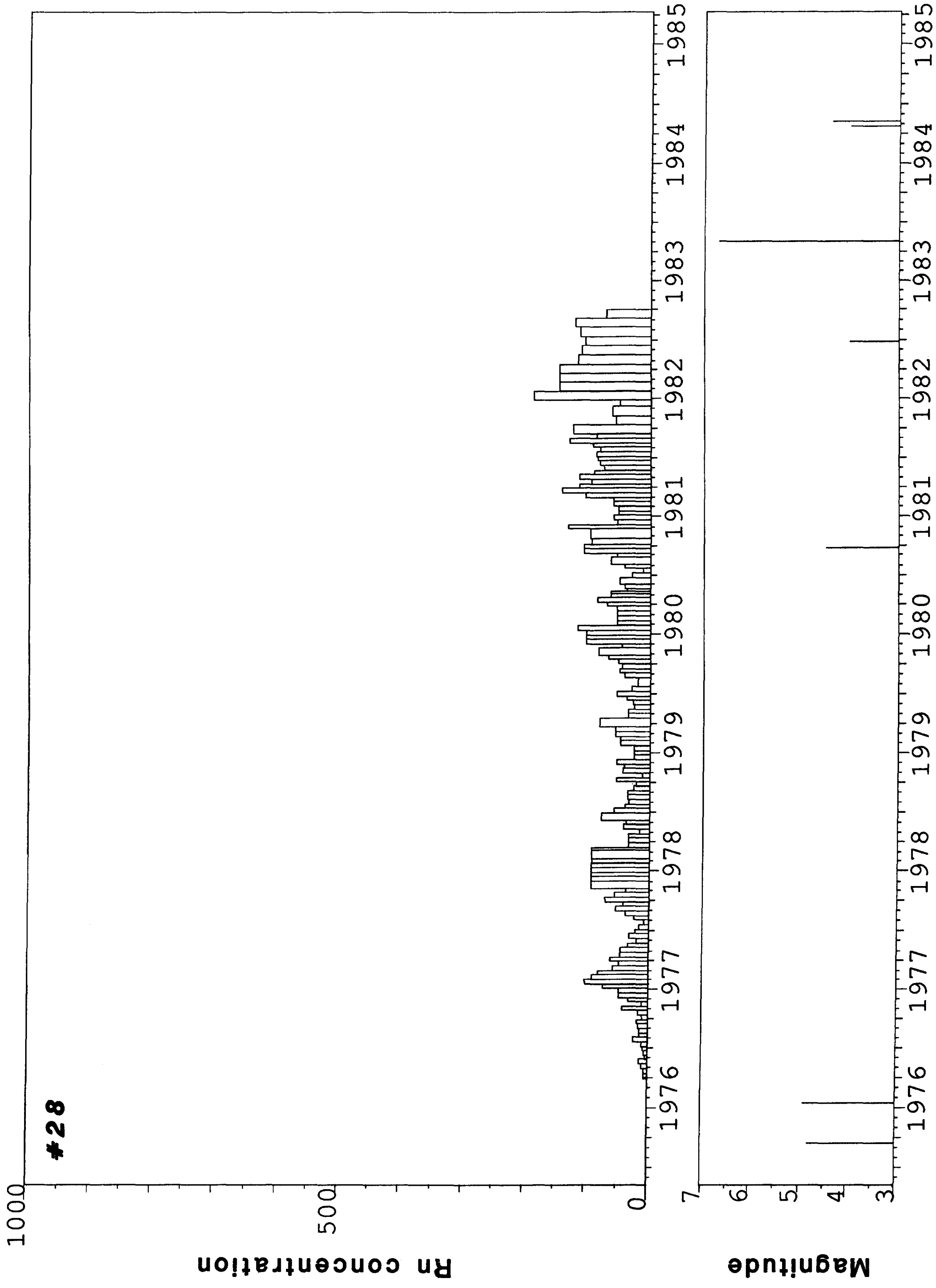

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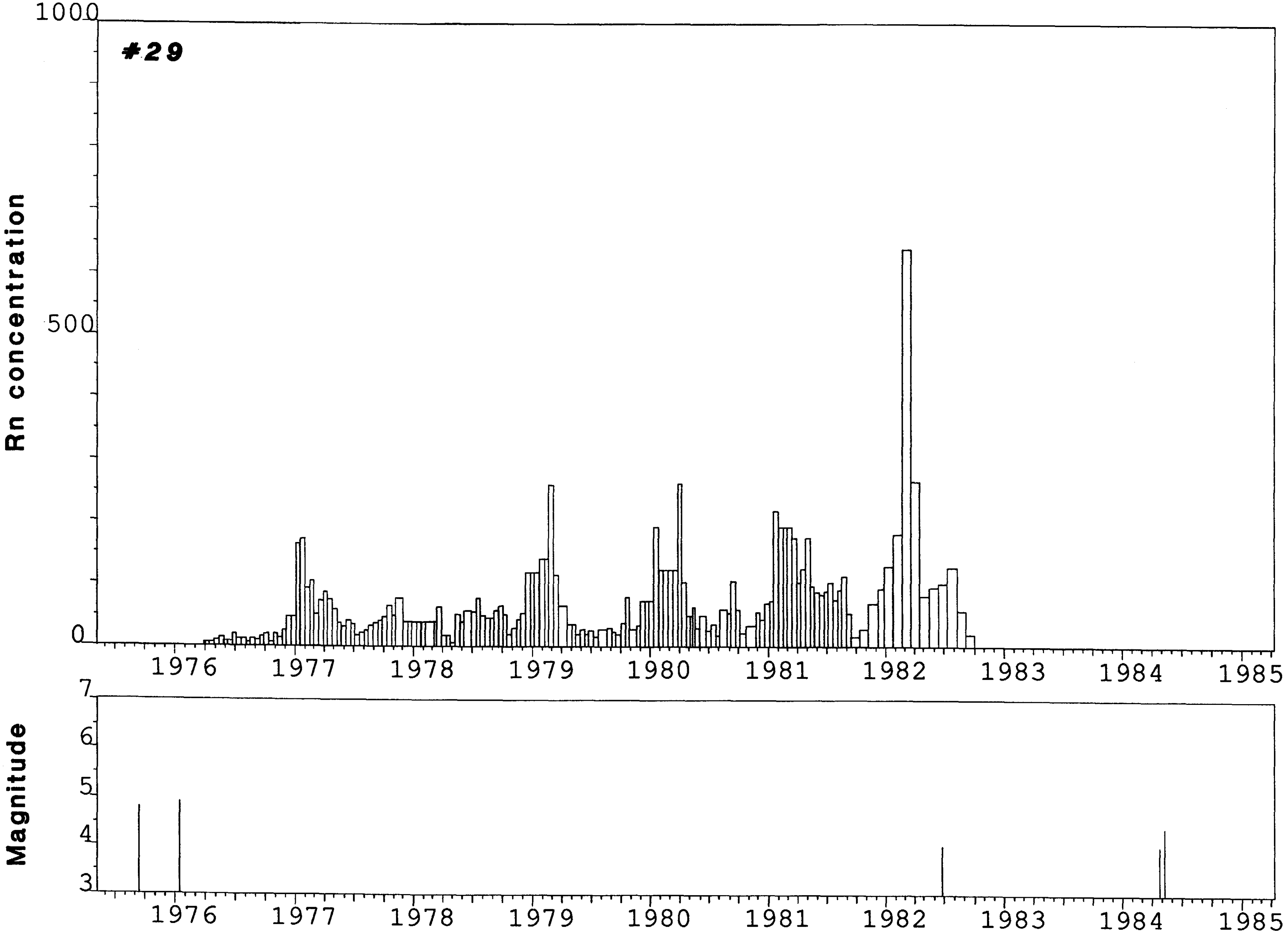

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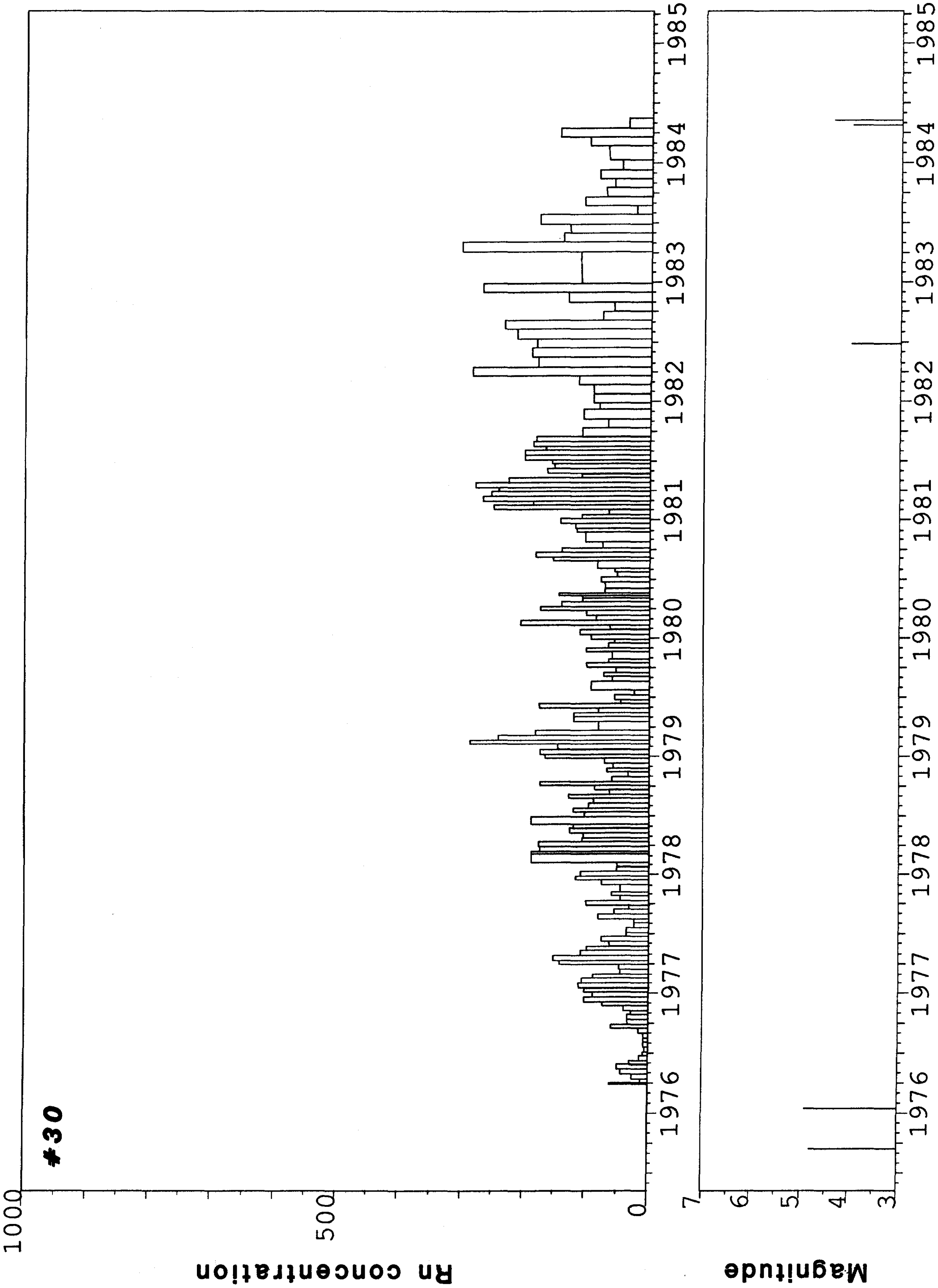

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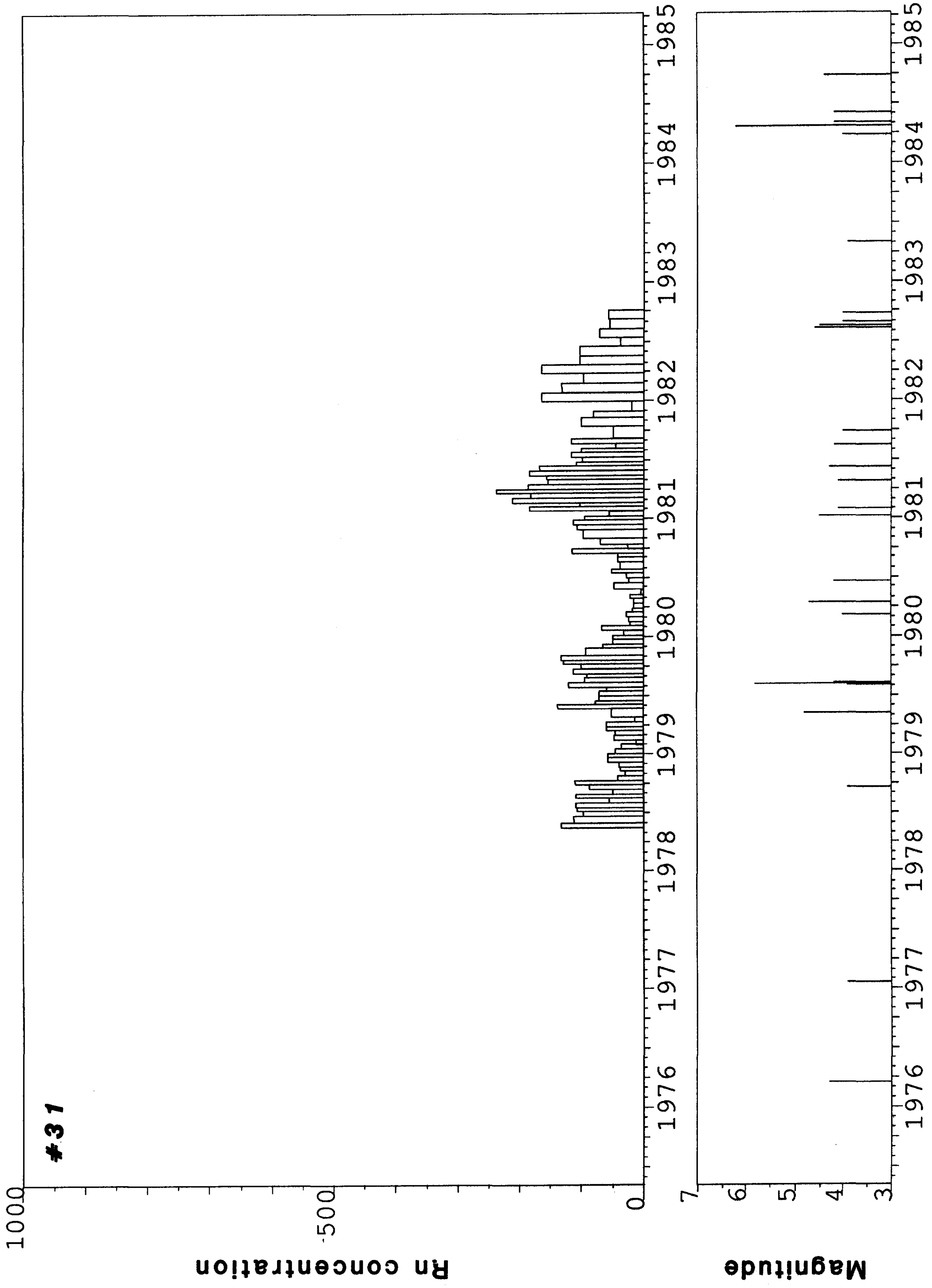

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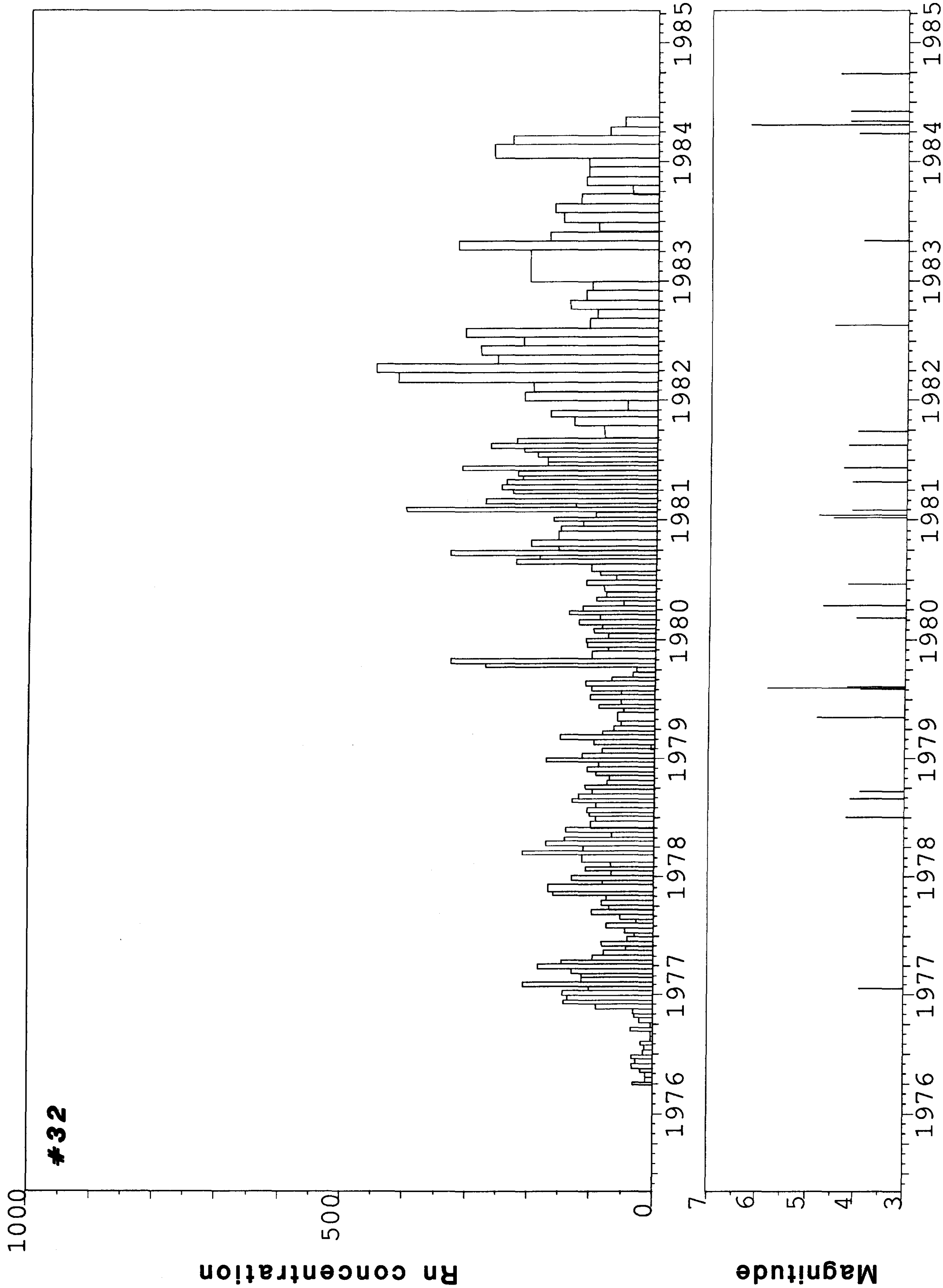

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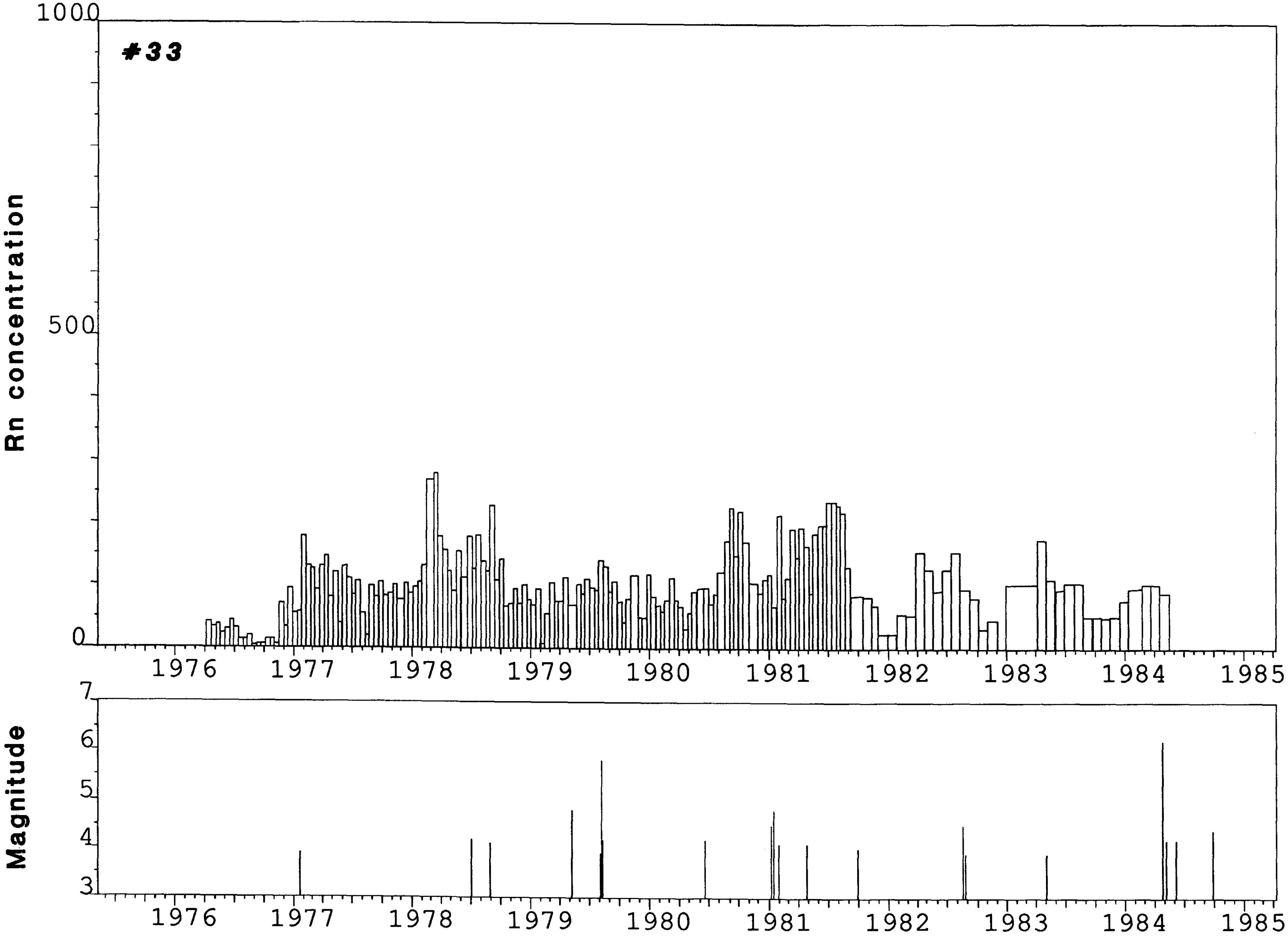

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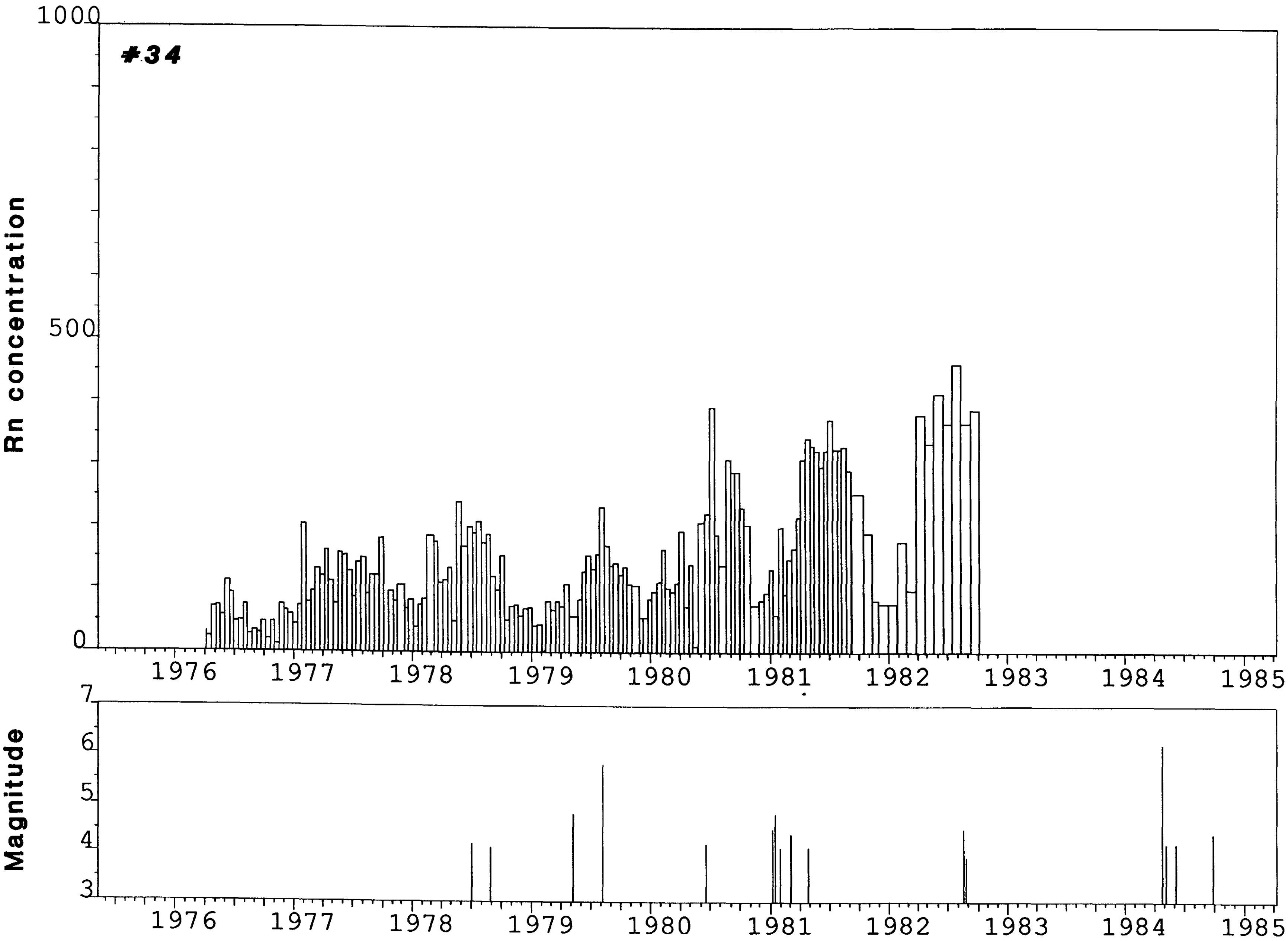

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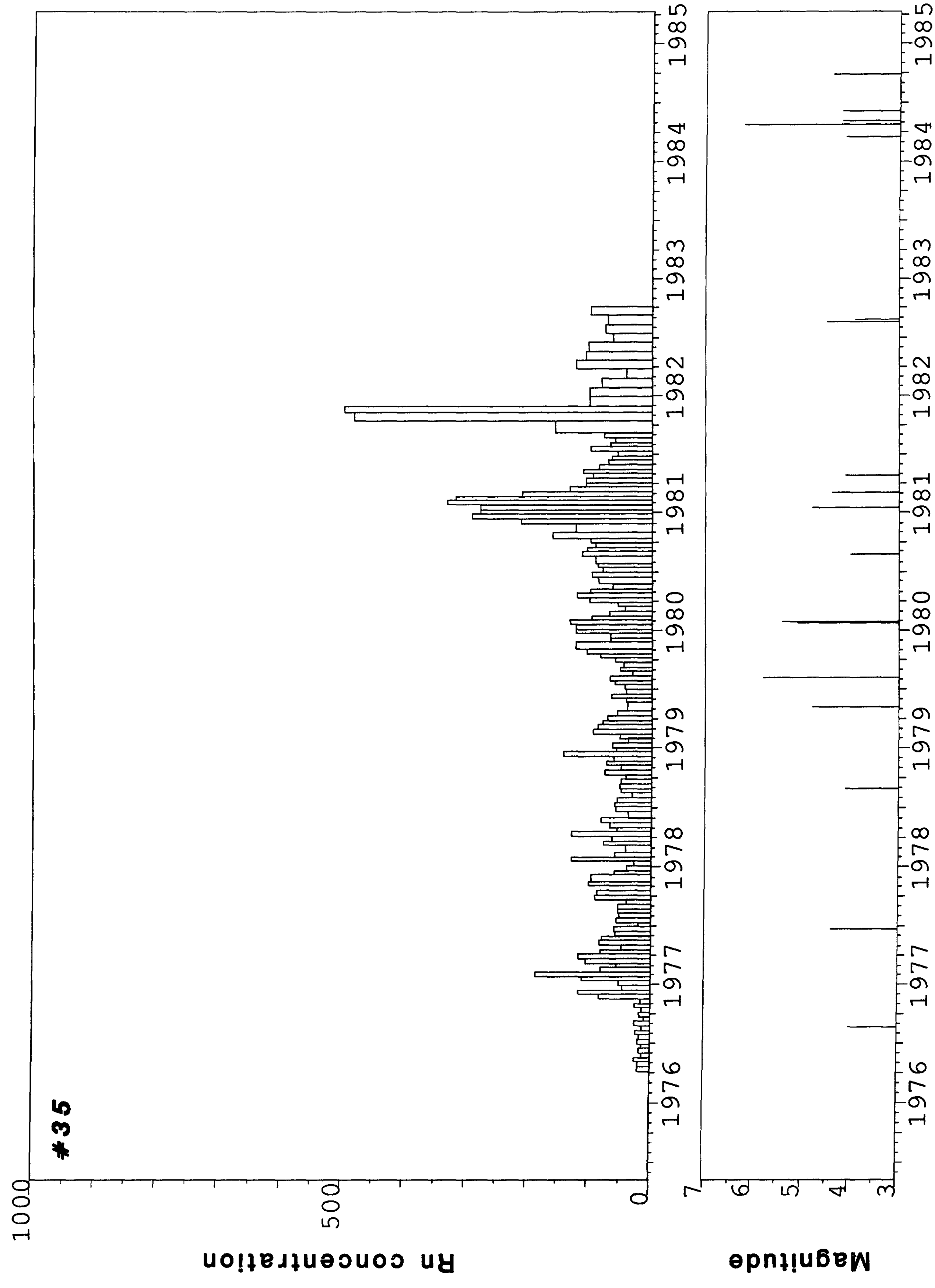

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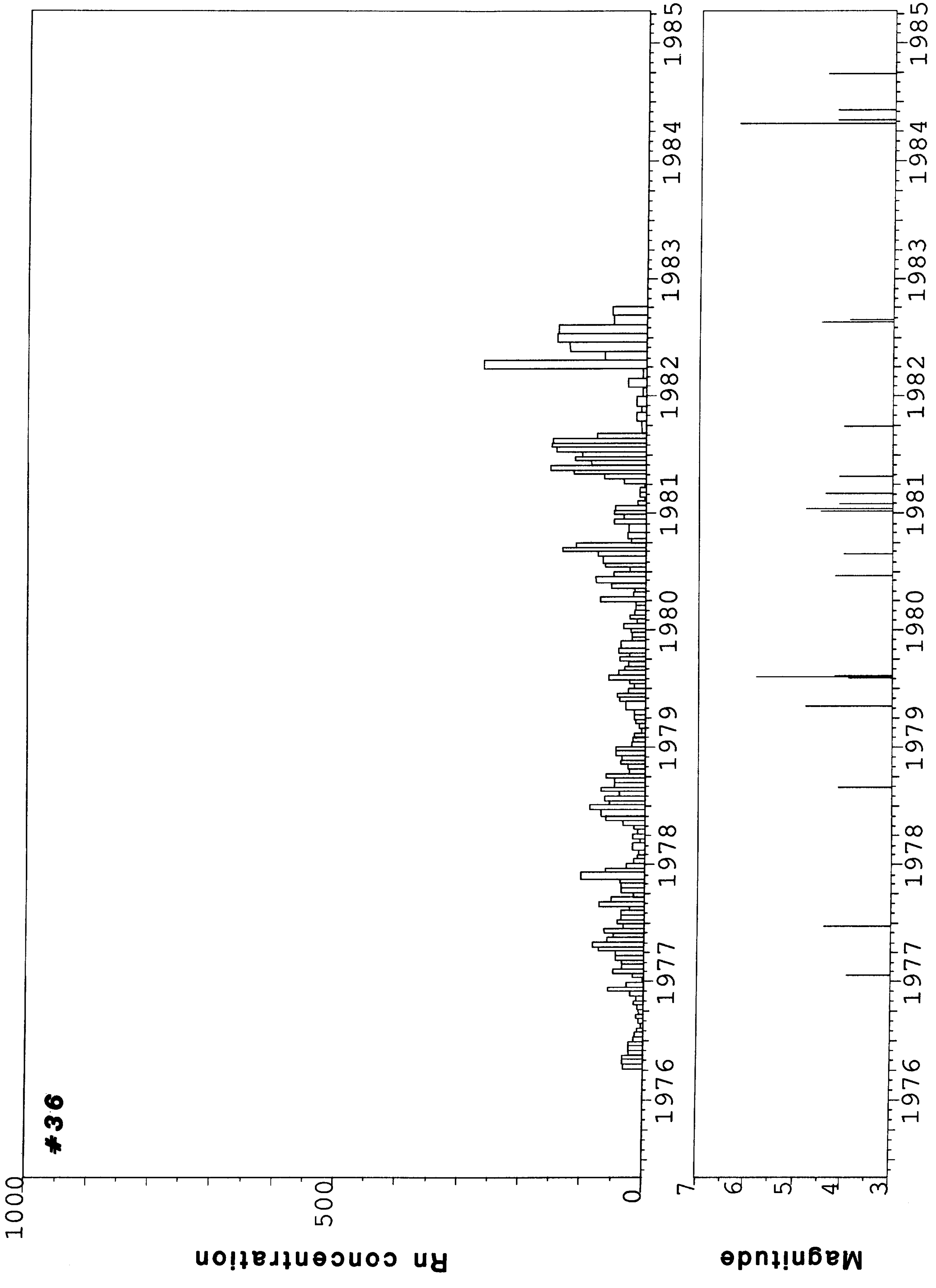

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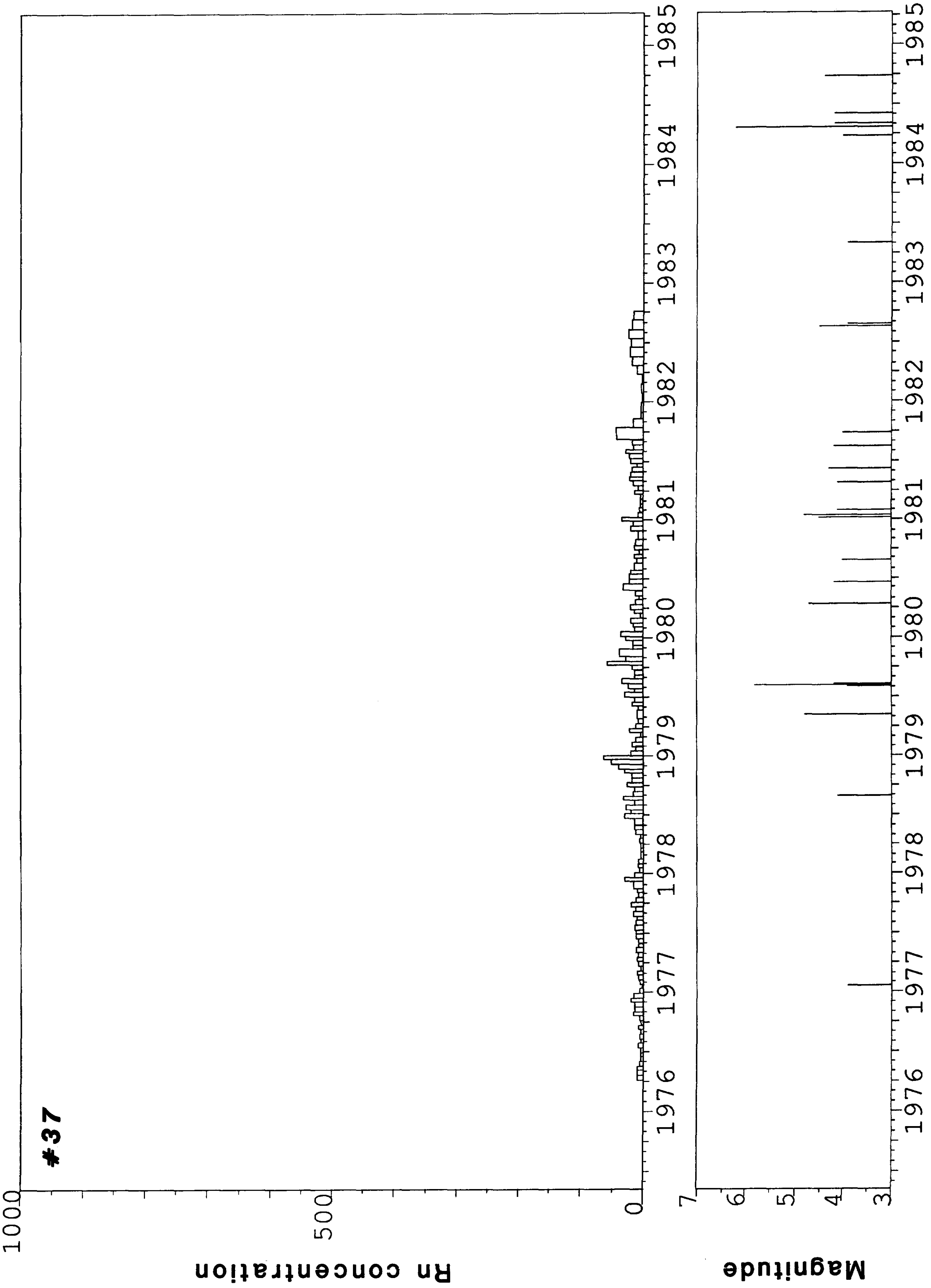

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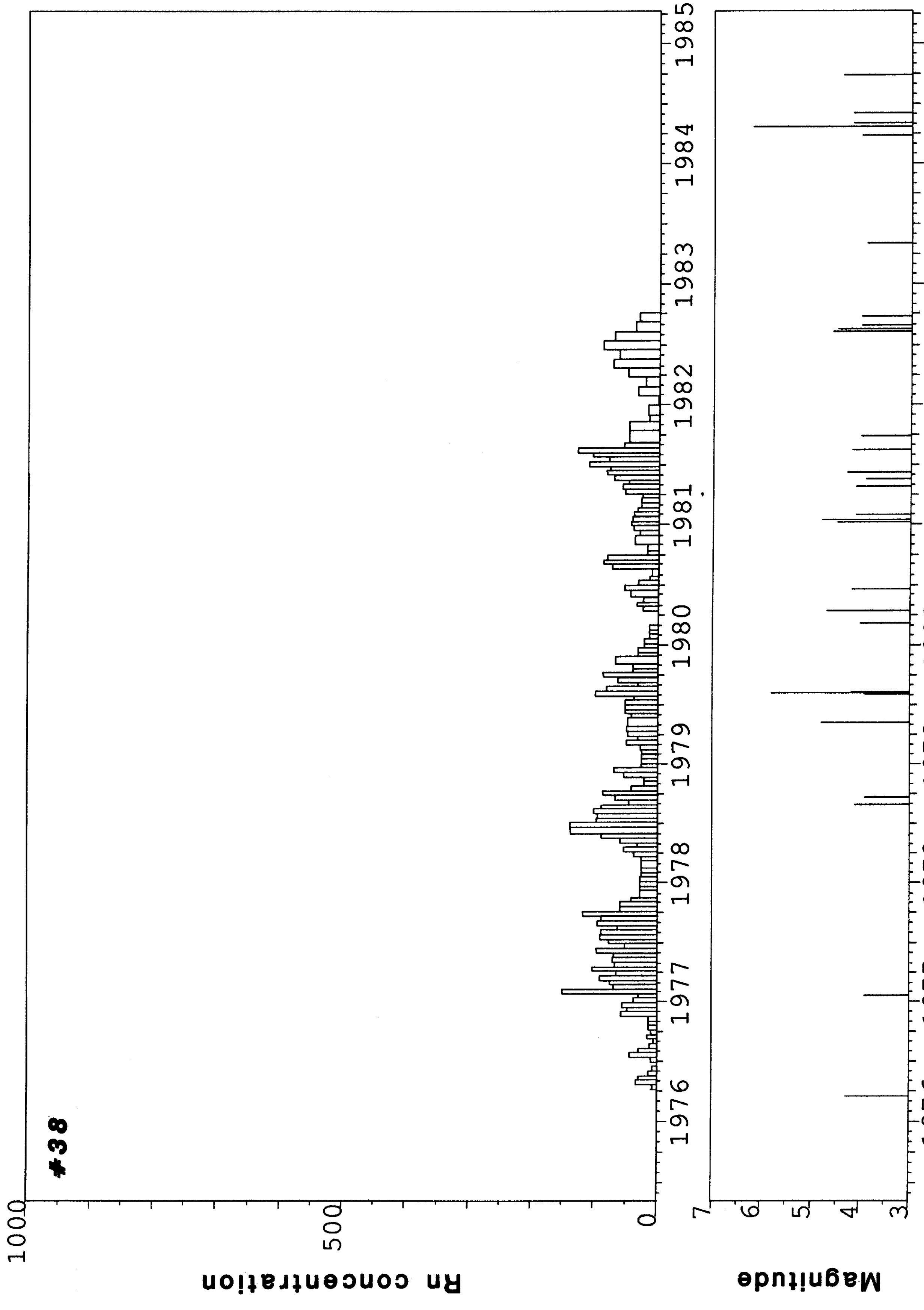

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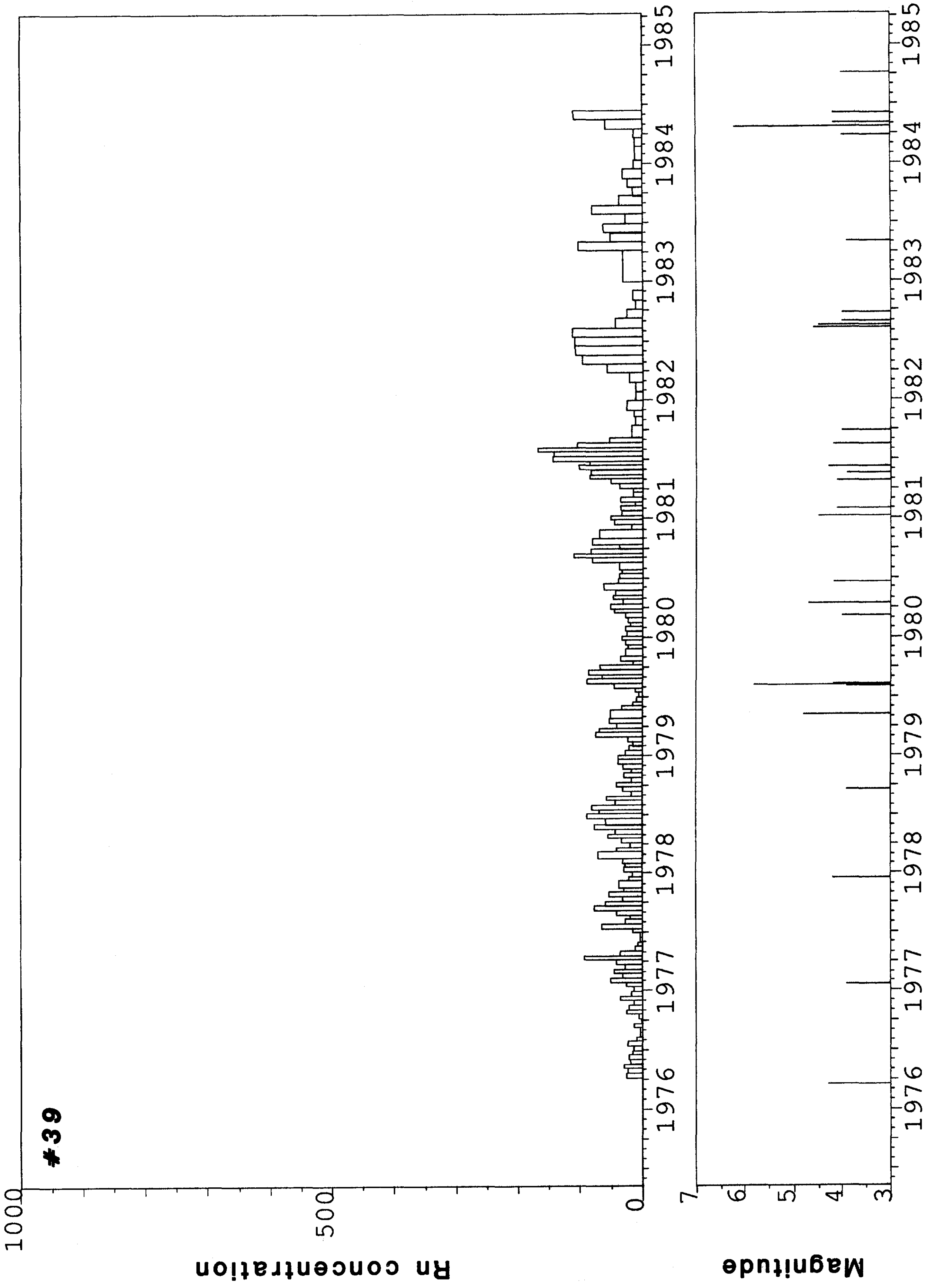

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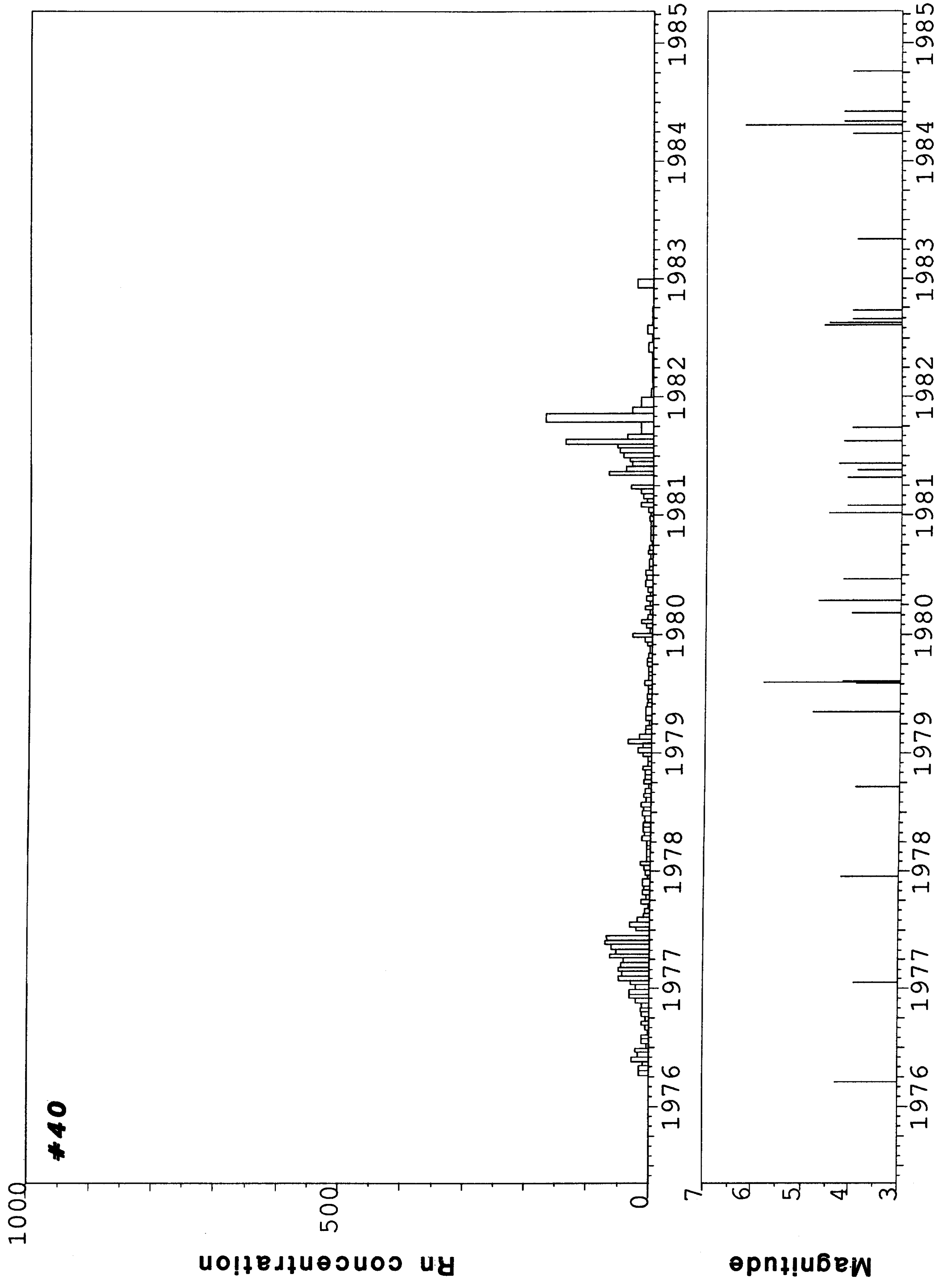

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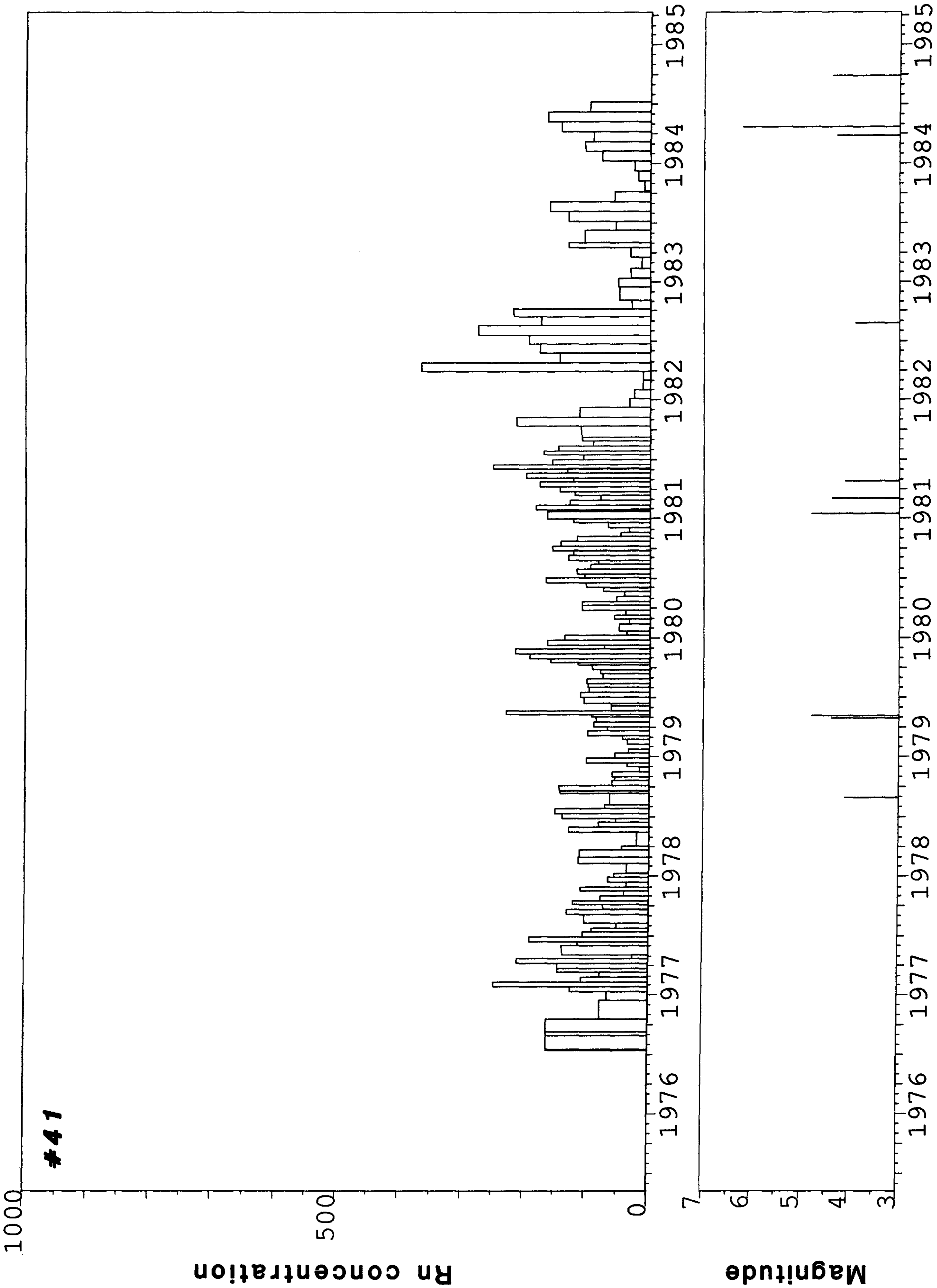

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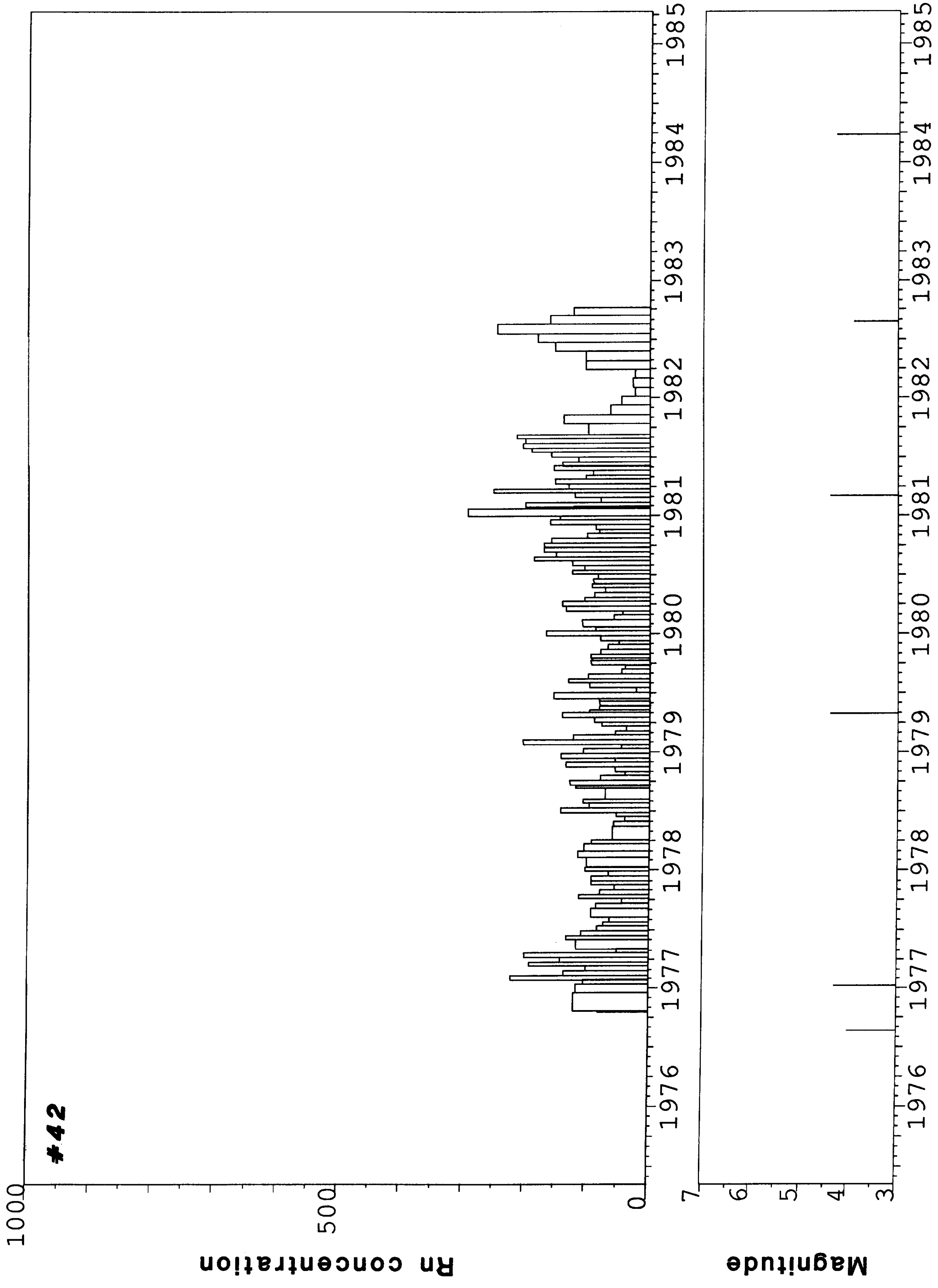

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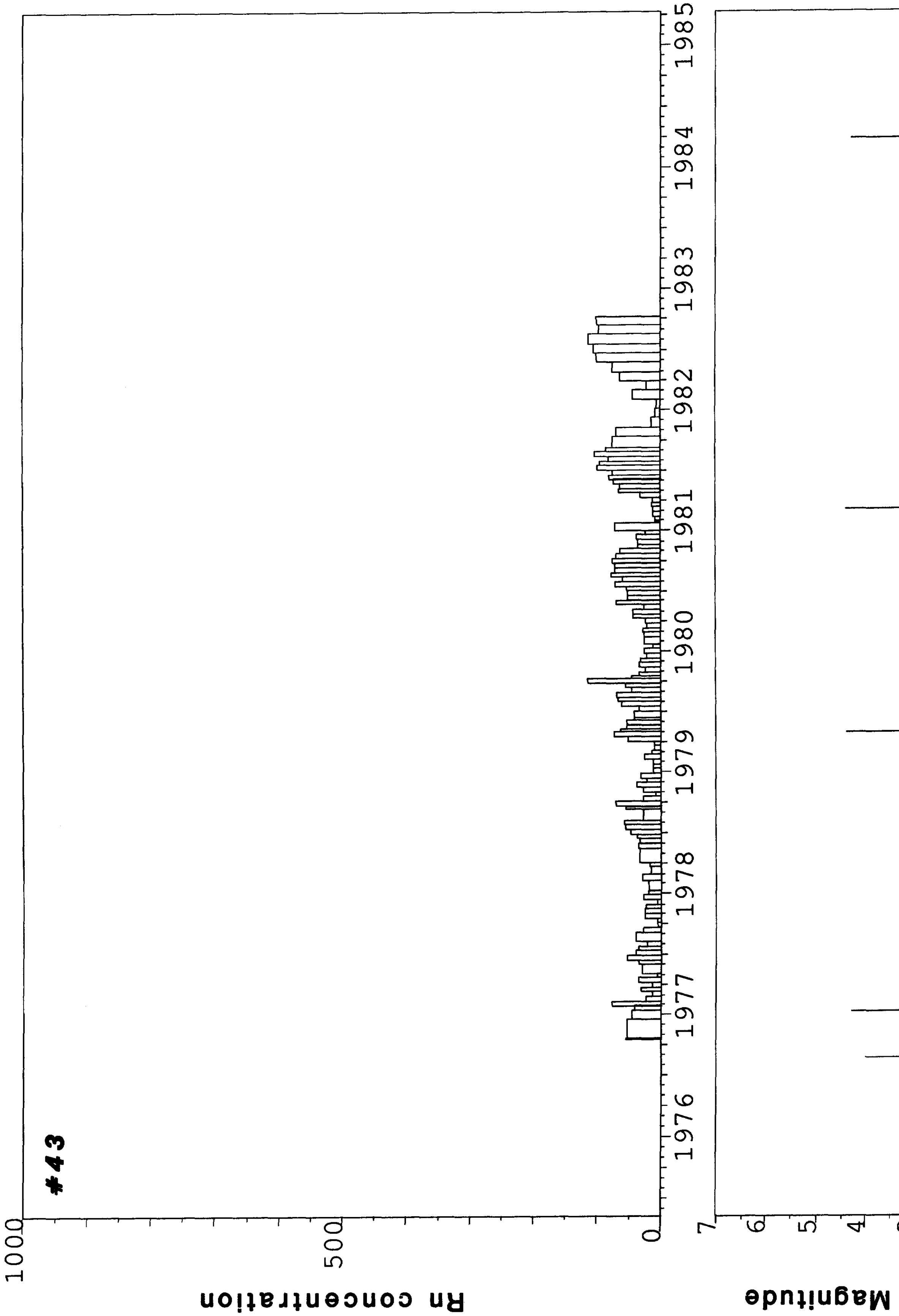

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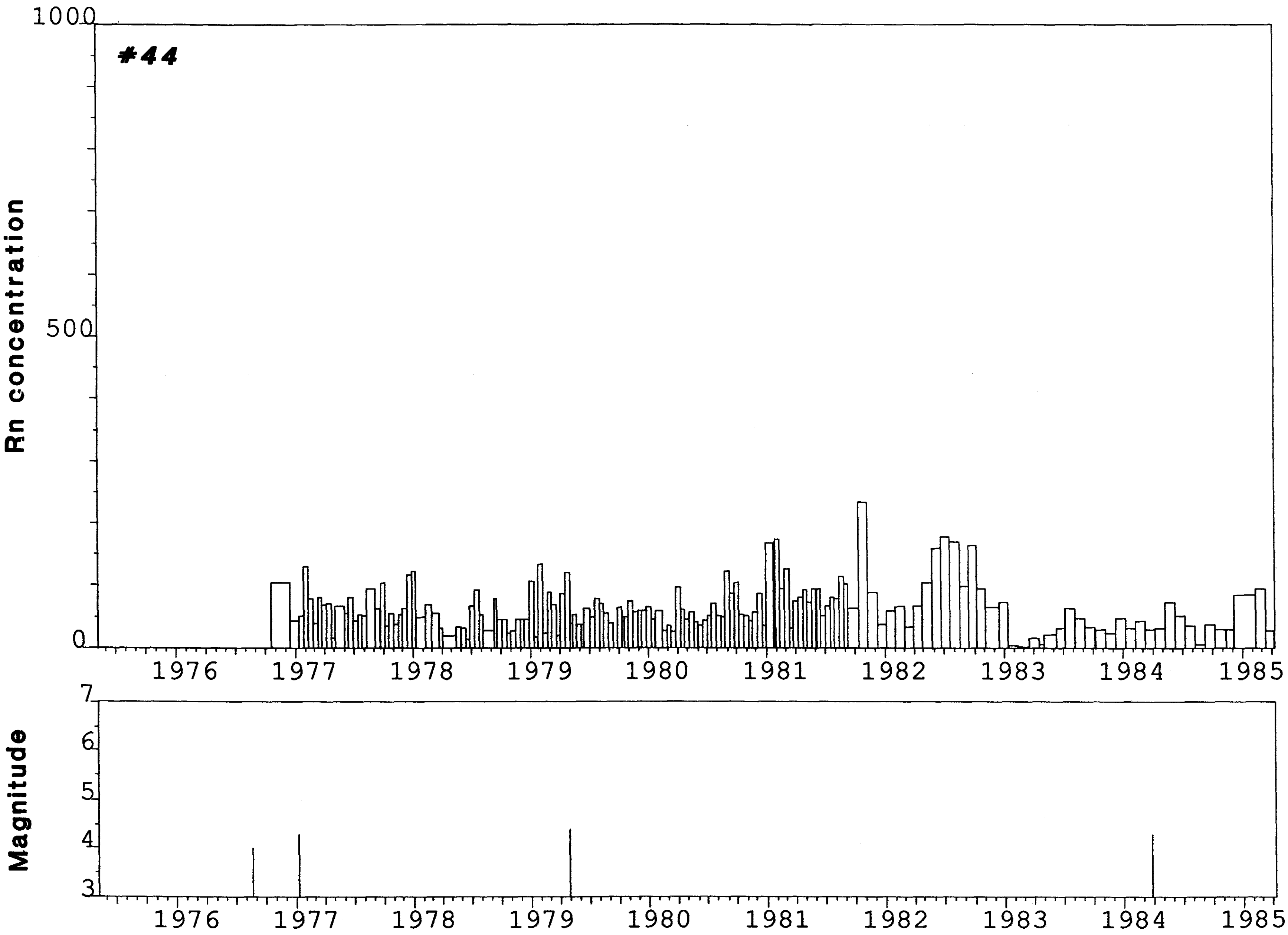

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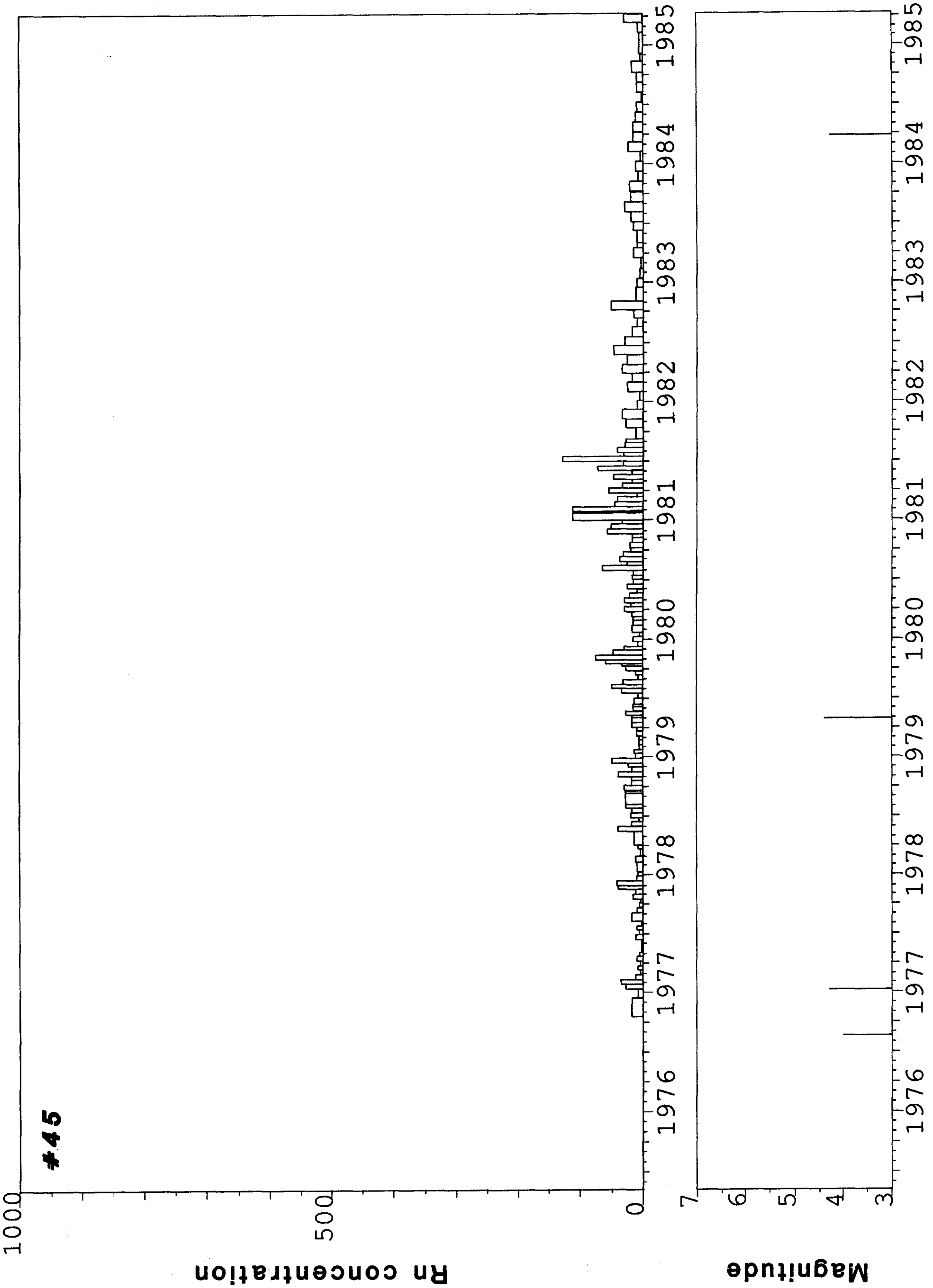

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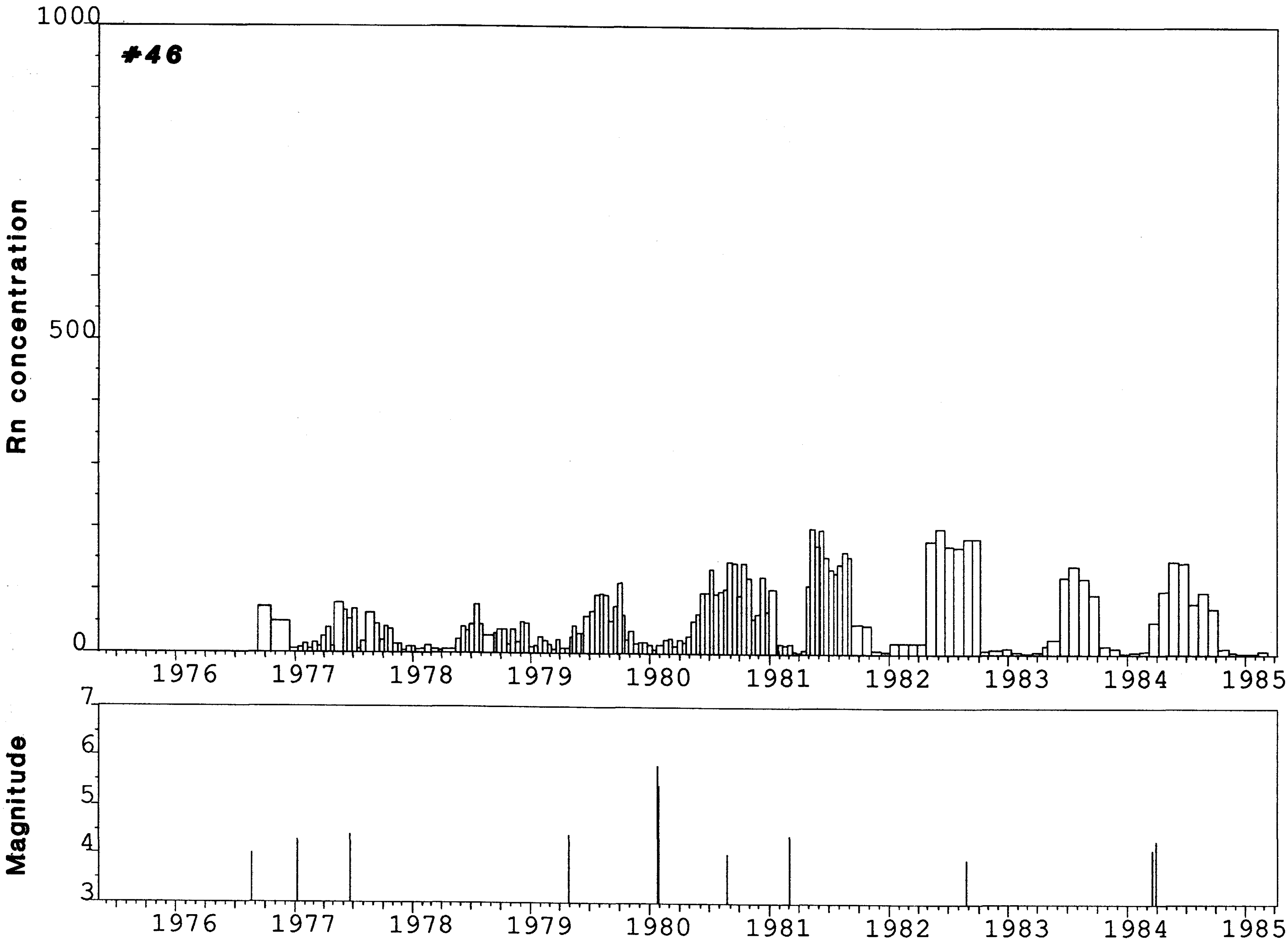

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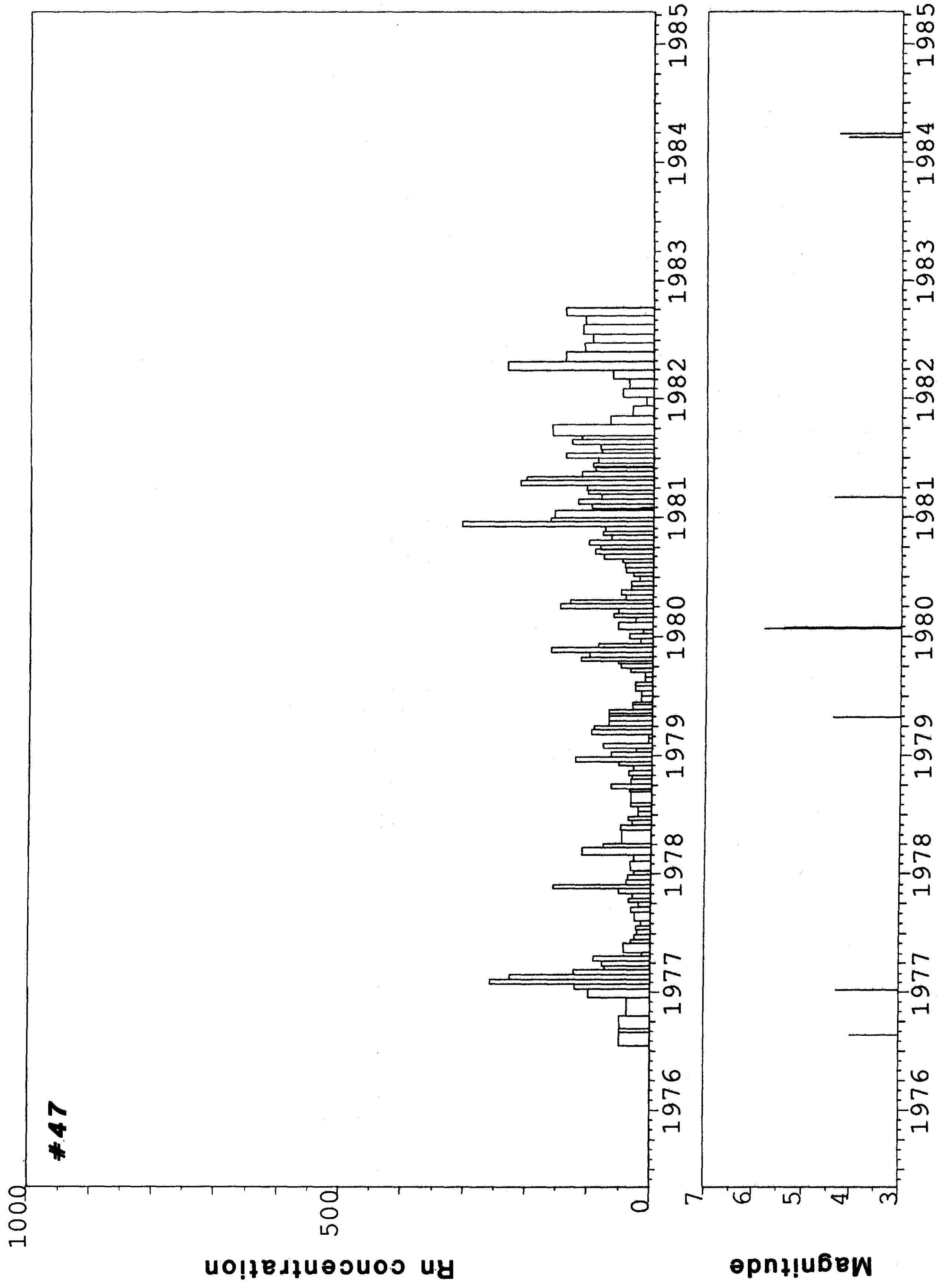

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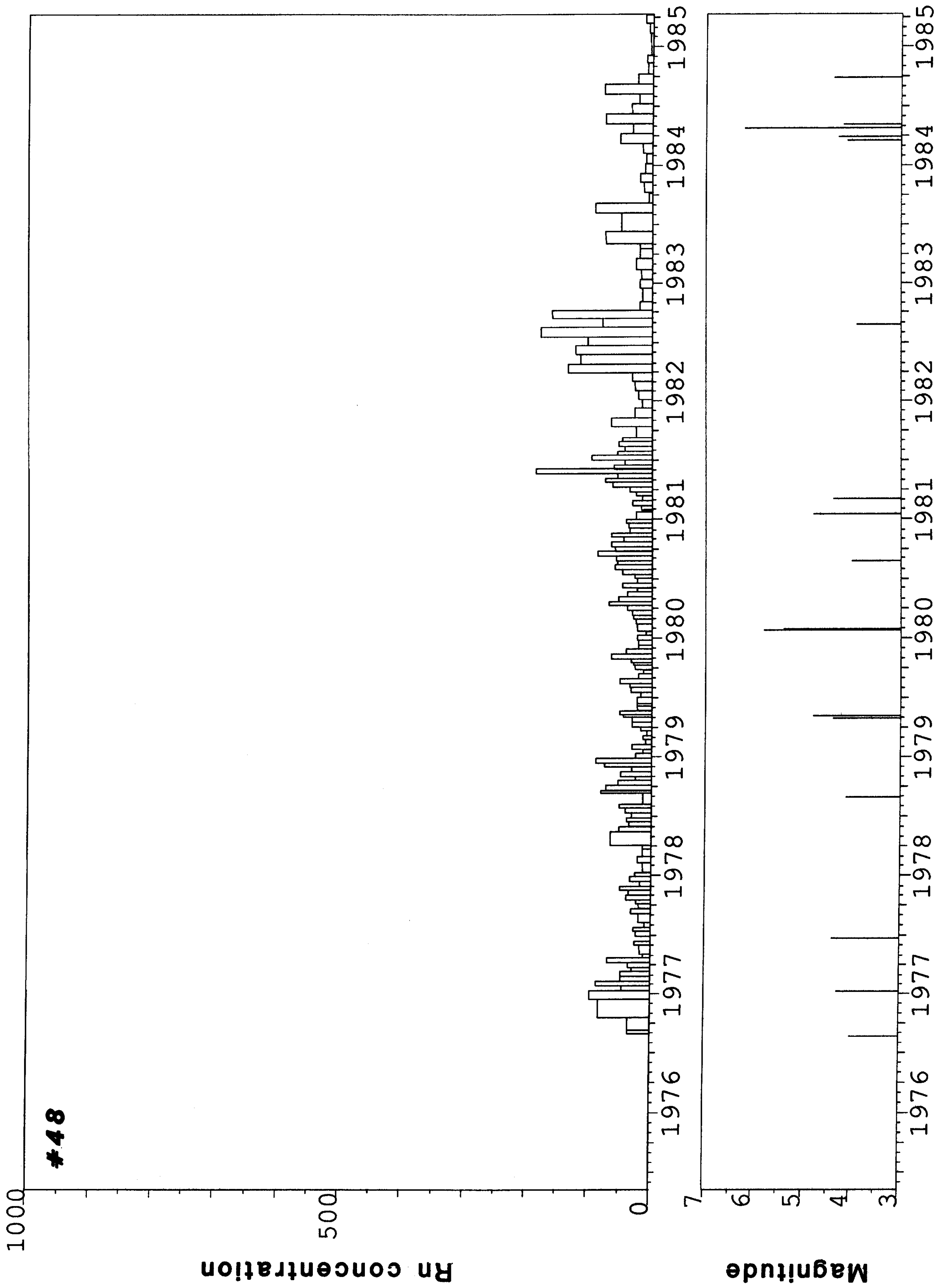

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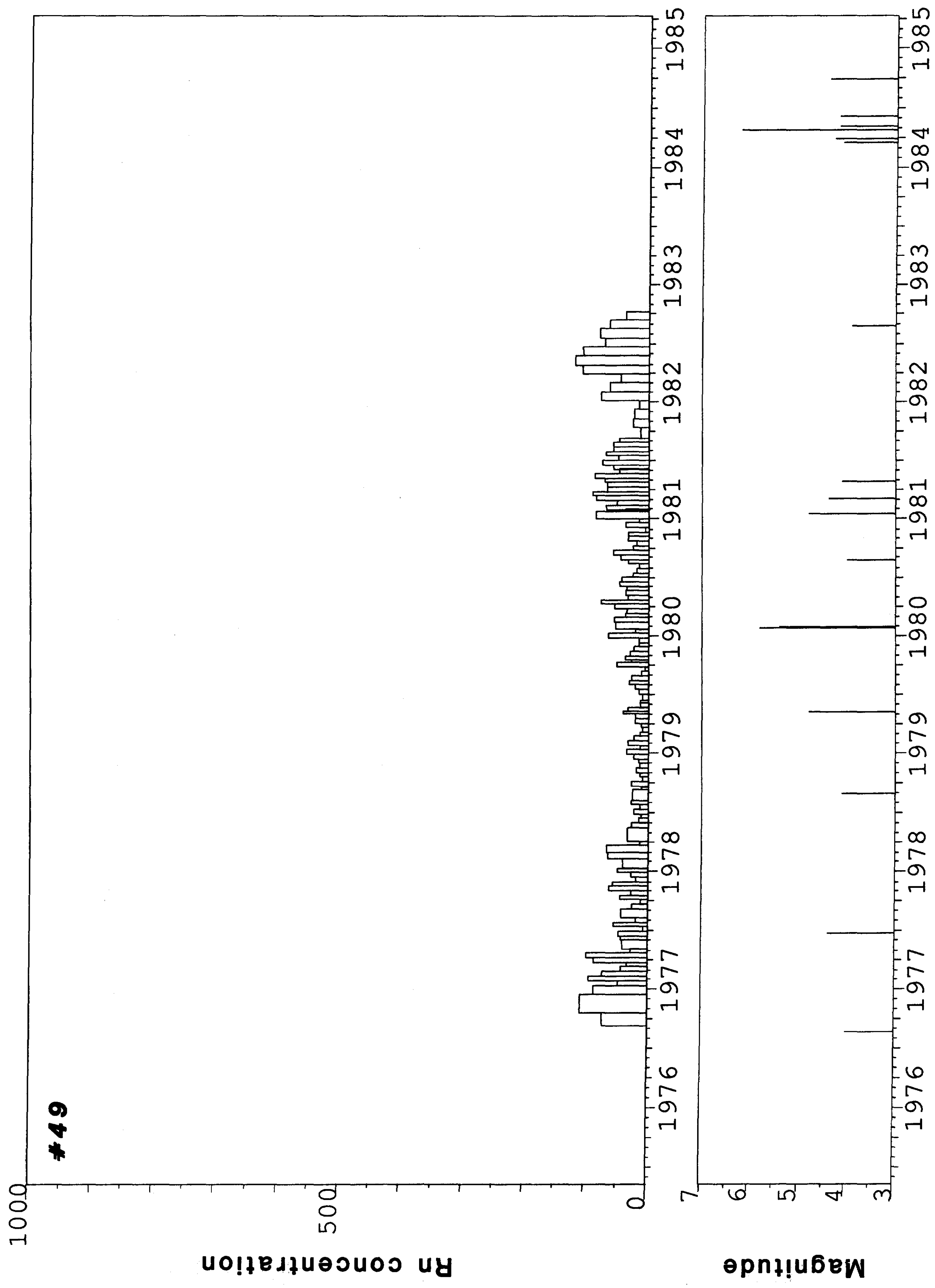

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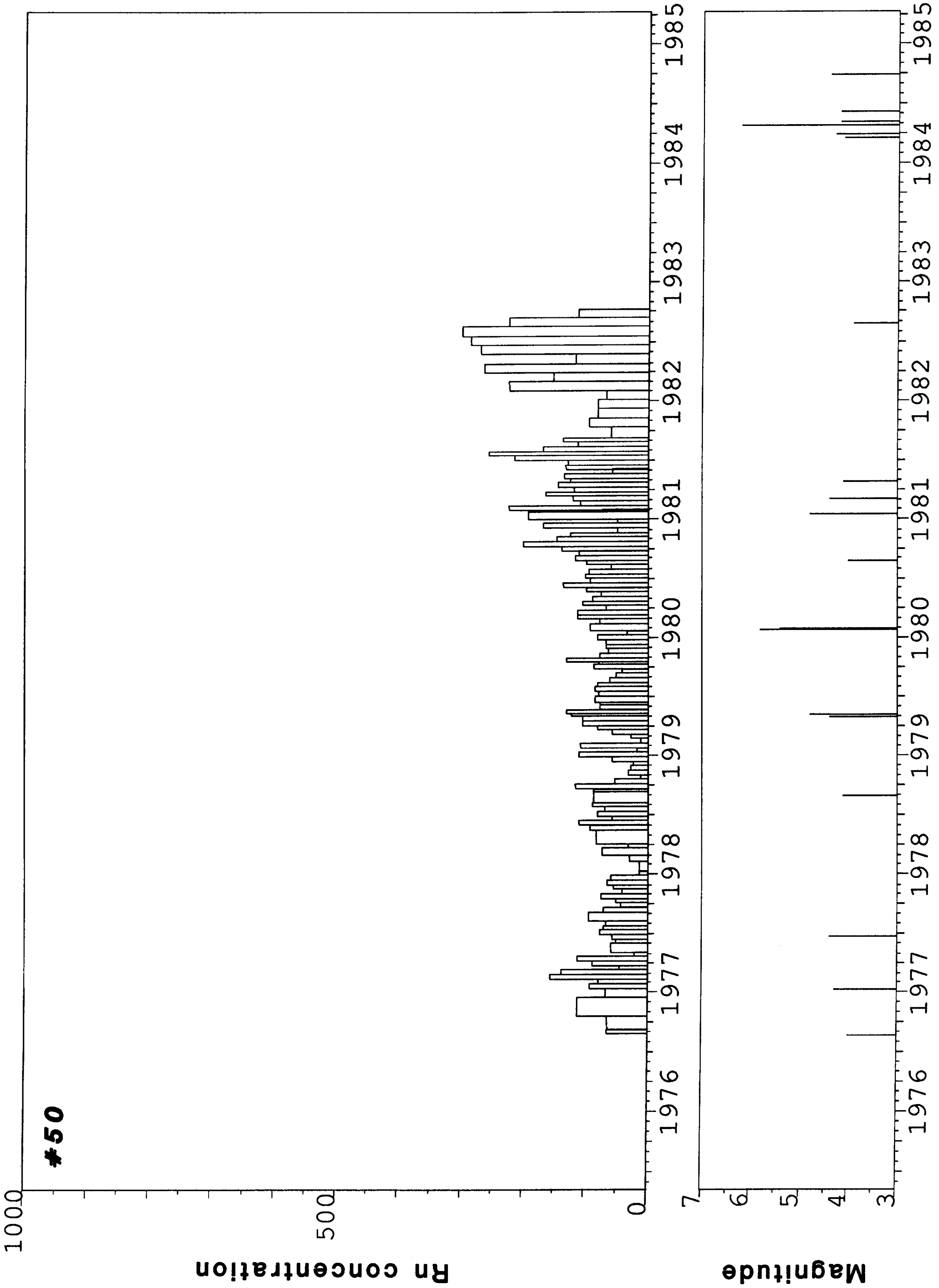

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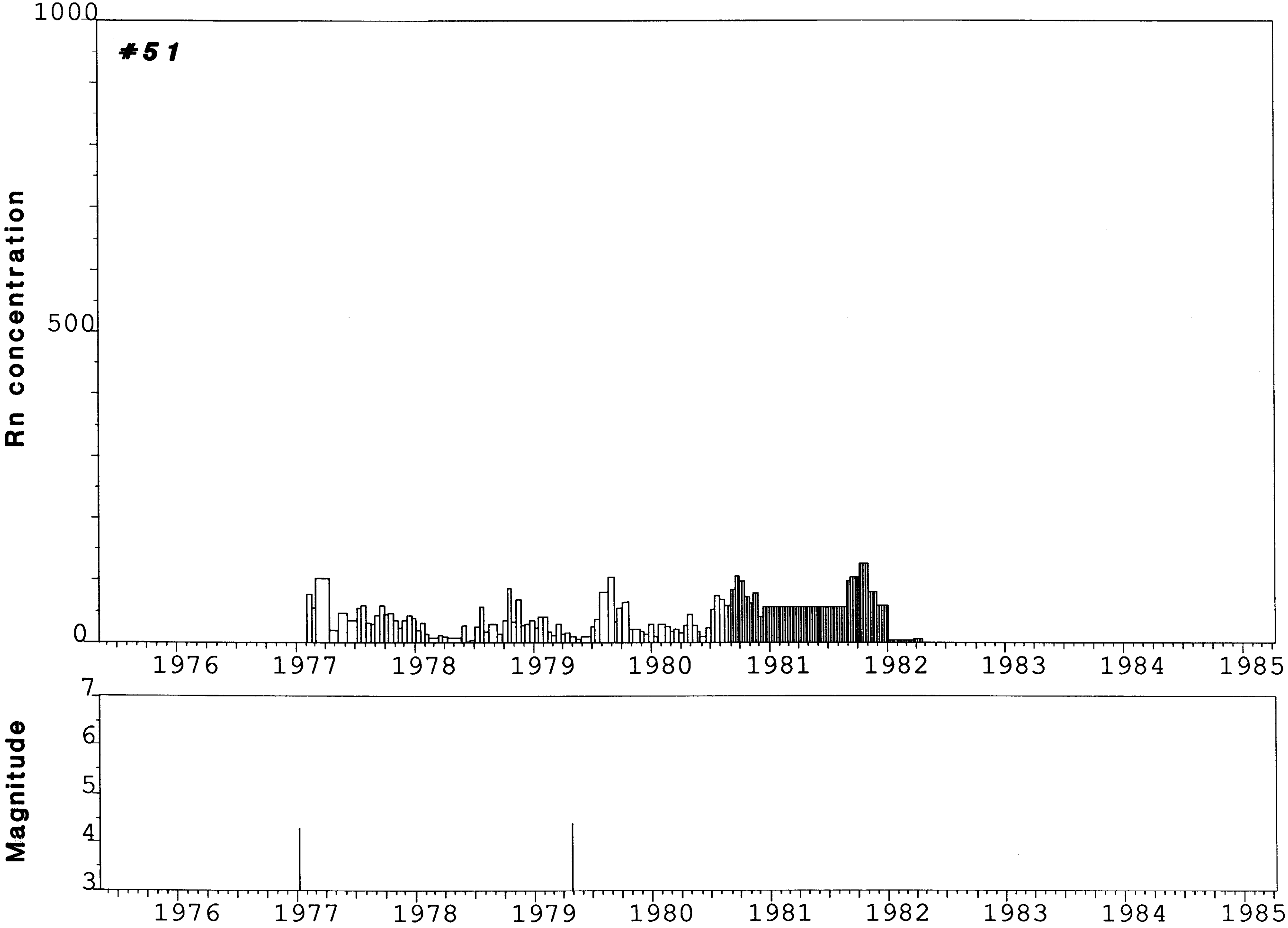

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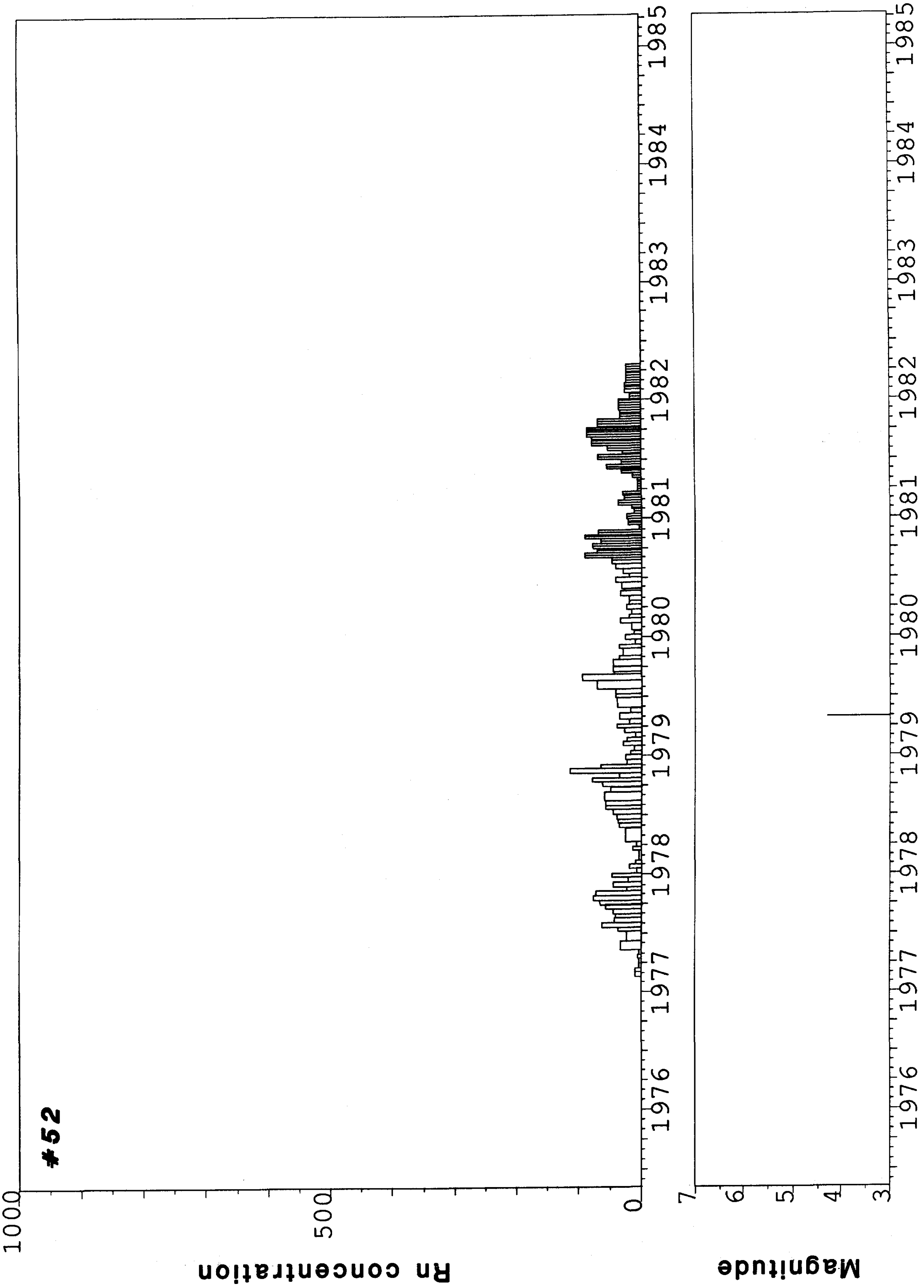

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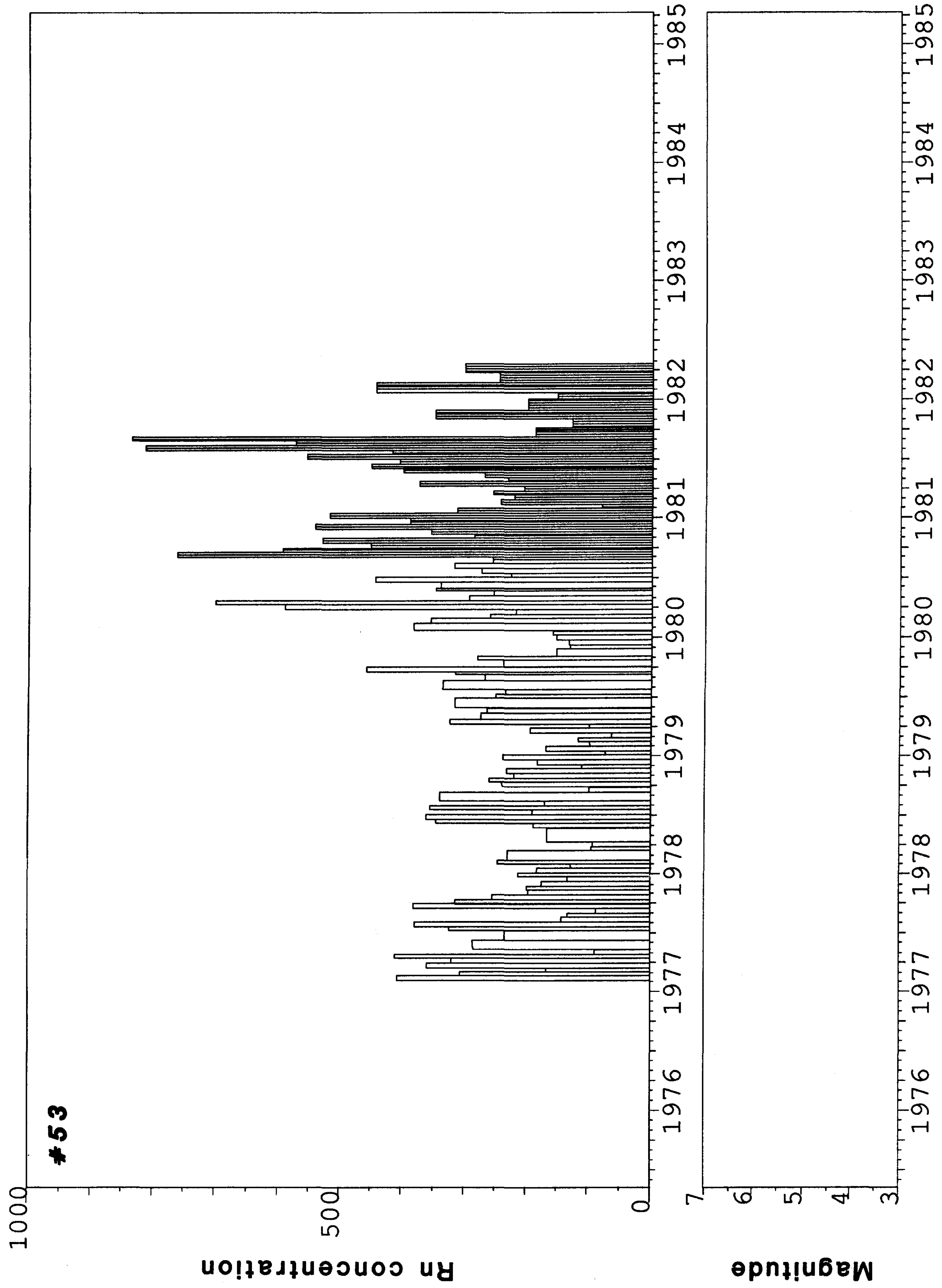

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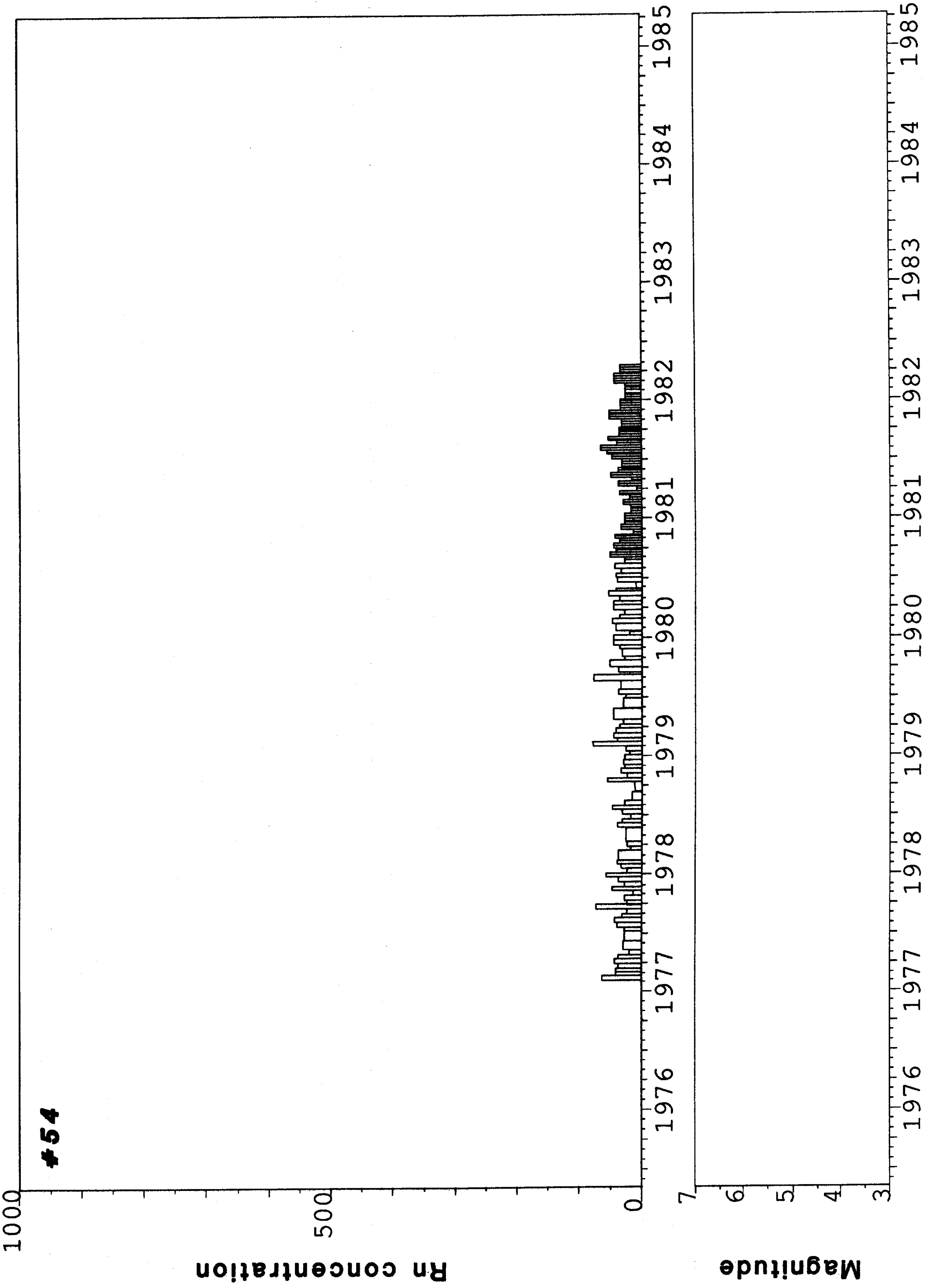

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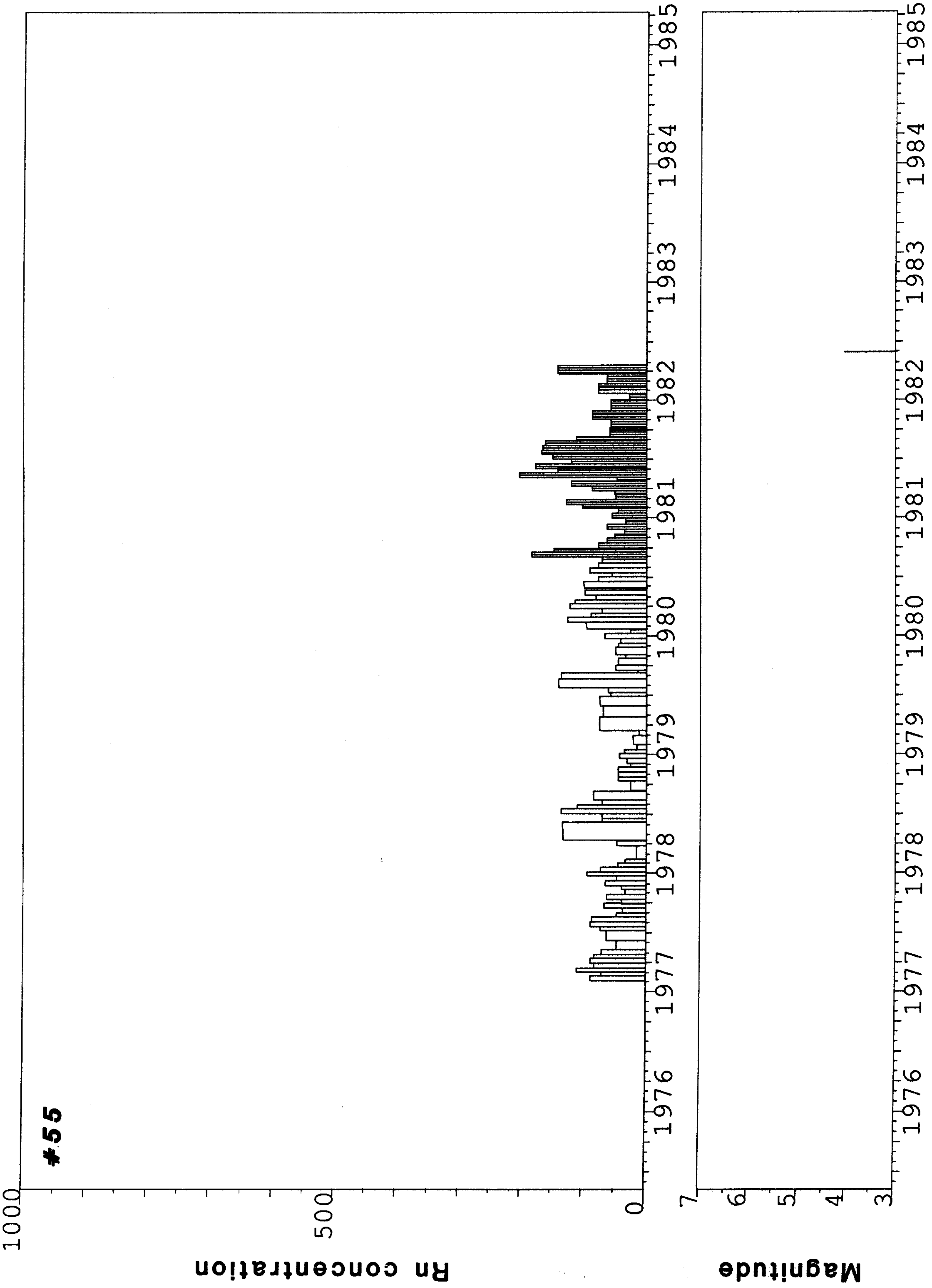

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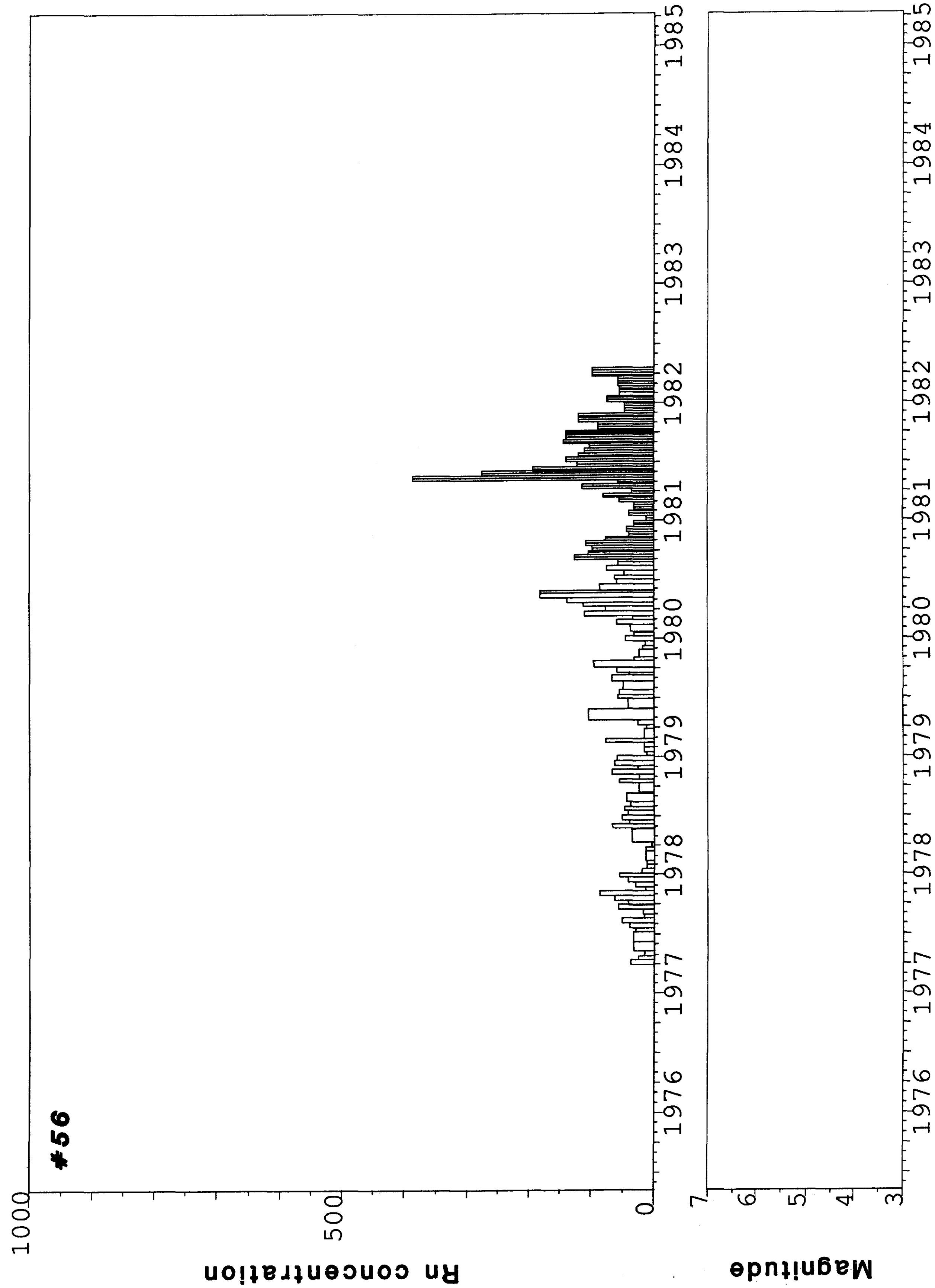

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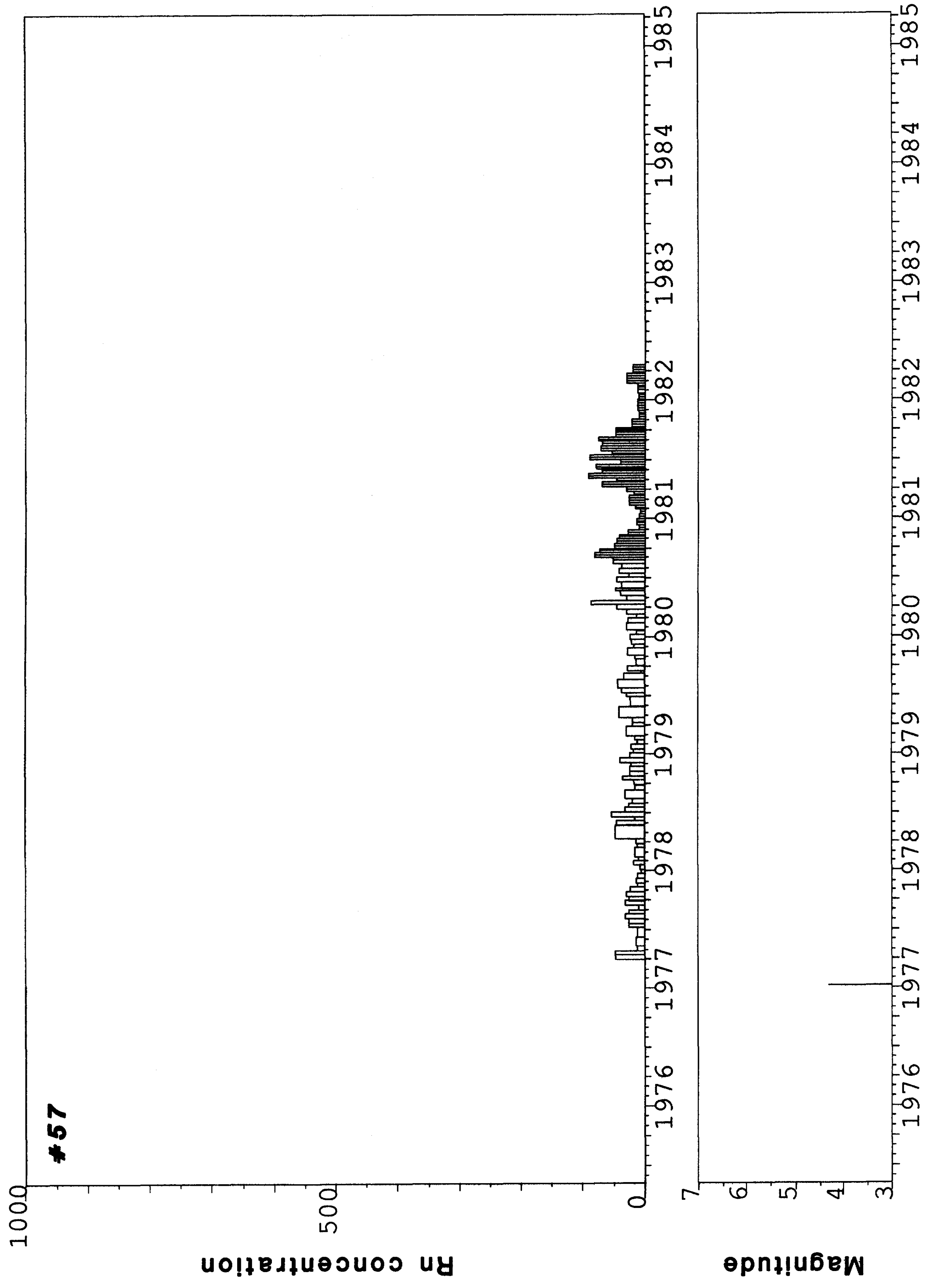

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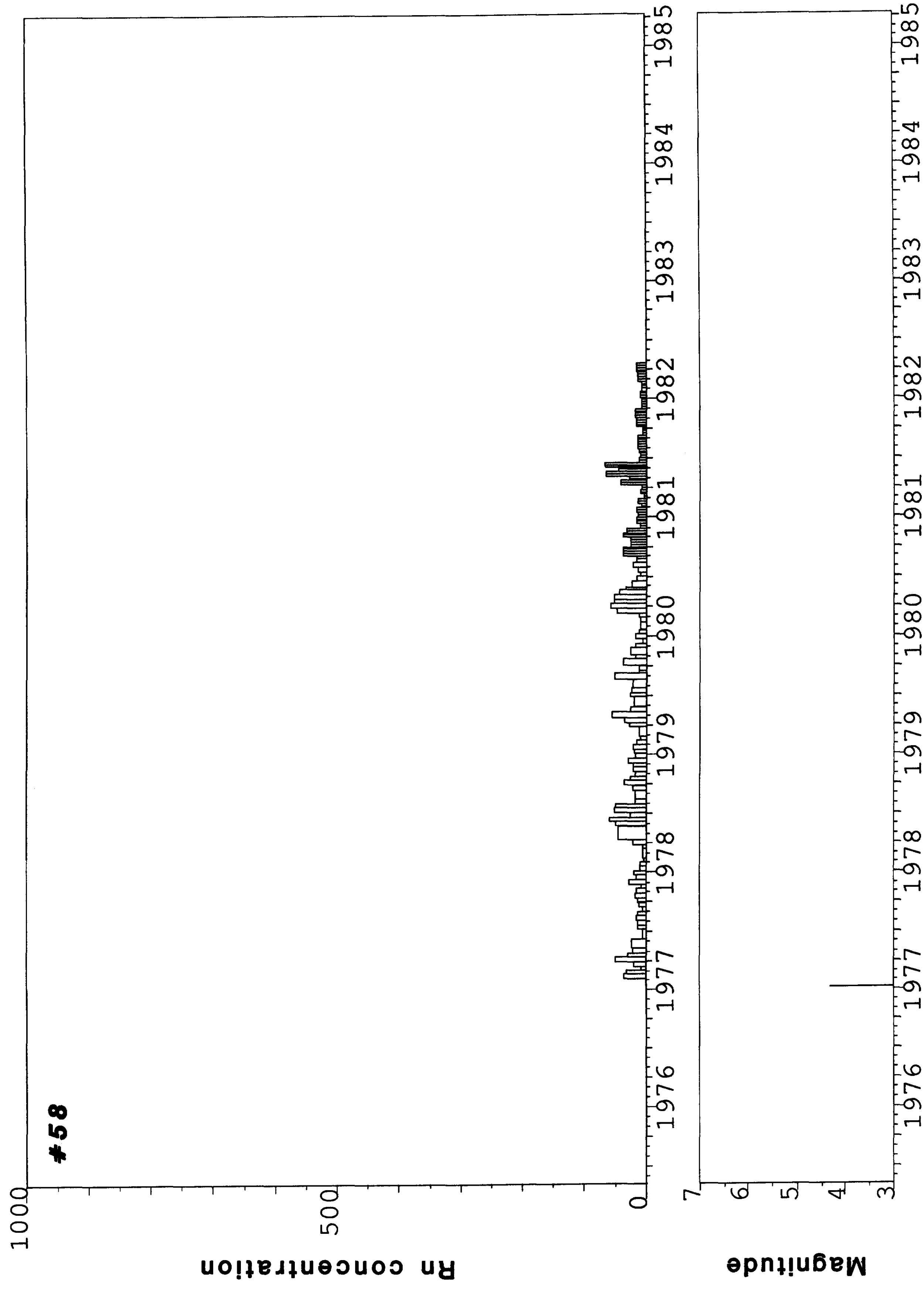

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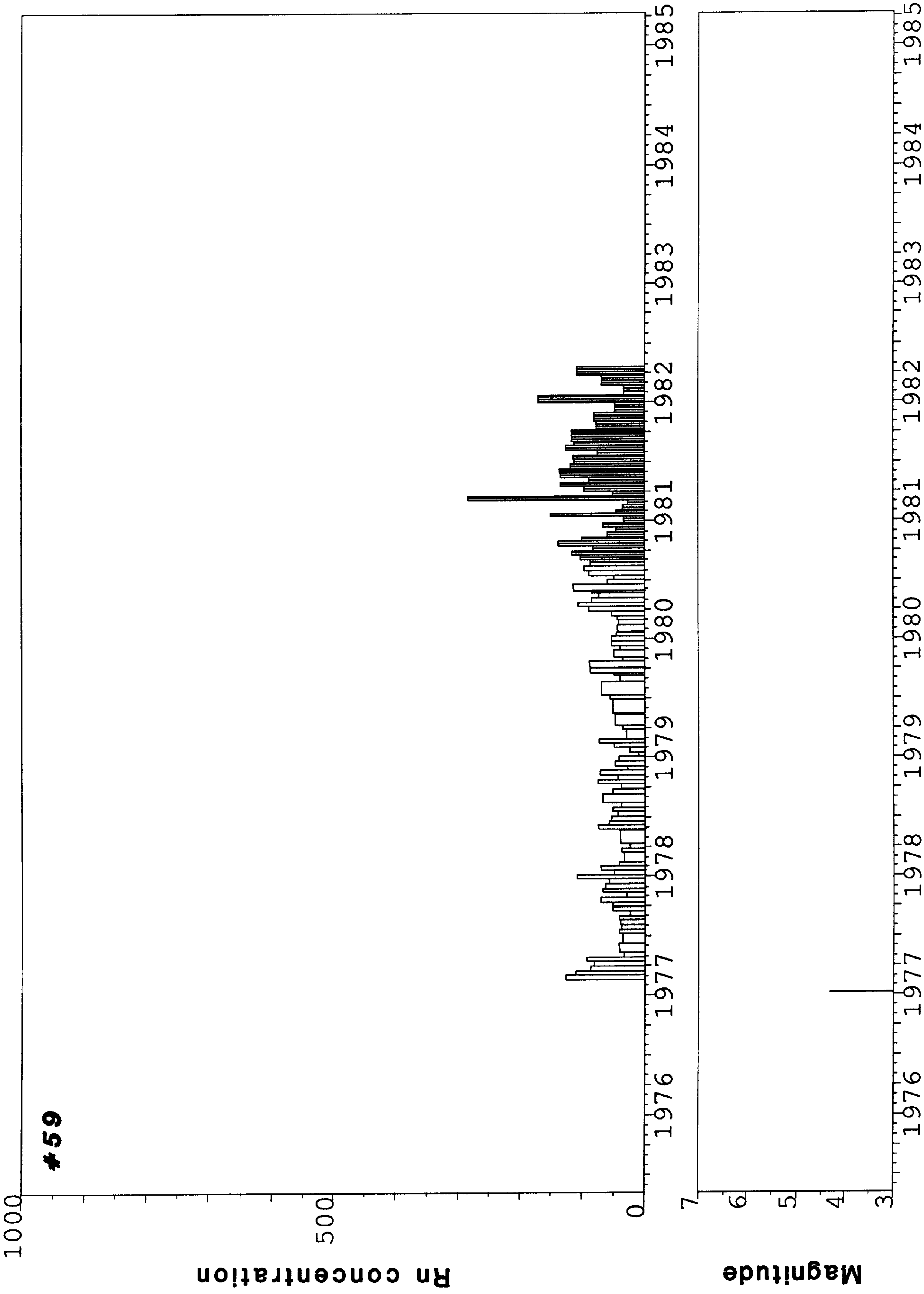

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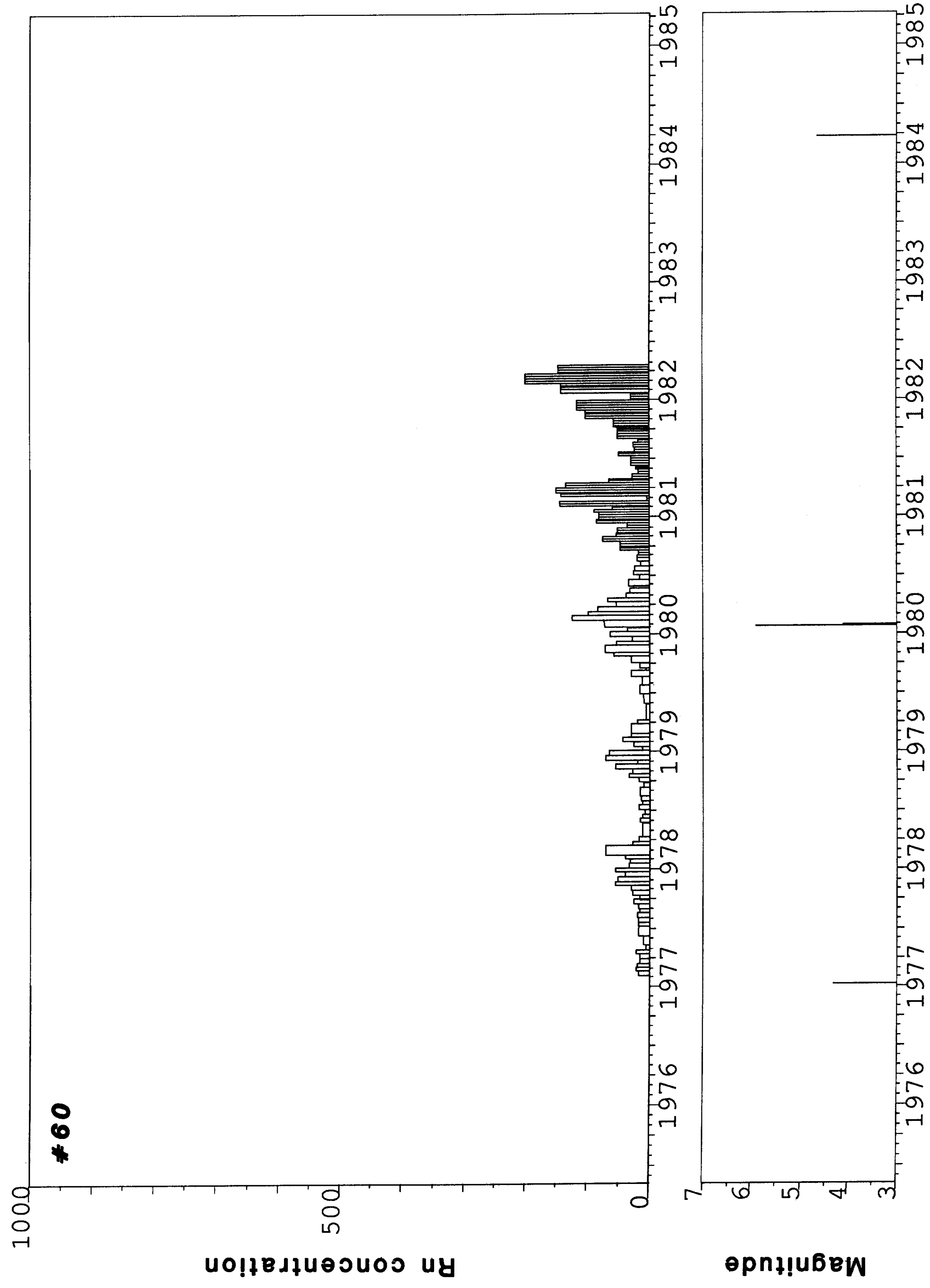

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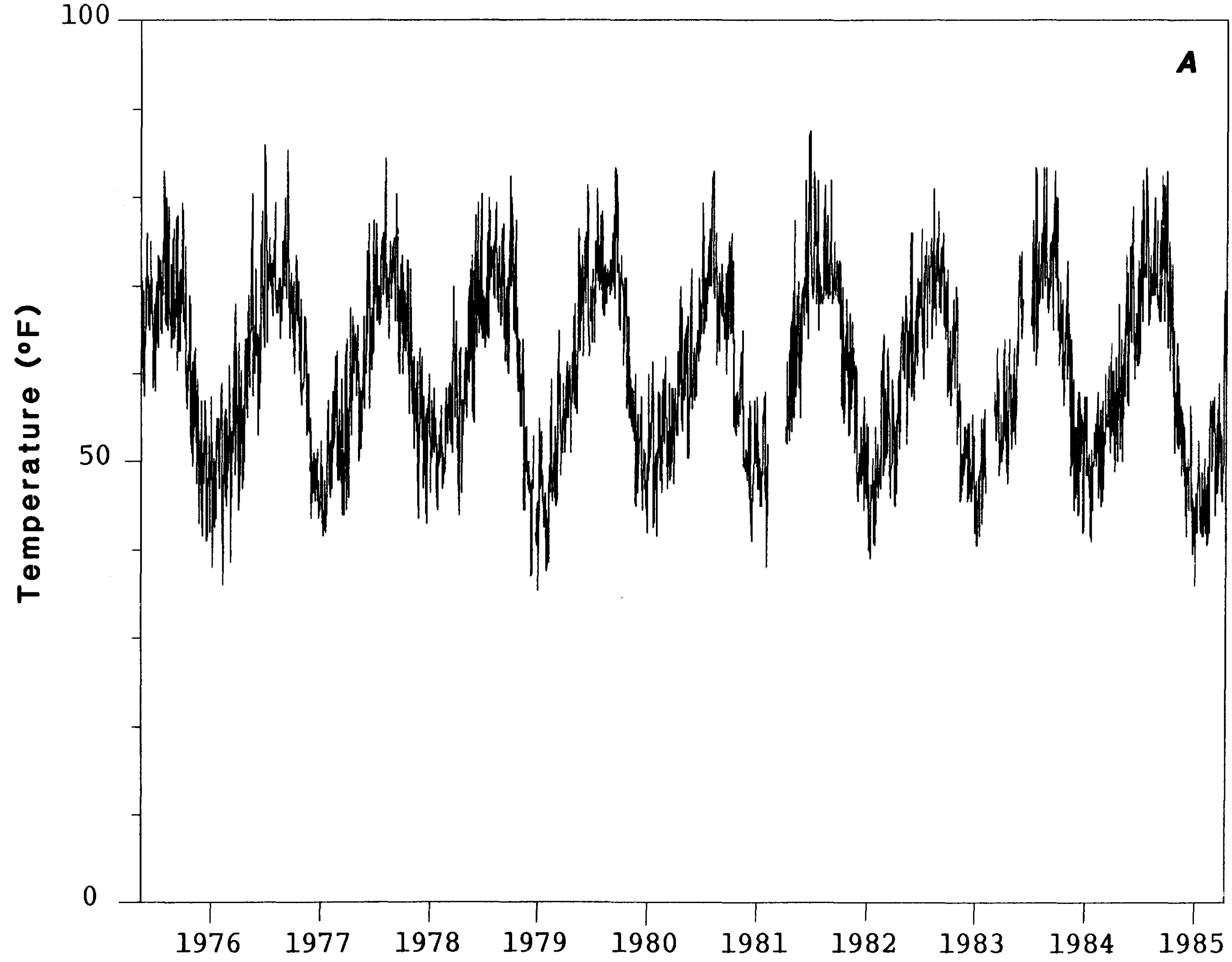
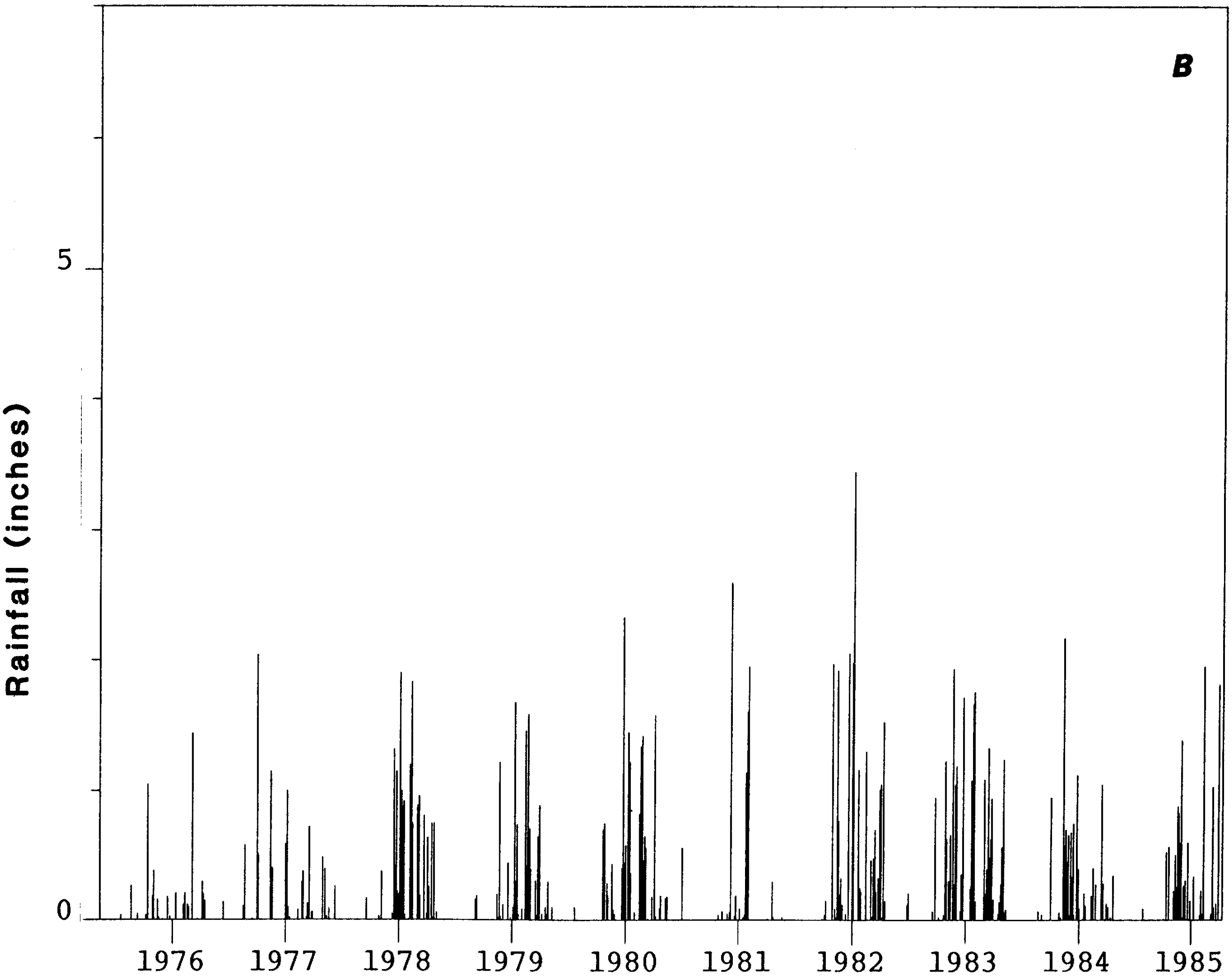


Figure 52. Time series of (*A*) atmospheric temperature (maximum/minimum) and (*B*) daily rainfall recorded at Gilroy, near the center of the radon-monitoring network.



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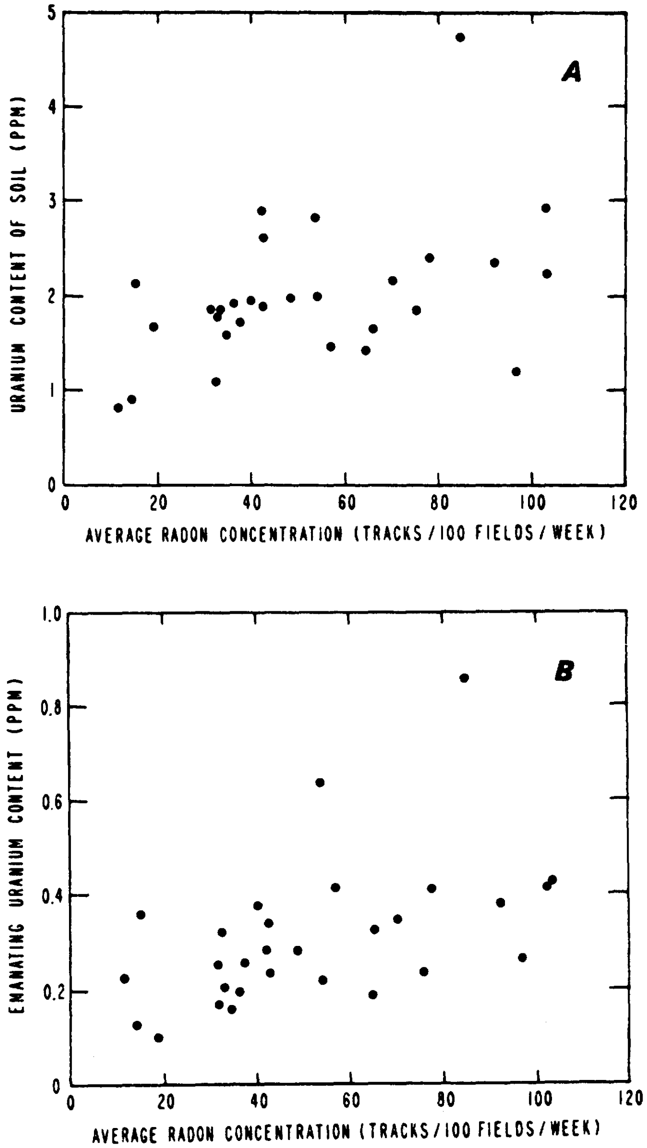

Figure 53. Average radon concentrations recorded from 1975 to 1978 at stations 1 to 30 versus (*A*) uranium content of local soil and (*B*) emanating part of the uranium content (after King, 1980). ppm, parts per million.

RADON EMANATION FROM URANIUM MILL TAILINGS

Edward R. Landa

Abstract

Alkaline circuit and a mixture of acid and alkaline circuit uranium mill tailings sampled at an inactive millsite near Monticello, Utah, and tailings from an active, acid-leach uranium mill were separated into particle-size fractions ranging from +10 mesh to −325 mesh by dry and wet separation techniques. The radium-226 contents and radon-222 emanation coefficients of these fractions were determined. Dry tailings show a high degree of aggregation that tends to mask the relationship of properties, such as radium content and radon-emanating power, to dispersed-particle size. Coarse-tailings fractions (+325 mesh) had emanation coefficients that were from 25 to 45 percent lower than those of their fine-fraction counterparts. Emanation coefficients measured for tailings derived from a salt-roast/carbonate-leach process suggest that such ore roasting does not lead to reductions in radon emanation in the tailings derived therefrom.

Three ore samples of differing uranium mineralogies and rock types were leached with either sulfuric acid or sodium carbonate solutions under laboratory conditions to simulate acid or alkaline uranium milling practices. The radon emanation coefficients of the initial ore and of the tailings derived from them were compared. No consistent pattern enabling the prediction of radon emanation coefficients of tailings on the basis of ore type was evident. The similarities in the coefficients of the acid-leach tailings suggest that the lixiviant solution plays an important role in determining the form, and hence, the emanating power of the radium retained therein.

INTRODUCTION

Although greater than 90 percent of the uranium is generally extracted during the milling of uranium ores, most of the daughter product activity remains with the tailings (Moffett, 1976). Because of this, uranium mill tailings constitute a voluminous form of low-level radioactive waste. The quantity of these tailings in the United States is presently about 200 million tons (U.S. Department of Energy 1981, 1986). Radon release to the atmosphere constitutes an important exposure pathway from these wastes, and studies have been undertaken to assess the roles of (1) particle size and (2) ore type and milling process on such release.

RADIUM-226 CONTENTS AND RADON EMANATION COEFFICIENTS OF PARTICLE-SIZE FRACTIONS OF ALKALINE, ACID, AND MIXED URANIUM MILL TAILINGS

For an unconsolidated earth material such as a soil, particle-size distribution can exert a strong influence on its geotechnical behavior and the relative susceptibility of its components to erosion by wind and water. Such relations also have been recognized in the management of uranium mill tailings (UMT). From the geotechnical perspective, "sand" and "slime" tailings have often been intentionally segregated prior to disposal by uranium mill operators to provide sand for tailings embankments (Merritt, 1971) and mine backfill (Markose and others, 1982) or to enable reprocessing of slimes at a later date for recovery of residual mineral values (Ford, Bacon and Davis Utah, Inc., 1978). From the geomorphological perspective, the preferential mobilization of fine particles by wind, raindrop impact, and flowing water is recognized as an important environmental transport phenomenon. As both operating practices at UMT disposal areas and natural forces tend to segregate tailings by particle size, it is important to understand how tailings' radiologic characteristics, such as the radium-226 (^{226}Ra) content and radon-222 (^{222}Rn) emanation coefficients,[1] may vary as a function of particle size. A knowledge of how these properties vary with particle size can also aid in modeling environmental impacts (for example, establishing emission-source terms in radon-flux models) and in designing mitigation measures (for example, requisite cover thicknesses for radon-flux control on sand and slime areas).

A longstanding truism of UMT management has been that the slimes (generally considered as the −200 mesh (<74 μ) fraction) have elevated contents of uranium daughter products compared to the sands (+200 mesh). However, studies by Guill (1978), summarized in the U.S. Nuclear Regulatory Commission's Final Generic Environmental Impact Statement on Uranium Milling (U.S. Nuclear Regulatory Commission, 1980) indicate that dry-sieved tailings may not show this pattern of increasing radionuclide content with decreasing particle size. The pattern observed by Guill is probably the result of aggregation of particles from either the cohesive properties of the slimes (Guill, 1978) or the cementing action of sulfate salts in acid-circuit UMT, carbonate salts in alkaline-circuit UMT, or a combination thereof in mixed or limed tailings. Methods of particle-size analysis that preserve such aggregates, and those that disrupt such aggregates, provide complementary approaches to evaluating particle-size relations in UMT. Dry-separation methods, including conventional sieving and air-elutriation techniques, enable one to examine mechanically stable aggregates that can persist under natural conditions and do not result in the loss of soluble constituents by leaching. Wet dispersion, followed by wet

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[1] The fraction of the radon produced that escapes from the solid matrix and thereby has the potential to reach the unconfined atmosphere.

sieving, sedimentation, or elutriation, enables one to isolate the discrete particles of which UMT are composed. In the modeling of the environmental transport of tailings particles, wind transport may best be evaluated by use of dry-separation size distributions, while with waterborne transport, the wet-separation size distribution should be considered.

The relation of the ^{226}Ra content and radon emanation coefficient (REC) to UMT particle size has received relatively little systematic investigation. The purpose of this study was to examine these relations, by use of both dry- and wet-separation techniques, in UMT derived from acid, alkaline, and mixed circuits.

Materials

Five uranium mill tailings samples were studied; four of these came from the Monticello, Utah, tailings site. Differences among these tailings reflect changes in milling practices and tailings disposal during the active life of that operation (1942–60). The following brief description of the Monticello operation is based largely on reports from the operating period, with extrapolation to the presently identified tailings area by the author.

The Monticello mill, constructed in 1942 and operated through 1944, used a salt roast-sodium carbonate leaching process to extract vanadium from Colorado Plataeu ores (Anonymous, 1944; U.S. Atomic Energy Commission, 1963). Some uranium was produced as a by-product (Butler, 1951), probably by leaching the salt roast tailings with sulfuric acid to produce a uranium-rich "green sludge" (Colorado Metal Mining Fund Board, 1961; Merritt, 1971). After a 5-year inactive period, a largely rebuilt mill opened in 1949 and operated through 1955; a salt-roast carbonate-leach circuit was used to recover uranium and vanadium (Butler, 1951; Merritt, 1971). The tailings from this period (1942–55) are designated the "carbonate" tailings at the site and by the acronym MCT in this report. In 1955, the salt roast and recovery of vanadium were abandoned and replaced by a direct carbonate-leach process for recovery of uranium only (Merritt, 1971). These mill tailings appear to be those called the "V_2O_5" tailings at the Monticello site and are designated MVT in this report.

Later in 1955, an acid-leach plant circuit (complementing the existing carbonate circuit) and new tailings pond were constructed (the "south" or "acid" tailings (MAT)). Despite the name, this pond apparently received both the alkaline- and acid-circuit tailings for a period of about a year (U.S. Atomic Energy Commission, 1963). Late in 1955, a new tailings pond (the "east" tailings (MET)) was built. Here, too, the tailings from the two circuits were combined, and additional lime was added to neutralize the waste stream (Whitman and Beverly, 1958). In 1958, the acid circuit was converted to a carbonate circuit, and discharge to the east tailings pond continued until the plant closed in 1960 (Merritt, 1971). During 1961–62, the 900,000 tons of tailings in the four disposal areas were recontoured and covered with rock and soil (U.S. Atomic Energy Commission, 1963).

In 1979, the U.S. Department of Energy did extensive drilling at the Monticello site and sampled each of the four tailings-disposal areas. Samples were dried at 110 °C and crushed with a cone and ring device set at −10 mesh, and then a composite sample from each tailings area was prepared (C.L. Bruner, Bendix Field Engineering Corp., personal commun., 1981). Portions of these composite samples were obtained for use in this study through the courtesy of the U.S. Department of Energy and Bendix Field Engineering Corporation, Grand Junction, Colo.

The fifth sample (CT1) was obtained from an active, acid-leach uranium mill in the Western United States. The tailings slurry was collected just prior to discharge to the pond, and the solids were collected by vacuum filtration in which Whatman no. 41 filter paper was used; the moist tailings were dried and crushed as described earlier.

Methods

For dry separations, 50-g (gram) aliquots of the bulk tailings samples were put through a nest of 20-cm (centimeter) in diameter stainless steel sieves that were mechanically shaken (500 excursions per minute, with intermittent tapping) for 30 minutes. The following size fractions were obtained (the numerical designations shown here are used in tables 9–14).

Size fraction	Mesh[1]	Size limits (in micrometers)
0	+10	>2,000
1	−10,+35	2,000–>500
2	−35,+60	500–>250
3	−60,+120	250–>125
4	−120,+200	125–>74
5	−200,+325	74–>44
6	−325	<44

[1] U.S. Standard series sieve designation.

Samples of dry fractions 0 through 5 were later dispersed in deionized water and wet sieved through a 325-mesh screen to determine the size distribution (>325 or <325 mesh) of the dispersed components of these dry fractions.

For wet separation, 15-g aliquots of bulk tailings samples were shaken overnight (180 excursions per minute) in 100 mL (milliliters) of deionized water in order to disrupt aggregates. These slurries were decanted onto a 325-mesh sieve. An electric sieve vibrator was used, and the material on the screen was washed with a fine stream of deionized water (approximately 375 mL per aliquot). The coarse

Table 9. Particle-size analysis (dry sieving) of uranium mill tailings

[MAT, MCT, MET, and MVT are the Monticello, Utah, "acid," "carbonate," "east," and "vanadium" tailings, respectively. CT1 are tailings from an active acid-leach uranium mill. These designations are used in tables 9–14]

Tailings	Dry-sieving size fraction (weight percent)[1]						
	0	1	2	3	4	5	6
MAT	1.8	20.4	10.9	14.9	16.1	24.8	11.0
MCT	5.5	12.0	12.0	26.3	20.9	17.4	5.9
MET	7.7	27.5	12.3	13.6	11.9	17.8	9.2
MVT	2.1	18.4	12.0	21.1	15.8	21.6	9.0
CT1	—	5.4	17.5	47.4	17.6	6.4	5.9

[1] Fractions 0 and 1 combined as the +10 mesh quantity are negligible.

Table 10. Quantity of fine particles (−325 mesh) in dry-sieved fraction as determined by subsequent wet dispersion and sieving

Tailings	Dry-sieving size fraction (weight percent)[1]					
	0	1	2	3	4	5
MAT	63	66	50	41	42	87
MCT	71	57	24	14	24	79
MET	69	65	46	35	36	82
MVT	69	70	53	29	32	80
CT1	—	17	11	11	18	63

[1] Weight percent −325 mesh following wet dispersion and sieving. Includes water-soluble material (<0.2 μm); see table 12 for total dissolved solids in water used to disperse and wet sieve the sample.

material (+325 mesh) on the screen was oven dried at 30 °C. The fine material (−325 mesh) passing the screen was allowed to settle overnight. The supernatant was then siphoned off and the sediment concentrated by high speed (10,000 revolutions per minute) centrifugation. In the case of slurry samples that did not clarify on standing overnight (MCT, MVT), the −325 mesh particles were collected by centrifuging (10,000 revolutions per minute) the entire slurry volume. These fine tailings solids were allowed to air dry, and the clods were hand crushed to pass a 120-mesh screen. The air-dried samples from the wet and dry separations had a mean (±1 standard deviation) moisture content (on 105 °C dry-weight basis) of 1.5 percent ±1.0 percent.

Samples of the siphoned and centrifuged supernatant from each tailings type were filtered (0.2-μm pore size); aliquots for ^{226}Ra determination were acidified prior to analysis by the radon emanation method (Yang, 1980). Radium-226 contents (expressed on vacuum dry-weight basis) and REC's of tailings were determined by Rogers and Associates Engineering Corporation (Salt Lake City, Utah) by use of a gamma-only assay (Thamer and others, 1981) on samples that were deemanated by evacuation at room temperatures.

Results And Discussion

Size Distribution

Table 9 shows the particle-size distributions of the five UMT samples determined by dry sieving. The +325 mesh fraction is about 90 percent of the total by weight. In contrast, table 10 shows that from about 10 to 90 percent of the particles in the +325 mesh-component fractions will pass through the 44-μm-diameter openings of the 325 mesh screen following wet dispersion and sieving. Thus, the +325 mesh dry-sieved fractions contain mechanically stable (persisting through 30 minutes of rapid oscillation) sand-sized (2,000 to >50 μm) aggregates composed of silt and clay (50 to >2 μm and ≤2 μm, respectively) primary particles. In the Monticello UMT, the highest proportion of these −325 mesh (wet-dispersed and sieved) particles occurs in the coarsest (#0, 1 fractions) and finest (#5) dry aggregates (about 60–90%), while the intermediate-size dry aggregates contain a lower proportion of −325 mesh particles (about 10–50%). Such a U-shaped size distribution may offer a partial explanation for the ^{226}Ra content distribution observed by Guill (1978) for dry-sieved acid-leached tailings; Guill observed a broad minimum in ^{226}Ra content around 500 μm and maxima around 100 and 900 μm.

The freshly produced tailings collected at the active, acid-leach uranium mill, CT1, show a differing pattern of aggregation (table 10), with only about 10 to 20 percent of the material in fractions #1–4 being −325 mesh after wet dispersion and sieving. The relative lack of aggregation in the CT1 tailings may be the result of an insufficient quantity of fine particles to provide cohesion. Acid-leach mills typically do not grind ores as finely as alkaline leach mills do. At the mill that produced the CT1 tailings, the ore was ground so that about 80 percent was −35 mesh. The exact grinding specifications used at the Monticello mills are not known and undoubtedly varied with ore type and leaching process. However, at a typical alkaline-leach mill, the ore is ground so that about 80 percent will be −100 mesh (International Atomic Energy Agency, 1980). Other possible reasons for the difference in aggregation pattern between the CT1 and Monticello UMT may include cementing materials unique to the acid-circuit-only CT1 tailings, differences in ores processed (for example, the presence of lignite particles in CT1), and possible diagenetic changes in the Monticello UMT during their 20 to 40 years of impoundment.

It is of interest to compare the known history of milling and tailings disposal at Monticello to the physical and chemical characteristics observed in the study. The

Table 11. Properties of wet-dispersed and wet-sieved uranium mill tailings

[Bq/g, becquerels per gram]

Tailings	Wet-sieved fraction[1]	Weight percent[2]	^{226}Ra (Bq/g)	Radon emanation coefficient (percent)[3]
MAT	+325	36.5	4.7	18(1)
	−325	55.6	41.9	24(1)
MCT	+325	57.6	8.8	16(1)
	−325	40.7	56.6	25(1)
MET	+325	41.1	3.6	13(2)
	−325	55.7	31.5	23(1)
MVT	+325	43.7	6.3	21(1)
	−325	54.6	24.5	33(1)
CT1	+325	80.1	6.1	12(1)
	−325	15.2	39.4	22(1)

[1] Soluble materials largely removed during wet processing.

[2] See table 12 for weight percent soluble material.

[3] The calculated uncertainty in the radon emanation coefficient based on 1 standard deviation counting statistic for counts before and after vacuum deemanation (Thamer and others, 1981) is shown in parentheses.

higher proportion, following wet sieving, of +325 mesh particles in the MCT tailings as compared to the other Monticello UMT (table 11) is a reflection of the finer grinding introduced in 1955 (U.S. Atomic Energy Commission, 1963) following conversion from the salt-roast/carbonate-leach circuit (MCT) to the direct carbonate leach circuit. The mixed nature (acid- and alkaline-circuit UMT) of the MAT and MET tailings is reflected in the pH and sulfate concentrations in the water used for dispersal and sieving, as is the alkaline character of the MCT and MVT tailings (table 12). Mixed tailings are not unique to the Monticello site but occur at several of the sites[2] being decommissioned under the Uranium Mill Tailings Remedial Action Project (UMTRAP) of the U.S. Department of Energy (Matthews, 1982).

Comparison of the ^{226}Ra distributions seen in tables 11 and 13 clearly shows the effects of aggregation of low-radium-content coarse particles with high-radium- content fine particles in the dry separates. It is also of interest to note that while about 1 to 6 percent by weight of the tailings are water-soluble materials removed during the wet dispersion and sieving process, only about 0.1 to 2 percent of the ^{226}Ra is thus removed (table 12). At much lower tailings-to-water ratios, wet separation can lead to major soluble losses of ^{226}Ra; studies by Skeaff (1977) involving cyclosizing of freshly collected acid-leach tailings showed the solubilization of about 30 percent of the ^{226}Ra at a tailings-to-water ratio of 0.21 g/L (grams per liter) (in contrast to the 31 g/L ratio used here).

Radon Emanation

With wet dispersion and sieving, it is apparent that for all of the UMT studied here, the fine fractions (−325 mesh) have higher radon emanation coefficients (REC's) than the coarse fractions (table 11). For dry-sieved samples (table 14), aggregation appears to mask any particle-size relation. The apparent lack of particle-size dependence of REC's noted in earlier studies (Macbeth and others, 1978) may, in part, result from such aggregation.

Previous studies have focused on UMT selectively collected from visually discernible sand and slime areas at tailings impoundments. Silker and Heasler (1979) examined dry beaches and dry berms at acid-circuit and alkaline-circuit mills in the vicinity of Grants, N. Mex., and made in situ, borehole measurements of REC's. The average REC for the beach and berm UMT at the alkaline-leach site were 43 and 40 percent, respectively, while for the acid-leach site they were 19 and 12 percent, respectively. The berms, by virtue of UMT disposal methods, are expected to contain the coarsest tailings. At the alkaline-leach site, a hydrocyclone separator was used to collect the coarse material for the berms while discharging the fines to the center of the tailings pond (beach). This method probably provided a relatively clean separation; thus, if it is assumed that any differences in the REC observed between the beach and berm are a function only of particle size, particle size evidently does not exert a major influence on REC's in these alkaline tailings. At the acid-circuit UMT site studied by Silker and Heasler (1979), the methods of berm construction (gravity flow of tailings from a discharge point at the outer edge of the impoundment and subsequent bulldozing of tailings from inside the pile to raise the berm height) would appear to result in a less well-defined particle size split between the beach and berm areas; hence, while the REC's of the materials from the two zones at this site are considerably different, it is difficult to unambiguously ascribe this effect to particle size. Using data of Rogers and others (1980) in 14 UMT samples, one obtains a linear correlation coefficient of +0.38 between the bulk sample REC (ranging from 6 to 31%) and the weight fraction of the sample passing a 200 mesh screen on wet sieving (ranging from 5 to 99%). While not statistically significant at the 0.1 level (Assaf and Assaf, 1974), it does suggest a weak association in which the REC increases as the proportion of fine particles in the UMT increases. Investigators at the Los Alamos Scientific Laboratory (Dreesen and others, 1981)

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[2] At Riverton, Wyo., a high-priority UMTRAP site (Matthews, 1982), both acid- and alkaline-circuit UMT were contemporaneously discharged to a common tailings pond (Ford, Bacon and Davis Utah, Inc., 1977); such UMT disposal also occurs in at least one presently licensed mill in the United States (U.S. Nuclear Regulatory Commission, 1979). At the Naturita and Durango, Colo., UMTRAP sites, sequential alkaline and acid treatments were used (Merritt, 1971), and liming of acid-circuit UMT, such as has been, or is, practiced at some mills in the United States (U.S. Department of the Interior, 1976; Ford, Bacon and Davis Utah, Inc., 1978) and elsewhere (LaRocque, 1978; Markose and others, 1982), creates still another form of this often-overlooked UMT class.

Table 12. Analysis of filtered water collected following aqueous dispersion and sieving of uranium mill tailings
[mg/L, milligrams per liter; Bq/L, becquerels per liter]

Tailings	pH	Sulfate (mg/L)	Total dissolved solids (mg/L)	Water-soluble material in tailings based on total dissolved solids (weight percent)	^{226}Ra (Bq/L)	Water-soluble ^{226}Ra as fraction of total ^{226}Ra in bulk tailings sample (percent)
MAT	7.0	1,300	1,880	6.0	1.63	0.19
MCT	9.2	91	193	.61	.78	.08
MET	7.8	540	875	2.8	3.26	.51
MVT	9.7	61	218	.69	.59	.11
CT1	4.0	920	1,370	4.3	5.92	1.6

Table 13. Distribution of radium-226 in uranium mill tailings as a function of particle size (dry sieving)
[The relative 1 standard deviation counting statistic for the values shown was 2 percent or less; all measurements are in becquerels per gram]

Tailings	Dry-sieving size fraction							Whole sample	
	0	1	2	3	4	5	6	By analysis	By summation of size fractions
MAT	19.0	22.1	22.6	20.2	20.3	31.1	40.0	26.6	23.6
MCT	10.7	25.3	22.7	17.9	27.0	54.4	67.0	31.1	30.1
MET	15.4	19.5	15.7	14.0	15.1	24.3	34.2	20.4	19.7
MVT	4.1	9.8	12.7	12.0	15.4	25.4	34.7	17.4	17.3
CT1	—[1]	9.0	8.5	7.7	10.9	25.5	32.7	11.5	11.0

[1] Fractions 0 and 1 combined, as the +10 mesh quantity are negligible.

Table 14. Radon emanation coefficient of particle-size fractions (dry sieving) of uranium mill tailings
[Values in percent; the calculated uncertainty in the radon emanation coefficient based on 1 standard deviation counting statistic for counts before and after vacuum deemanation (Thamer and others, 1981) is shown in parentheses]

Tailings	Dry sieving size fraction							Whole sample	
	0	1	2	3	4	5	6	By analysis	By summation of size fractions
MAT	4(2)	11(1)	20(1)	21(1)	19(1)	18(1)	19(1)	21(1)	17
MCT	22(1)	21(1)	21(1)	20(1)	19(1)	20(1)	20(1)	21(1)	18
MET	12(1)	15(1)	14(1)	16(1)	11(1)	13(1)	14(1)	14(1)	14
MVT	4(3)	29(1)	27(1)	27(1)	25(1)	26(1)	23(1)	27(1)	27
CT1	—[1]	7(1)	7(1)	9(1)	11(1)	12(1)	10(1)	9.4(0.7)	9.1

[1] Fractions 0 and 1 combined, as the +10 mesh quantity are negligible.

have studied "sands" (about 80 percent sand-sized particles) and "fines" (about 80 percent silt-sized particles) collected from select areas of the inactive UMT piles at Shiprock, N. Mex. (acid-leach site), and Durango, Colo. (mixed UMT). The Shiprock sands and fines have nearly identical RECs (10 and 11% , respectively), while the REC of the Durango sand was less than that of the fines (19 and 26, respectively). Ryon and others (1977) have investigated nitric acid leaching of uranium ores and mill tailings as a radium removal technique and have determined REC's on the +140 mesh (105 μm) and −140 mesh wet-sieved fractions of these nonconventional UMT (in which about 95 % of the radium, as opposed to about 1% for conventional UMT, is removed). In all cases, the REC of the +140 mesh fraction (5 to 36%, average 15%) was less than that for the −140 mesh fraction (17 to 57% , average 37%). Taken together, these studies suggest that where wet segregation of UMT has occurred, fines tend to have somewhat higher REC's than coarse tailings, a finding that is in general agreement with the findings of the present study.

The reason for the observed particle size to REC relationship is probably more than just a surface area effect. In some sedimentary rock samples, the REC has been shown (Andrews and Wood, 1972) to be proportional to the square root of the particle diameter. However, as demonstrated in several investigations with soils and rocks (Barretto, 1971; Andrews and Wood 1972; Megumi and Marumo, 1974), a direct correlation between radon emanating power and particle size (over a large range of particle sizes) can be expected only where the radium isotope precursors are uniformly distributed in the soil or rock. Such a distribution is not the norm in soils and rocks (Tanner, 1980), and certainly not in UMT, where redistribution of ^{226}Ra during both acid-circuit milling (Whitman and Beverly, 1958; Skeaff, 1981) and alkaline-circuit milling (Lakshmanan and Ashbrook, 1978), and inherited patterns from ore, tends to concentrate ^{226}Ra in the fine fraction of the UMT. Thus, the tacit assumptions that the higher REC of the fines is purely a function of their smaller particle dimensions and that the higher emanation coefficients generally observed for alkaline-circuit, as compared to acid-circuit, UMT are due only to their finer grinding, should be examined. Other factors, perhaps related to the chemical or mineralogical form of ^{226}Ra in the fine UMT (and in alkaline-circuit UMT), may also enhance their emanating power.

Dreesen and others (1981) have demonstrated reductions of 80 to 90 percent in the REC upon annealing UMT at temperatures around 900 °C. The mean REC determined by Austin and Droullard (1978) for the Colorado Plateau ores that were exploited during the operating era of the Monticello mill (Fischer, 1956) is about 20 percent. With these facts in mind, let us consider the REC's of the MCT tailings, whose production at the Monticello mill included an 840 to 910 °C (terminal temperature) salt roast (Anonymous, 1944; Philippone, 1955); these UMT have a bulk REC of 21 percent (table 14). Similar REC's are also observed for the salt-roast UMT at the Durango, Colo., mill, which also processed these Colorado Plateau ores (Dreesen and others, 1981). Thus, it would appear that for ore roasting conditions that include the presence of salt, followed by quenching and leaching, marked reductions in the REC of the tailings relative to the ore do not occur or do not persist. Studies by the U.S. Bureau of Mines (Austin and Droullard, 1979) on the effects of heating (without salt) on radon emanation from uranium ores (not subsequently put through a mill circuit) indicate that decreases in the REC induced by heat treatments may be partially restored upon aging at room temperature. Also, the redistribution of radium during milling noted above probably mitigates any initial reduction in the REC of the ore.

INFLUENCE OF ORE TYPE AND MILLING PROCESS ON RADON-222 EMANATION COEFFICIENTS OF URANIUM MILL TAILINGS

The chemical form of ^{226}Ra in precipitates (Hahn, 1936; Wahl 1951) and in natural geologic materials (Austin and Droullard, 1978) can influence the emanation of ^{222}Rn produced in such materials. Radon-222 emanation from uranium ores (Leach and others, 1982) and from UMT (U.S. Nuclear Regulatory Commission, 1980) is a radiological health concern. In making prospective radiological impact assessments of uranium milling operations, a fundamental question is how does the milling process influence the environmental mobility of radionuclides associated with the uranium ores being treated? Although the ^{226}Ra inventory in UMT solids generally accounts for about 98 percent of that in the parent ore, extensive solubilization and redistribution between particle-size fractions have been demonstrated to occur in both acid (Whitman and Beverly, 1958; Skeaff, 1981) and alkaline (Lakshmanan and Ashbrook, 1978) milling circuits. Such redistribution undoubtedly involves changes in the geochemical hosts of the ^{226}Ra in the ores versus the tailings (Landa, 1982); hence, changes in radon emanation characteristics may be anticipated. In the only previous study known to the author relating to this problem, workers at Oak Ridge National Laboratory (Ryon and others, 1977) sampled ore and tailings at three acid-leach and two alkaline-leach uranium mills in New Mexico, Utah, and Wyoming. In all cases, the REC's of the tailings at the acid-leach mills were less than those of the corresponding ore, while at the alkaline-leach mills, the REC's of the tailings were higher than those of the corresponding ore.

This section describes a laboratory study in which uranium ore samples of distinct mineralogies, rather than composite mill feeds, were subjected to acid and alkaline leaching for uranium extraction. In this way, both the ore type and extraction process serve as experimental variables, and the problems (Landa, 1982) in obtaining a matched set of ore and its resultant tailings at mills processing blended ores at rates of several hundred to several thousand tons per day are avoided.

Materials and Methods

The following mineral samples were obtained[3]: (1) autunite/meta-autunite in pegmatite, Ruggles Mines, near Grafton Center, N.H., (2) carnotite in sandstone, San Rafael Swell, Utah, and (3) uraninite in a matrix of iron and manganese carbonates, and quartz, Northwest Territory, Canada.

Samples were crushed and initially reduced to a nominal −28 mesh (590 μm) by ceramic plate grinder. A split of this material was ground to −100 mesh (149 μm), the finer particle size being required for effective alkaline leaching of uranium. Aliquots of about 300 g each were prepared by splitting the −28 mesh and −100 mesh materials. Two such aliquots of each grind size of each ore type were analyzed for ^{226}Ra content (expressed on vacuum-dry-weight basis) and the REC by Rogers and

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[3] Ward's Natural Science Establishment, Rochester, NY 14692.

Table 15. Characteristics of ore samples and leaching conditions used

[Values for uranium and vanadium in parts per million; all other values in percent]

	Ore type		
	Autunite	Carnotite	Uraninite
Uranium	18,200	857	27,000
Carbonate carbon	<.01	<.01	3.12
Aluminum	8.6	1.4	1.5
Iron	.66	.91	8.5
Manganese	.23	.0032	>10
Calcium	2.3	.02	.84
Phosphorus	2.0	.18	<.01
Vanadium	<4	170	<4
Leaching conditions (see footnotes for descriptions)	acid[1] alkaline[4]	acid[1] alkaline[3]	acid[2] alkaline[4]

[1] 0.5 M H_2SO_4; 2 hours in autoclave (122 °C).

[2] 1.25 M H_2SO_4; 4 hours at room temperature to initiate carbonate digestion, then overnight (16 hours) in 80 °C oven, and finally 2 hours in autoclave.

[3] 0.5 M Na_2CO_3 + 4.2 g $NaHCO_3$ per 100 mL; 4 hours in autoclave.

[4] 0.5 M Na_2CO_3 + 4.2 g $NaHCO_3$ per 100 mL; 9 days in 80 °C oven; deionized water added to bottles every 2–3 days to adjust to initial volume.

Associates Engineering Corporation (Salt Lake City, Utah) by use of a gamma-only assay on dry samples that were de-emanated by evacuation at room temperature (Thamer and others, 1981). These same aliquots were then leached as described here.

Simple laboratory-scale uranium extraction procedures using only air oxidation were developed to approximate commercial practices (Merritt, 1971). Acid leaching was done on the −28 mesh ground material, while alkaline leaching was done on the −100 mesh ground material. Leaching was done in loosely capped polypropylene bottles containing 30 g of ore, 100 mL of either sulfuric acid (H_2SO_4) or sodium carbonate (Na_2CO_3) solution, and, in the case of the alkaline leaching, 4.2 g of sodium bicarbonate ($NaHCO_3$). Bottles were placed in either an autoclave operated at 122 °C and 1.27 kg/cm^2 (kilograms per square centimeter) or a conventional oven operated at 80 °C. Solution concentrations and leaching conditions are given in table 15. For acid leaching of the autunite and carnotite samples, 0.5 M H_2SO_4 was used. For the uraninite sample, a modified procedure was necessary to compensate for acid consumption by carbonates. In the alkaline leaching case, the carnotite ore was amendable to a 4-hour autoclave procedure, but uranium extraction from the other two ores was only about 60 percent under these conditions. Experimentation showed that the extraction could be increased to about 80 percent by treatment for about a week at 80 °C in a conventional oven, and such a procedure was adopted. Immediately following the heating/leaching period, the warm slurries were vacuum filtered by use of a Buchner funnel and highly retentive, hardened filter paper (Whatman no. 50). The collected tailings (1 bottle per filter) were filter washed with about 75 mL of deionized water, allowed to air dry, and lightly crushed by hand. These samples (mean moisture content = 0.09% ±0.04% (1 standard deviation)) were then analyzed for uranium and ^{226}Ra content and for the REC.

Uranium content of ore and tailings was detemined by the delayed neutron method (Millard and Keaton, 1982), and carbonate content of ore was determined by the acid-neutralization method (Allison and Moodie, 1965). Other constituents were determined by induction-coupled argon plasma spectrometric analysis (Taggart and others, 1982). Particle-size distribution (±325 mesh (44 μm)) of ground ore and tailings samples was determined by wet dispersion and sieving.

Results and Discussion

A comparison of the uranium contents of the ores (table 15) and the tailings (table 16) shows removal of from 78 to 99 percent of the uranium by the milling treatments used. The agreement between the REC's measured (table 16) for the −28 mesh and the −100 mesh grind-size samples of each ore type (for example, −28 mesh autunite versus −100 mesh autunite) suggests that finer grinding of ores (table 17) is not necessarily associated with higher REC's. These findings are in general agreement with those of Austin and Droullard (1978), who compared emanation coefficients of whole rock versus disaggregated samples, although they did observe significant increases (34 to 227 percent) in the REC's of some samples after going from whole rocks to −100 mesh (nominal) pulps. In the case of UMT, the finer grinding of alkaline-leach tailings has been suggested as the reason for the higher REC's reported, compared to the coarser ground, acid-leach tailings (U.S. Nuclear Regulatory Commission, 1980). While such a relationship may be an indirect result of the greater available surface area on which redistributed ^{226}Ra can take up residence, it appears unlikely that the effect is purely physical and due to comminution of the ore.

In the case of acid leaching, the REC of the autunite-derived tailings is about the same as that of the ore, whereas for the carnotite- and uraninite-derived tailings, the REC's are considerably lower than those of the respective ores. In the case of alkaline leaching, the REC of the autunite-derived tailings increased, while that of the carnotite-derived tailings decreased considerably with respect to the ores. The REC's of the alkaline-leach, uraninite-derived tailings are about the same as those of the ore. Tailings from the acid treatment of the uraninite ore showed a decrease in REC despite an increase in the −325 mesh size fraction from about 13 weight percent in the ore to 22 weight percent in the tailings (table 17). This shift toward finer grained material was associated with the digestion of carbonate and

Table 16. Radium-226 content and the radon-222 emanation coefficient (REC) of ore and tailings, and the yield and uranium content of tailings

[Bq/g, becquerels per gram; %, percent; ppm, parts per million]

Ore type	Sample mesh/split no.	Ore		Acid-leach tailings				Alkaline leach tailings			
		^{226}Ra[1] (Bq/g)	REC[1] (%)	Yield[2]	Uranium (ppm)	^{226}Ra (Bq/g)	REC (%)	Yield[1]	Uranium (ppm)	^{226}Ra (Bqt/g)	REC (%)
Autunite	−28/1	214±0.3	4±1	94.6	2,840	183±1.1	5±1	—	—	—	—
	−28/2	216±0.1	2±1	94.1	3,990	184±1.1	2±1	—	—	—	—
	−100/3	212±0.1	4±1	—	—	—	—	96.9	3,450	176±1.1	12±1
	−100/4	209±0.1	4±1	—	—	—	—	96.8	3,130	173±1.1	12±1
Carnotite	−28/1	9.4±0.04	12±1	99.0	10.5	9.9±.1	4±2	—	—	—	—
	−28/2	10.2±0.04	17±1	99.1	7.9	10.0±.2	7±3	—	—	—	—
	−100/3	9.8±0.04	18±1	—	—	—	—	99.0	15.4	8.9±.1	6±2
	−100/4	9.8±0.04	19±1	—	—	—	—	99.3	13.5	8.3±.1	4±3
Uraninite	−28/1	302±0.2	12±1	63.1	1,160	310±1.5	7±1	—	—	—	—
	−28/2	322±0.3	14±1	63.2	1,310	329±1.8	5±1	—	—	—	—
	−100/3	309±0.2	13±1	—	—	—	—	96.6	6,290	229±1.5	12±1
	−100/4	311±0.2	15±1	—	—	—	—	96.7	5,990	221±1.1	11±1

[1] Uncertainty shown based on 1 standard deviation counting statistic.

[2] $\text{Percent yield} = \frac{\text{g tailings solids}}{\text{g ore}} \times 100.$

other acid-soluble phases in the ore. For the other ore-tailings pairs, the acid or alkaline leaching did not influence the −325 mesh size distribution.

No consistent pattern enabling the prediction of the REC's of tailings on the basis of ore type emerges from the analysis of the data presented in table 16. It is apparent that the REC of the tailings cannot be assumed, a priori, to be the same as that of the ore from which it is derived. In the acid-leach case, there is a clustering of tailings' REC's around a value of about 5 percent, despite starting ores of differing uranium mineralogies and rock types. Such behavior might possibly be expected if all the ^{226}Ra present in the ore were released to solution and subsequently precipitated or retained by some other mechanism in a specific chemical/mineralogical form in the tailings dictated largely by the lixiviant solution. Nirdosh and others (1984) have discussed in detail the various mechanisms by which ^{226}Ra can be retained in UMT (for example, coprecipitation, surface adsorption on amorphous silica and hydrated metal oxides, ion exchange on clay minerals). The emanating power of these retained forms can vary widely at room temperature; for example, while ^{226}Ra coprecipitated with barium sulfate generally emanates less than 1 percent of the ^{222}Rn generated, that coprecipitated with or sorbed on hydrous iron oxide can emanate nearly 100 percent of the ^{222}Rn produced (Hahn, 1936). While REC's of dry samples are reported here, increasing moisture content can be expected to enhance radon emanation from both ores and tailings (Thamer and others, 1981).

Removals of ^{226}Ra from the ore by the leaching processes (table 18) are generally higher than the 1 to 2 percent commonly reported on the basis of in-mill sampling

Table 17. Particle-size distribution of ore and tailings samples as determined by wet sieving

Sample	Weight percent −325 mesh	
	Ore	Tailings
28-mesh ground, acid-leach tailings		
Autunite	4.9	5.4
Carnotite	6.5	6.3
Uraninite	12.8	22.2
100-mesh ground, alkaline-leach tailings		
Autunite	12.9	14.4
Carnotite	13.8	13.5
Uraninite	27.6	25.9

Table 18. Percentage of radium-226 (initially in ore) found in tailings solids (adjusted for tailings yield)

[Value shown is mean of duplicate samples]

Ore type	Acid-leach tailings (percent)	Alkaline-leach tailings (percent)
Autunite	80.5	80.3
Carnotite	100.9	87.1
Uraninite	64.6	70.2

(Moffett, 1976). One can only speculate on the reasons for this difference (for example, variance from actual mill leaching practices); however, it is interesting to note that under certain milling conditions, considerable quantities of ^{226}Ra can enter the liquid effluent waste stream.[4]

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[4] Skeaff (1981) presents an excellent discussion showing that the quantity of ^{226}Ra measured in solution at various stages of an acid-leach uranium mill circuit was orders of magnitude lower than the limits imposed by the solubility of $RaSO_4$ in sulfuric acid (75 °C). Recently, Torma and others (1985) have reported extraction of about 40 percent of the ^{226}Ra from uranium ore leached with sodium carbonate-bicarbonate in laboratory tests.

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PART 1: STUDIES OF RADON IN ROCKS AND SOIL

SECTION 3: ESTIMATING RADON POTENTIAL

USE OF AERIAL GAMMA-RAY DATA TO ESTIMATE RELATIVE AMOUNTS OF RADON IN SOIL GAS

Joseph S. Duval

Abstract

Because aerial gamma-ray measurements of equivalent uranium are an indirect measure of radium-226 and a direct measure of bismuth-214, which is a daughter isotope separated from radon-222 by short-lived isotopes of polonium and lead, gamma-ray data can be used to estimate the relative amounts of radon in soil gas. Theoretical calculations are available that enable an interpreter to define the limitations on the coverage provided by an aerial survey. Aerial survey data for the State of New Jersey are compared to indoor radon measurements. The results of this comparison support the use of aerial gamma-ray data to estimate the relative concentrations of radon in soil gas, and published data comparing radon concentrations in soil gas to surface gamma-ray measurements provide corroborating evidence. The application of Swedish criteria for assessing radon hazards to the data for New Jersey suggests that hazard-assessment criteria from one area may not be correct for other, geologically different areas.

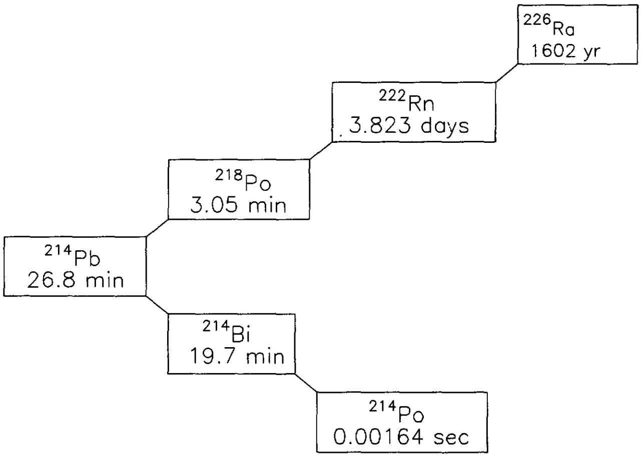

Figure 54. Part of the radioactive decay series of uranium-238 showing radium-226 (^{226}Ra) and daughter nuclides including radon-222 (^{222}Rn); ^{218}Po, polonium-218; ^{214}Pb, lead-214; ^{214}Bi, bismuth-214; ^{214}Po, polonium-214.

INTRODUCTION

In 1985, a house located within the geological province known as the Reading Prong was discovered to have indoor radon-222 (^{222}Rn) concentrations exceeding 2,000 pCi/L (picocuries per liter), and other houses having elevated indoor radon concentrations were quickly identified in the same neighborhood (Gaertner, 1987). The State of Pennsylvania immediately began a large program to determine the extent of the problem by distributing radon detection kits to homeowners located within the Reading Prong (Gerusky, 1987). The results of this program showed that more than 50 percent of the houses measured had indoor radon concentrations exceeding the 4.0 pCi/L guideline set by the Environmental Protection Agency. Data from other parts of the Nation soon made it obvious that the Reading Prong is not the only place that has an indoor radon hazard. The possibly widespread nature of the indoor radon hazard made efficient identification of those parts of the nation that have the greatest potential for indoor radon problems a primary concern. Because aerial gamma-ray measurements provide an estimate of the surface concentrations of radium-226 (^{226}Ra), which is a precursor of radon, the aerial gamma-ray surveys conducted as part of the U.S. Department of Energy National Uranium Resource Evaluation (NURE) Program can be used to make maps showing the distribution of ^{226}Ra. These maps can be used to estimate relative concentrations of radon in soil gas. This report presents an example for the State of New Jersey that illustrates the utility of such maps.

AERIAL GAMMA-RAY SURVEYS AND RADON

Aerial gamma-ray surveys are generally used to estimate the apparent surface concentration of uranium by measuring the flux of 1.76-MeV (million electron volt) gamma rays produced by the decay of bismuth-214 (^{214}Bi). The survey data can also be expressed as apparent concentrations of ^{226}Ra, which is the last long-lived (1,602 yr) member of the uranium-238 (^{238}U) radioactive decay series. Because ^{214}Bi is directly linked to ^{222}Rn (hereafter designated simply as radon) by short-lived isotopes and because ^{222}Rn is the direct daughter of ^{226}Ra (fig. 54), the gamma-ray data provide an estimate of the near-surface concentration of radon. Because the radium is located inside mineral grains as well as in soil pore spaces and on grain surfaces, some fraction of the total radon is locked inside the soil materials and is unable to diffuse into the soil gas. The emanation power is defined as the fraction of the total radon produced that escapes into the soil gas and is largely controlled by the distribution of radium within the rocks and soils (Tanner, 1964). The relative concentrations of radon in the near-surface materials can, however, be used as a first-order estimate of the relative radon concentrations in the soil gas. Other factors that affect the radon concentrations in the soil gas are the soil moisture content and the soil permeability. Soil moisture can, to some extent, increase the emanation power of a material (Hahn, 1936; Starik and Melikova, 1957), but high soil moisture causes the effective emanation power (radon availability) to approach zero because the water fills the soil pores, replaces the soil gas, and inhibits diffusion of the radon (Morozova and Mukhraneli, 1971). Soils or rocks having high gas permeability are affected by convective processes that reduce the radon concentrations in the top 1 to 2 m (meters) and allow the radon to move distances of 5 m or more (Tanner, 1964, 1980; Schery and others, 1984).

The NURE aerial gamma-ray surveys were flown at a nominal altitude of 122 m above the ground, a height that provides a measurement path approximately 250 m wide beneath the flight line (Pitkin and Duval, 1980). The gamma-ray signals from the ground are attenuated by the ground itself and the air. The effective thickness of material measured is about 30 cm (Duval and others, 1971). The flight-line spacings of the NURE surveys range from 1.6 to 10 km, with most of them having either 5- or 10-km intervals. These flight-line spacings (5–10 km) result in a total ground coverage of less than 10 percent (Pitkin and Duval, 1980); thus, maps made from these data are generally not suitable for site-specific studies. The data along the flight lines could, however, be used to provide site-specific information.

DISTRIBUTION OF RADIUM IN NEW JERSEY

Figure 55 shows the NURE aerial gamma-ray surveys that were computer processed and compiled to make the map of the distribution of radium in New Jersey shown in figure 56. The NURE flight-line data were first inspected, and all data measured at ground clearances greater than 200 m were rejected as invalid because of the low signal-to-noise ratio caused by air attenuation of the gamma-ray flux. The data were then filtered by use of a 3-point median filter as a spike rejection technique followed by a 17-point Gaussian filter to smooth out statistical fluctuations in the data. The data were then gridded by use of a minimum curvature algorithm (Briggs, 1974; Webring, 1981) and a grid interval of 3.2 km. The gridded data were examined as a gray-scale image by use of image processing techniques, and some of the data sets were found to have relative level errors. The term "relative level error" is used here to describe the fact that data sets from different NURE contractors frequently exhibit obvious disagreements when the data are merged and portrayed in an image format.

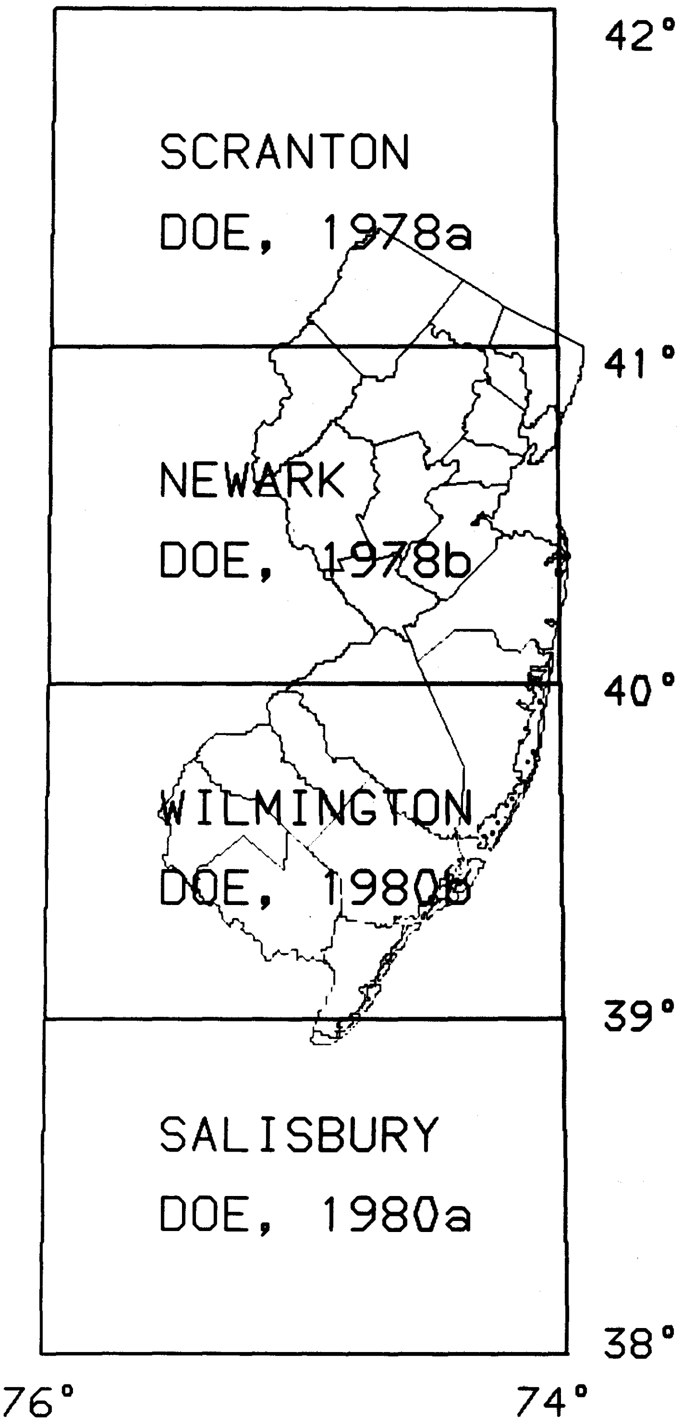

Figure 55. Index map showing the NURE aerial gamma-ray surveys used to compile the data for New Jersey. DOE, U.S. Department of Energy.

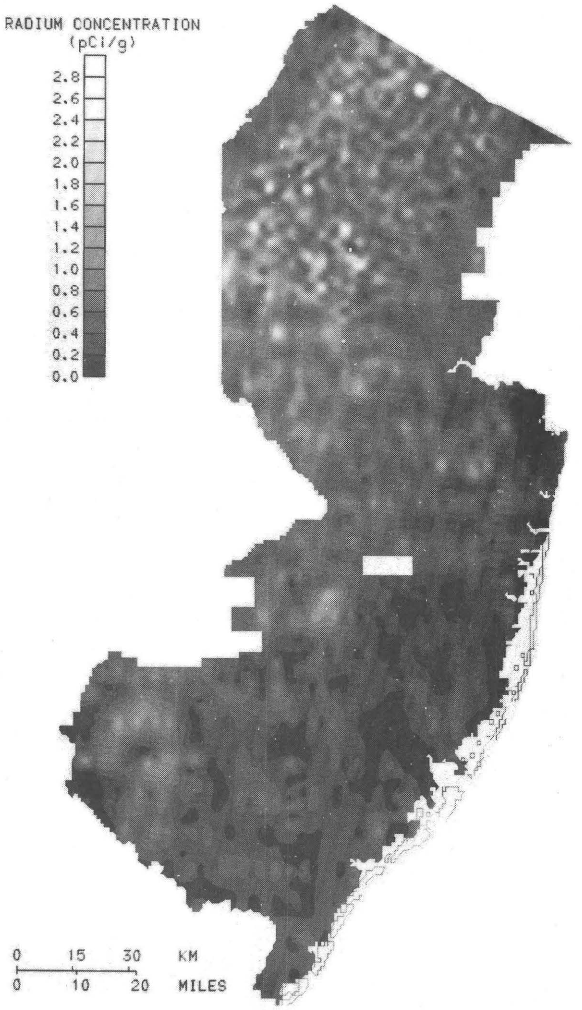

Figure 56. Contour map of the near-surface concentrations of radium in New Jersey. pCi/g, picocuries per gram.

Because these data had been corrected for background by use of on-board measurement techniques, the level differences were treated as scaling errors caused by calibration errors or soil moisture differences, and the data were multiplied by a constant.

Because the NURE data sets do frequently show disagreements, multiple data sets should be processed simultaneously; for this example, data outside the quadrangles shown in figure 55 were included in the data compilation. The resulting image was used to determine that all of the data sets covering New Jersey needed to be multiplied by a factor of 0.9 to achieve agreement with the adjacent data sets. The determination of this value was based upon a subjective examination of the gray-scale image. Experience with this approach has shown that relative level differences on the order of 5 percent can be seen in the image.

INDOOR RADON IN NEW JERSEY

In trying to utilize the aerial gamma-ray data, one must determine the appropriate scale at which to make comparisons and predictions. Because of convenience and ease of representation, one is tempted to use broad regional categories such as geologic provinces or physiographic regions. Political boundaries are also viewed as convenient for summarizing data. To some extent the use of physiographic provinces, county boundaries, and localized sites has been dictated by the availability of radon data.

In September of 1987, the New Jersey Department of Environmental Protection (NJDEP) released data showing the regional distribution of indoor radon concentrations in New Jersey (NJDEP, 1987). The data consisted of more than 5,000 measurements of indoor radon concentrations. The sample locations are distributed nonuniformly across the State, and the data are summarized by physiographic provinces in figure 57. The average radon values were taken from the NJDEP press release, and the average radium values were calculated from the data of figure 56. The boundaries of the physiographic provinces were taken from the "Geologic Map of New Jersey" (Lewis and Kummel, 1912) and were defined as follows: "outer Coastal Plain" corresponds to the Tertiary rocks, "inner Coastal Plain" contains the Cretaceous rocks, "Triassic basins" includes the Triassic rocks, "Reading Prong" includes the Precambrian crystalline terrane, and the "Valley and Ridge" province includes the Devonian, Silurian, and Ordovician rocks. An examination of the information presented in figure 57 shows that the average indoor radon level does tend to increase as the average surface radium concentration increases, but the relationship is not consistent. Part of the reason for this apparent inconsistency is the fact that the radium concentrations within the physiographic provinces are not uniform. Figure 58 presents the frequency histogram of radium concentrations within the Valley and Ridge province and shows a bimodal distribution. Although the distribution functions of the other provinces are not obviously bimodal, they are similarly skewed to a greater or lesser extent. This skewing suggests that the comparison should be made by using smaller geographical areas. Because the NJDEP press release included a summary by counties (table 19), average radium values were calculated for the counties with available data. Figure 59 shows the average radium concentrations versus the average indoor radon concentrations for all of the counties in New Jersey except Hudson County, for which the aerial gamma-ray data are not available. These data exhibit an approximately linear trend up to radium concentrations on the order of 1 pCi/g (picocurie per gram).

DISCUSSION

Although the comparisons presented above support the use of aerial gamma-ray data to estimate relative indoor

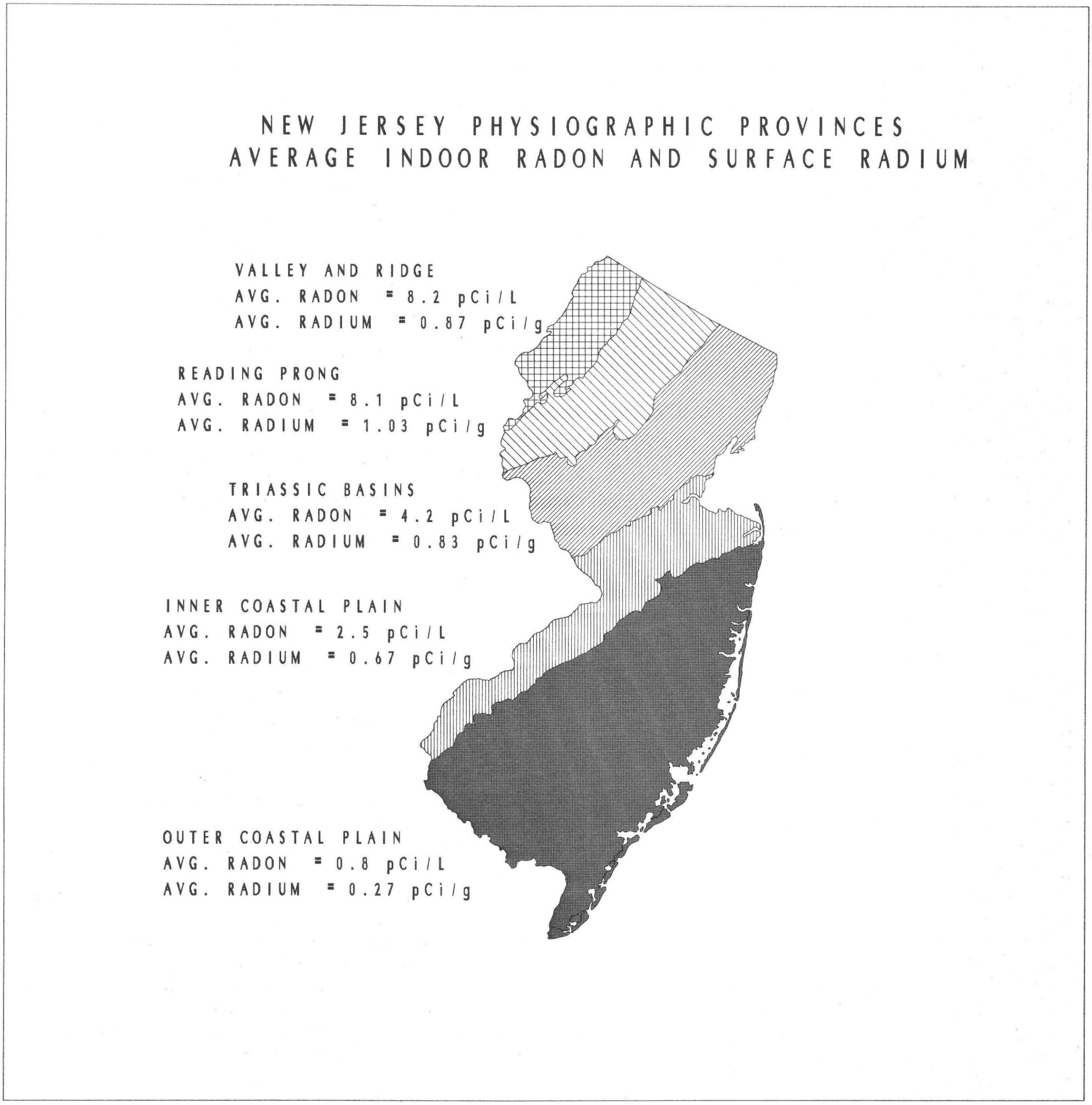

Figure 57. Summary of average indoor radon and surface radium concentrations for physiographic provinces in New Jersey. The labels correlate with the map patterns from north to south. pCi/L, picocuries per liter; pCi/g, picocuries per gram.

radon potential of large areas such as geologic provinces or counties, the data are subject to the following criticism: (1) the use of areas defined by political or other arbitrary boundaries results in the lumping of diverse geologic materials with significantly different radium concentrations and reduces the validity of the comparison and (2) indoor radon measurements include variance that is a function of the building construction, which is largely independent of the geologic environment. The use of indoor radon data can be eliminated by making comparisons with measurements of radon concentrations in soil gas. Gunderson and others (1988) have made measurements of radon concentrations in soil gas as well as surface radium concentrations for different geologic materials in Montgomery County, Md.,

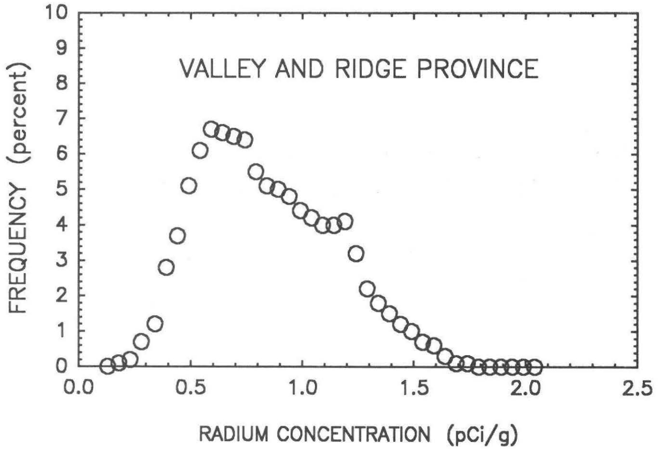

Figure 58. Frequency histogram of radium concentrations within the Valley and Ridge province. pCi/g, picocuries per gram.

Table 19. Average indoor radon levels for counties in New Jersey

[pCi/L, picocuries per liter]

County	Average indoor radon (pCi/L)	County	Average indoor radon (pCi/L)
Atlantic	0.75	Middlesex	2.16
Bergen........	1.85	Monmouth	2.68
Burlington.....	1.80	Morris	5.13
Camden.......	2.27	Ocean	.95
Cape May.....	1.43	Passaic	3.67
Cumberland ...	1.79	Salem	2.49
Essex.........	1.23	Somerset......	5.20
Gloucester.....	3.12	Sussex........	6.47
Hudson	2.47	Union	2.32
Hunterdon.....	6.88	Warren	11.83
Mercer........	4.46		

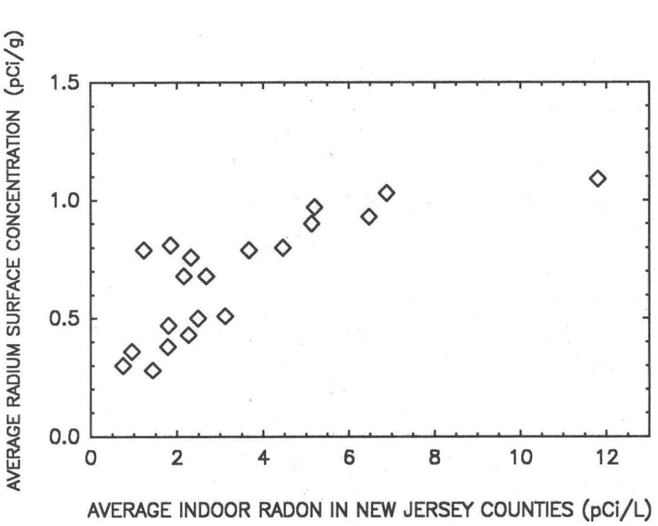

Figure 59. Average radium concentration versus the average indoor concentrations in counties in New Jersey. pCi/L, picocuries per liter; pCi/g, picocuries per gram.

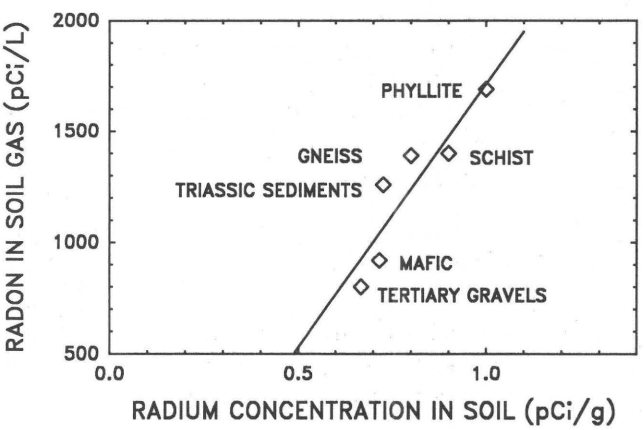

Figure 60. Average radon concentrations in soil gas versus average surface radium concentrations for various geologic materials in Montgomery County, Md. (adapted from Gunderson and others, 1988). The straight line is a linear least-squares fit to the data. pCi/L, picocuries per liter; pCi/g, picocuries per gram.

and figure 60 presents those data with a calculated linear fit. The linear fit results in the equation

$$\mathrm{Rn} = -654.2 + 2367.5\ \mathrm{Ra}. \qquad (1)$$

The use of a linear equation to fit the data was predicated by the following: (1) the available data are few in number and do not cover a large range of values and (2) a linear dependency is the simplest case. Because radium is the direct parent of radon, it is possible to calculate the total amount of radon produced by the decay of radium, but not all of that radon escapes into the soil gas. For that reason an equation of the general form given by equation 1 should apply to any geologic material. There is, however, no reason to expect that the same equation will apply to various geologic materials unless the radon emanation powers of the different materials are approximately the same. The fact that the linear equation reasonably represents the data suggests that the general geologic and geochemical conditions of the measured geologic units have resulted in near-surface radium distributions that produce approximately equal radon emanation powers. Because the equation provides a predictive capability, these data provide corroboration and additional support for the use of surface gamma-ray measurements to predict relative amounts of radon in soil gas.

RADON HAZARD ASSESSMENT

Radon hazard assessment in the United States is not yet well developed. Recent efforts by Gunderson and others (1988) and Otton and others (1988) combine radon soil-gas

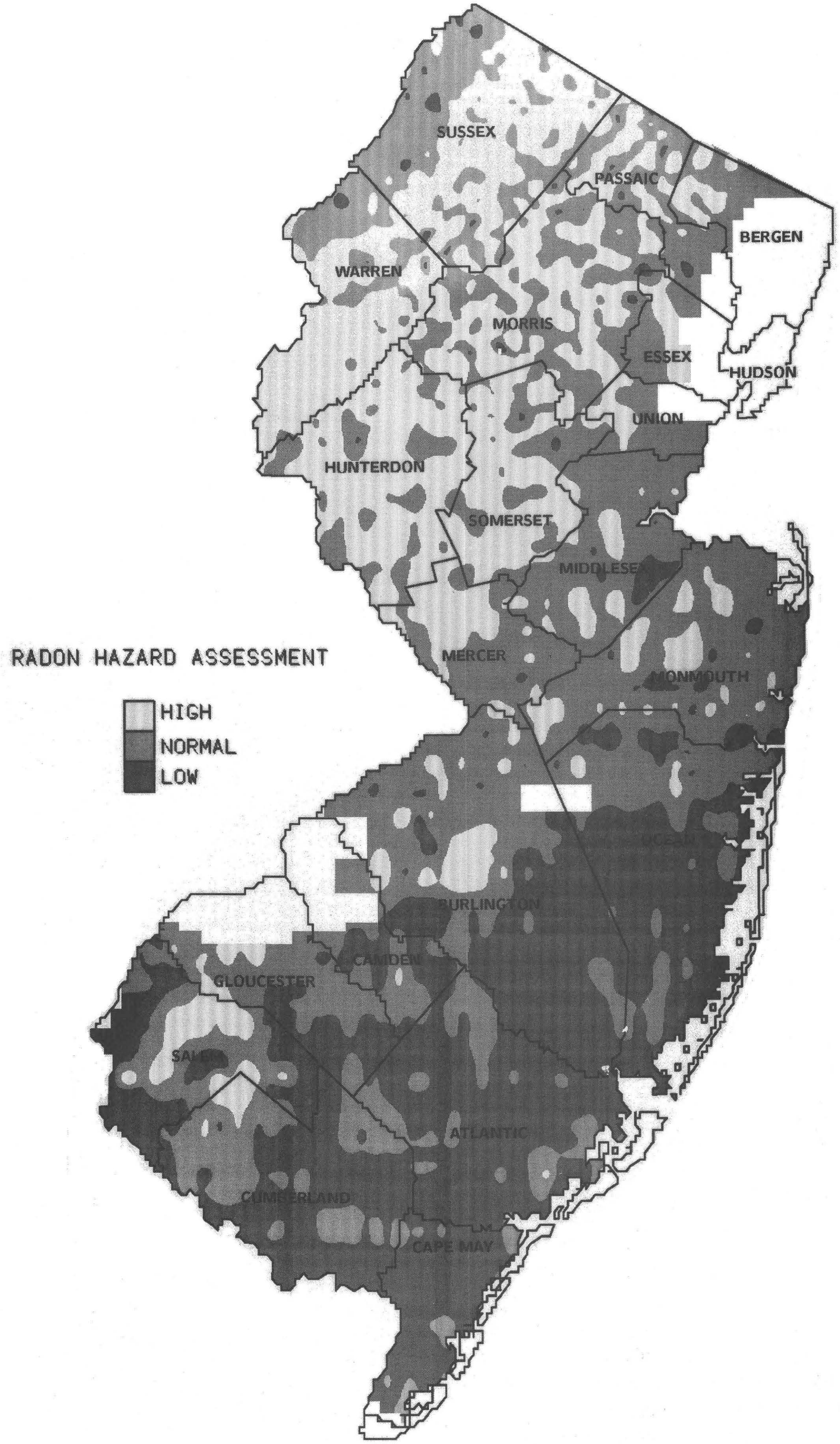

Figure 61. Radon assessment map based upon the Swedish criteria, the data of figure 60, and figure 56.

data, surface gamma-ray measurements, and geologic data to define a relative ranking within the areas studied. These studies do not provide any specific rules for assessing radon hazards. Sweden has established criteria for radon hazard assessment. These criteria consider that concentrations of radon in soil gas less than 270 pCi/L are "low" and require no special construction, the range 270–1,350 pCi/L is classified as "normal" and requires "radon protective" construction, and values greater than 1,350 pCi/L are "high" and require "radon safe" construction (Åkerblom, 1986; Wilson, 1987). The natural tendency is to try to apply these same criteria to other geologic environments. The validity of applying the Swedish criteria to New Jersey can be tested if we make some assumptions about the radon in the soil gas.

If the soil-gas data from Montgomery County, Md., are assumed to apply to New Jersey, the radium data of figure 56 and the Swedish criteria can be used to define a radon assessment map as shown in figure 61. The validity of this map can be tested by comparing it to the data for the New Jersey counties listed in table 19. In figure 61, Atlantic and Cape May Counties are largely classified as "normal," and application of the Swedish criteria would not require any special new home construction methods. The indoor radon data of table 19 suggest that this classification is correct. Salem, Gloucester, Monmouth, and Middlesex Counties are largely classified as "normal" to "high," and application of these criteria would require most new home construction in these counties to be either "radon protective" or "radon safe." The data in table 19 do not support this classification. Burlington and Ocean Counties have over half of their area classified as "low" with the remainder being mostly "normal." The data in table 19 suggest that much more than half of these counties should be classified as "low." Warren, Hunterdon, Morris, Sussex, Passaic, and Somerset Counties are largely classified as "high," and the Swedish criteria would require that most new home construction be "radon safe." The data in table 19 suggest that a significant fraction of the areas classified as "high" probably should be so classified, but it is not clear how much of the "high" area is correctly classified.

The radon assessment presented in figure 61 is not consistent with the available indoor radon data. This inconsistency must be at least partly caused by the use of criteria that are based upon only one of the factors that control radon availability. This exercise in radon hazard assessment is useful because it points out that the use of inadequate criteria can result in requirements for potentially expensive construction techniques in areas where they may not be needed. The Swedish scientists did include geologic data in their work and stated that permeability and rock type must be considered, but the criteria given herein are based solely upon the radon concentrations in the soil gas.

Tanner (1988) discusses a technique that provides a measure of the amount of radon available for entry into a building. This measurement of "radon availability" includes the emanation power, soil permeability, and soil moisture effects in a single measurement and would be a better basis for hazard assessments.

CONCLUSIONS

The data and comparisons presented in this paper support the following conclusions: (1) although absolute estimates of radon in soil gas may not be possible, aerial gamma-ray measurements of near-surface concentrations of radium can be used to estimate relative levels of radon in soil gas, (2) aerial gamma-ray data are positively correlated with indoor radon levels, but the use of political or physiographic boundaries for data summaries may result in significant errors because of geologic variations within those boundaries, (3) the application of radon hazard assessment criteria from one area to another will not be valid unless both areas have similar climatic, geologic, and geochemical histories that produce similar radium distributions, emanation powers, soil moisture, and soil permeability, (4) simplistic radon hazard criteria should not be used, and any criteria that are developed should include the radon concentration in the soil gas, soil permeability, and soil moisture, and (5) aerial or ground gamma-ray data should be used only to estimate the relative amounts of radon in soil gas until more data are available to better test the calculation of actual radon concentrations. The data needed to establish whether surface gamma-ray measurements can be used to estimate absolute amounts of radon in soil gas are similar to the data collected by Gunderson and others (1988). Because the emanation power of geologic materials depends upon the geologic history of the lithologic units, data need to be gathered for a variety of geologic and climatic environments. In the collection of these data it is critical that the surface gamma-ray and soil gas measurements be made simultaneously and that various soil gas measurements be made at a single site so as to reduce the variance of the measurements.

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REGIONAL RADON CHARACTERIZATIONS

R. Thomas Peake[1] and R. Randall Schumann[2]

Abstract

Indoor radon is a potential health threat in a significant number of homes across the United States. Twenty-one percent of the homes tested in the 1986–87 Environmental Protection Agency/State Indoor Radon Survey of 10 States contained screening indoor radon levels greater than the Environmental Protection Agency's recommended action level of 4 picocuries per liter. However, the distribution of indoor radon screening levels, and, therefore, the potential health risk, is not uniform everywhere. These differences are related to geology and house construction.

Characterizations of radon potential can be made by use of geologic, soil, and radiometric data. National Uranium Resource Evaluation equivalent uranium concentrations of 2.0 parts per million or greater indicate areas that have the potential to produce a substantial number of elevated indoor radon levels. Equivalent uranium concentrations less than 1 part per million may indicate low radon potential. Soil permeability greater than 6.0 inches per hour may increase the radon potential of an area, except in cases where soil radium concentrations are very low.

INTRODUCTION

Almost every State has some homes that contain radon levels greater than 4 pCi/L (picocuries per liter) (Alter and Oswald, 1987). Nero and others (1986) estimated that an annual indoor radon average for the United States is 1.5 pCi/L and that 7 percent of the homes in the United States, or approximately 5 million single family homes, may have annual averages above 4 pCi/L. These may be underestimates because the Nero distribution did not account for the large number of radon concentrations that have been found above 20 pCi/L (Napolitano and others, 1988). Twenty-one percent of the homes sampled in 10 States during the 1986–87 Environmental Protection Agency (EPA)/State Indoor Radon Survey were found to contain screening levels above 4 pCi/L (Ronca-Battista and others, 1988). A screening measurement is defined as a short-term (2–3 day) radon measurement made in the lowest livable area in the the house (EPA, 1986). By definition, therefore, screening measurements are made in basements or on the first floor of homes that do not have basements.

The distribution of indoor radon levels is not uniform across the country. On a regional scale, the percentage of homes having screening levels above 4 pCi/L is an indicator of the differing radon potential. In the 1986–87 EPA/State Indoor Radon Survey, fewer than 10 percent of the homes sampled in Alabama and Michigan were found to contain screening radon levels above 4 pCi/L. In contrast, 26 percent of the homes sampled in Wyoming were found to have screening indoor radon concentrations greater than 4 pCi/L. These differences may be related to regional geology and differences in house construction characteristics.

Preliminary radon characterizations can aid in the planning of regional indoor radon testing programs or in allocating resources to those areas that have the most severe radon problem. This paper describes the concepts and methods used for preliminary regional radon characterizations and discusses the results of comparisons between geologic and indoor radon data. The views presented in this paper do not necessarily reflect formal EPA policy.

FACTORS CONTROLLING RADON CONCENTRATIONS IN SOILS

Radon-222 (^{222}Rn) is produced from the radioactive decay of radium-226 (^{226}Ra). Other isotopes of radon occur naturally, but they are of lesser importance in terms of indoor radon risk because of their short half-lives and uncommon occurrence. In general, the concentration and mobility of radon in a soil are dependent on several factors, the most important of which are the concentration of radium in the soil and the soil's permeability. Soil moisture conditions, presence and types of soil structures, and weathering profile characteristics also influence radon availability.

Although there are few data on background levels of soil radium in the United States, a reasonable estimate for surface soils is less than 1 pCi/g (picocurie per gram) (Myrick and others, 1983; Sextro and others, 1987), which corresponds to 3 ppm (parts per million) uranium when uranium and radium are in secular equilibrium. Various equations (Bruno, 1983; Sextro and others, 1987) and empirical data (Peake, unpub. data, 1988) indicate that at 1 m (meter) below the soil surface, 1 pCi/g radium in natural soils can produce 700–1,400 pCi/L radon in soil gas. Typical concentrations of radon in natural soils may average approximately 900 pCi/L.

High radium concentrations have been closely associated with high indoor radon levels in the United States. Examples include the "index" home in Boyertown, Pa. (indoor radon concentration of 2,600 pCi/L), and the "discovery" house in Clinton, N.J., where the radon concentration reached similar levels (2,200 pCi/L) before mitigation. The Boyertown home is situated on a shear zone containing as much as 50 ppm uranium (Gundersen and

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[1] Environmental Protection Agency, Washington, D.C.

[2] U.S. Geological Survey.

others, 1988a). One soil sample collected near the "discovery" house contained 85 pCi/g radium, and a soil-gas radon sample at the same site had a concentration of 120,000 pCi/L. Soil samples collected around 12 homes with elevated indoor radon levels in the Reading Prong in eastern Pennsylvania had a median radium concentration of 2.4 pCi/g (Peake, 1988). The median radium concentration for soil samples collected by Gundersen and others (1988a) around other homes with elevated radon levels in the Reading Prong is also greater than 2 pCi/g.

Radon transport in soils occurs by two processes, diffusion and convective flow (Tanner, 1964). Diffusion is the dominant radon transport process in soils of low permeability (generally less than 10^{-7} cm^2 (square centimeters), or approximately 14.0 in/hr (inches per hour)), whereas convective transport processes tend to dominate in highly permeable soils (generally greater than $10^{-7}cm^2$; Sextro and others, 1987). Highly permeable soils, such as those derived from glacial outwash and on eskers, have been associated with high indoor radon levels in Europe and in the United States (Åkerblom and others, 1984; Sextro and others, 1987; Kunz and others, 1989). In areas of karst topography, high secondary porosity is postulated to increase soil permeability at depth to the extent that Soil Conservation Service (SCS) soil permeability data for the regolith may provide an incorrect estimate of radon availability. Solution cavities and caverns may provide routes for free movement of soil gas, especially when a building exerts pressure on the openings. In low-permeability soils, much of the radon decays before it is able to enter a structure because the radon's transport distance is limited.

DRIVING FORCES AND BASEMENT VERSUS NONBASEMENT HOMES

A driving force (negative pressure in the house relative to the soil) and entry points must exist for radon to enter a home from the soil. The negative pressure caused by furnace depressurization and the stack effect during cold winter months are common driving forces. Sump-pump holes and slab to foundation wall joints are common entry points.

EPA/State Indoor Radon Survey data indicate that homes with basements are more likely to have elevated indoor radon measurements than nonbasement homes (Ronca-Battista and others, 1988). The term "nonbasement" applies to slab-on-grade or crawl space construction. Two areas having the same geologic radon potential, one primarily with basement homes and the other primarily with nonbasement homes, would likely be characterized by two different radon frequency distributions because the measurements would be made on different floors. Basement homes generally have higher average indoor radon levels because basement homes provide more entry points for radon, commonly have a more pronounced stack effect, and typically exert more negative pressure on their foundations than nonbasement homes. Another important point, however, is that basement homes may tend to have higher indoor radon levels simply because the measurement is made in the basement rather than on the main floor. In many cases, the main floors of homes with basements have radon levels similar to those on the main floors of nonbasement homes; the implication in those cases is that occupants of basement homes are at greater risk from radon exposure only if they spend a significant amount of time in their basements. Information on foundation types (fig. 62), organized by State, can be obtained from Sterling and others (1985) or from major builders or Realtor associations.

ASSESSMENT SCALES

The types of data used for assessments of radon potential are dependent in large part on the scale of the area being evaluated. Larger areas require less detail and provide more general estimates, because the resulting maps show areas in which homes are more or less likely to have elevated indoor radon levels, but such maps cannot be used to predict the radon levels accurately in individual homes. The main purpose of such a map is to serve as a planning guide for radon testing programs, but these maps have also been used as preliminary maps of hazard potential because they can often be compiled from existing geologic data. To predict an individual home site's indoor radon level accurately, on-site field testing must be done to provide information on the physical and chemical characteristics of the soil. In addition, data on the variability of each soil property must be collected.

Understanding the variability of properties within units is more important when relatively small areas are evaluated. For example, although relatively minor variations in permeability within a soil unit may have little effect on the ranking of an area in a statewide or countywide evaluation, they may be critical to determining an individual home site's radon availability. State-sized areas and individual home sites are representative of the extremes of radon assessment scales; the intermediate scales may then be added to produce a list of typical assessment scales, as follows (Schumann, unpub. data, 1988):

1. State
2. County
3. Bedrock/soil units
4. Individual building sites (house construction characteristics)
5. Macroscopic soil properties (permeability, soil profiles, meteorologic effects, radon transport processes)
6. Molecular-scale properties of radon and soil particles (radon emanation processes)

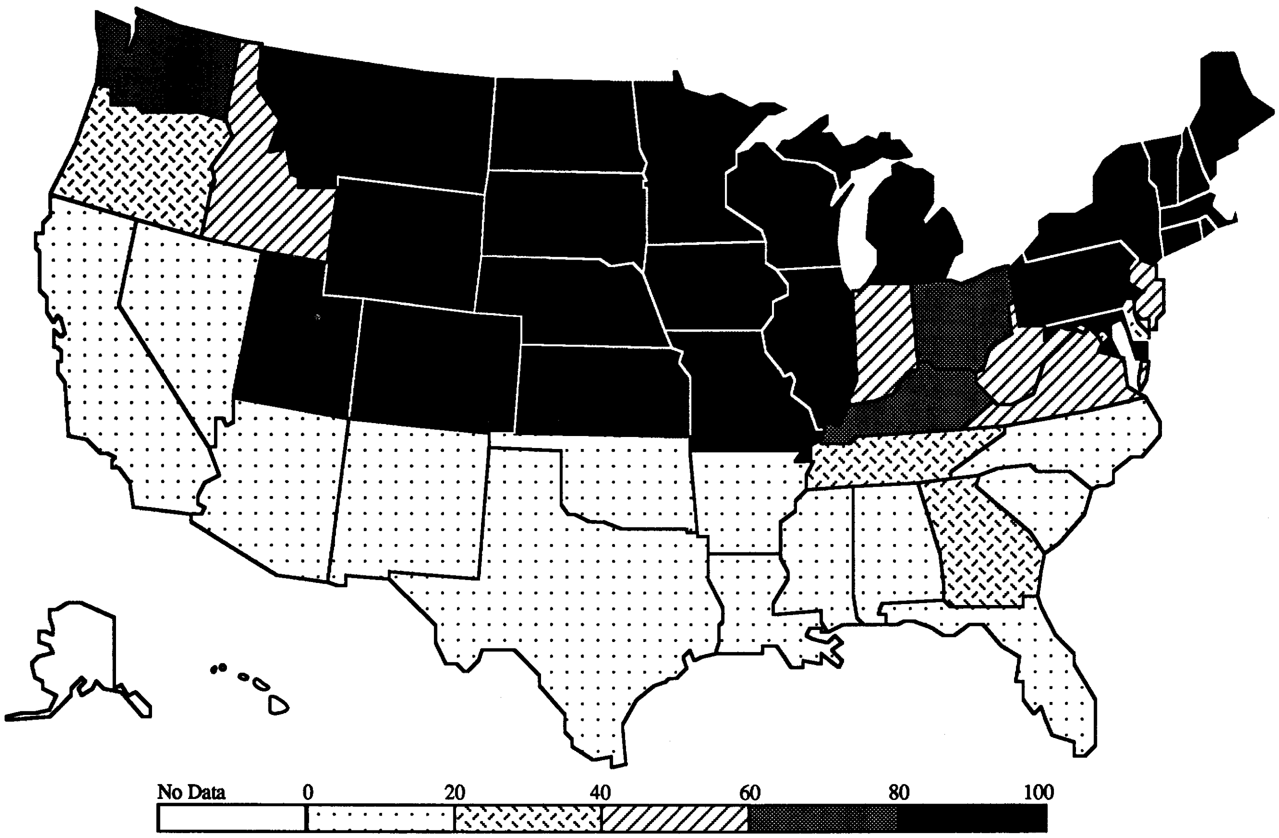

Figure 62. The percentage of homes having basements in the United States, by State (after Sterling and others, 1985). Homes that do not have basements are categorized as slab-on-grade or crawl space construction.

As a general rule of thumb, it is necessary to understand processes and provide detail at two scales larger than the scale of the area to be assessed. To evaluate a State-sized area (category 1), one would need to evaluate the radon potential of individual bedrock and soil units within that area (category 3), whereas an evaluation of radon potential within a single rock or soil unit (category 3) would require additional information on the variability of physical and chemical properties of the bedrock and soil within the unit (category 5), as well as on house construction characteristics (category 4). In this report we discuss the methods and results of large-scale (State) characterizations of radon potential, such as those compiled for the EPA/State Indoor Radon Survey, so the following discussions will focus on elements of evaluations at this scale.

ASSESSMENT TECHNIQUES

An assessment of geologic radon potential can be made from four main types of data: geologic (lithologic), radiometric, soil permeability, and available indoor radon data. These elements can be integrated to produce an estimate of radon potential.

Indoor radon data are an important element of geologically based assessments of radon potential. Existing indoor radon data for an area of interest can give clues to the overall distribution of indoor radon concentrations and provide a basis for preliminary comparisons with geologic data. Possible sources of indoor radon data include local, State, and Federal agencies, power companies, private contractors, and the homeowners themselves. One example of a readily available summary of regional indoor radon information is a series of reports released quarterly by the Bonneville Power Administration for the Northwestern United States (U.S. Department of Energy, Bonneville Power Administration, 1988). When using data of this type, however, it is important to remember that the data set may not necessarily be statistically representative of the actual distribution of indoor radon levels in an area. Volunteers are often motivated to test their homes when they are aware of other homes in the area that have elevated radon levels (Ronca-Battista and others, 1988). Another problem with many indoor radon data sets is that the data points may be only generally located (by Zip Code, for example), so that direct comparisons of indoor radon values of homes with their underlying geology may be difficult.

Information on the type and distribution of lithologic units and other geologic features in an assessment area is of primary importance. The most common radon source rocks include many granitic and metamorphic rocks, black shales, phosphatic rocks, and some carbonate rocks. Granitic rocks commonly concentrate uranium in specific minerals during initial crystallization; uranium in metamorphic rocks has typically been remobilized and concentrated by heat, pressure, and fluid migration during metamorphism. The organic compounds in black shales are natural concentrators of uranium and other metals; phosphates also provide favorable chemical conditions for concentrating uranium and radium. Although carbonate bedrock is generally low in radium, areas underlain by carbonates can generate elevated indoor radon concentrations. One reason may be that soils formed from carbonate bedrock tend to be enriched in uranium during weathering (Kukoc, 1980; Rose and others, 1988). Some continental sandstones contain localized deposits of uranium ("roll front" deposits, for example) that may generate locally elevated concentrations of radon in soil gas. Although many cases of elevated indoor radon levels can be traced to high radium and (or) uranium concentrations in parent rocks, some structural features, most notably faults and shear zones, have been identified as sites of localized uranium concentrations and have been associated with some of the highest known indoor radon levels (Deffeyes and MacGregor, 1980; Gundersen and others, 1988a,b; Gundersen, this volume). Investigating faults and shear zones is an important, but often overlooked, part of the preliminary characterization process, because in favorable geochemical settings, they can be likely sources of radon "hot spots."

Aerial radiometric data are used to describe the radioactivity of rocks and soils. Although radiometric data usually comprise spectral as well as total-count gamma-ray data, equivalent uranium (eU) data are typically used for radon characterizations because they provide an estimate of the surficial concentrations of radon parent materials (uranium, radium) in rocks and soils. Equivalent uranium is calculated from the counts received by the gamma-ray detector in the wavelength corresponding to bismuth-214 (^{214}Bi), with the assumption that uranium and its daughters are in secular equilibrium. The primary source of aerial radiometric data in the United States is reports of the National Uranium Resource Evaluation (NURE) program of the 1970's and early 1980's. The purpose of the NURE program was to identify and describe areas in the United States having potential uranium resources (U.S. Department of Energy, 1976). Hill (1986) lists aerial radioactivity and aerial magnetic data available for the United States. Ground-based radiometric data have also been used for preliminary assessments in some areas (Cannia and Case, 1986; Pennsylvania Topographic and Geological Survey, 1985). In this discussion we will focus primarily on the use of NURE aerial radiometric data for area characterizations.

Use of NURE Data for Radon Characterizations

Although radon is highly mobile in soil, and its concentration is affected by meteorologic conditions (Kovach, 1945; Jaacks, 1984; Schery and others, 1984; Schumann and others, 1989), a relatively good correlation between average soil-gas radon concentrations and average eU values for some soils has been noted (Gundersen and others, 1988b; Schumann and Owen, 1988). Aerial radiometric data can provide an estimate of radon source strength over a region, but the amount of radon that is able to enter a home from the soil is dependent on several additional factors, including soil structure, grain size distribution, moisture content, and permeability, as well as type of house construction and structural condition, and these factors must also be considered when using radioactivity data for radon characterizations.

The types of data in, and the presentation format of, NURE reports are variable, depending on the contractor that performed the survey. The data are presented in 1°×2° format and include graphs of the flight-line data, uranium anomaly maps, standard deviation maps, and histograms of the radiometric data grouped by lithology. In addition, many NURE quadrangle reports also include pseudocontour maps of the spectral gamma-ray data; some reports include maps of known uranium occurrences and rocks having favorable uranium potential.

NURE aerial radiometric data are best used for characterizing large areas such as a State or region (Peake, 1988) because of the relatively wide spacing between adjacent flight lines. The primary NURE flight lines are oriented east-west and are generally spaced 3 mi (miles) apart in the Western United States and 6 mi apart in the Eastern United States. With this flight-line spacing, a large unmeasured area exists between flight lines. This may be less important when data from several flight lines, covering a large area of relatively uniform geology, are integrated; however, localized anomalies may be overlooked. For example, the uranium anomaly underlying homes that have the highest indoor radon values in the Boyertown, Pa., area was not shown in the NURE reports because it is relatively small and located between flight lines; however, the entire Reading Prong displays an elevated ^{214}Bi signature.

The shallow (20–25 cm) depth of investigation of gamma-ray spectrometers, either ground-based or airborne (Duval and others, 1971; Durrance, 1986), suggests that gamma-ray data may sometimes provide an underestimate of radon source strength, especially in soils with deep weathering profiles or where the surficial materials are derived from a different source than the underlying bedrock (for example, transported soils). As a soil weathers, some minerals in the near-surface soil layers are leached and moved downward in the soil profile, where they may be concentrated in the B horizon, whereas the concentration of radionuclides in the C horizon and below may be relatively unaffected. In these cases the surface gamma-ray signal indicates a lower radon source concentration than actually exists in the deeper soil layers. Therefore, soil profile data should be considered and correlated with radioactivity data whenever possible. Schumann and Owen (1988) describe an example of relatively high soil-gas radon concentrations generated by a soil derived from a weathered granite with low surface radioactivity in Fairfax County, Va.

The initial phase of a regional evaluation begins by selecting the type of NURE data to be used—flight-line data, anomaly maps, standard deviation maps, or pseudocontour maps—and comparing them with geologic maps. NURE aerial radiometric data are typically presented in the 1°×2° format reduced to a scale of 1:500,000, which corresponds to, or is easily converted to, the scale of many State geologic maps. Correlations between eU signatures and outcrop patterns of geologic units should be noted, as well as any anomalies corresponding to structural features such as faults or shear zones. Areas with less than 1.0 ppm eU are delineated and assigned a low radon potential, those with 1.0 to 2.0 ppm eU are assigned a moderate potential, and areas with greater than 2.0 ppm eU are designated areas of high radon potential. These rankings may be modified later as soil permeability data are integrated and compared; low soil permeability generally reduces the radon risk, whereas high permeability increases the radon risk. Pseudocontour maps of eU may often be used without modification, except for adjustments between adjacent or nearby quadrangles for contractor differences or other inconsistencies. These maps provide a quick and easy preliminary method for determining the radon potential of an area or region but are less detailed than maps interpreted from flight-line anomaly maps, standard deviation maps, or flight-line data because of the data smoothing that is performed as part of the contouring process. An alternate method involves calculating a weighted mean eU value for a 1°×2° quadrangle by using the median eU values from the histogram data and weighting them by the number of adequate samples for each lithologic unit. This method provides an average eU value for the quadrangle that can be used for comparison with eU values in other quadrangles.

Use of SCS Soil Survey Data

Soil surveys prepared by the Soil Conservation Service (SCS) provide data on soil characteristics, including soil cover thickness, grain size distributions, permeability, shrink-swell potential, typical vegetative cover, generalized ground water characteristics, and land use. The reports are typically available in State and county formats and usually contain both generalized and relatively detailed maps of soils in the area. Quantitative soil characteristics are presented in tables following descriptions of the physiography, geology, climate, and soils of the area.

For regional-scale radon characterizations, a generalized soil map should be compared to a geologic map of the area, and the soil descriptions, shrink-swell potential, and permeability of each soil group should be noted. The permeabilities listed in the SCS surveys are for water, but they are generally related to gas permeability. Permeabilities reported by the SCS as greater than 6.0 in/hr may be considered high and may increase the radon potential of areas with low to moderate radium concentrations (less than 2.0 ppm eU). In contrast, permeabilities less than 0.6 in/hr may be considered low in terms of soil-gas transport. Soils with permeabilities in this range may have a lower radon potential than more permeable soils with similar eU concentrations.

Soil permeability is generally highest near the surface and decreases with depth. Many well-developed soils contain a clay-rich B horizon that may impede vertical soil-gas transport. Radon generated below this horizon cannot readily escape to the surface, so it would instead tend to move laterally, especially under the influence of a negative pressure exerted by a structure. Soil permeability data and weathering profiles thus provide important information for regional radon assessments. Depth to water table can also be an important parameter to consider in some areas. Because water in soil pores inhibits gas transport, the amount of radon available to a home is effectively reduced by a high water table. Areas likely to experience effects of high water tables are river valleys, coastal areas, and some areas overlain by deposits of glacial origin (for example, loess).

Shrink-swell potential is an indicator of the abundance of smectitic (swelling) clays in a soil. Soils with a high shrink-swell potential may cause building foundations to crack and thus create pathways for radon entry into the structure. In addition, swelling soils often crack as they dry; as a result, they provide additional pathways for soil-gas transport and effectively increase the gas permeability of the soil (Schumann and others, 1989).

CORRELATIONS BETWEEN EQUIVALENT URANIUM AND INDOOR RADON DATA

Nine 1°×2° quadrangles exhibiting high and low radioactivities in geologically and geographically diverse regions of the United States were selected to illustrate the relation between indoor radon measurement data and eU concentrations (table 20). The quadrangles were selected on the basis of available NURE, soil, and randomly collected indoor radon data. The EPA/State Indoor Radon Survey was the primary source of indoor radon data. Data from counties within each of the areas were combined and grouped by house type (basement versus nonbasement) for comparisons with geologic and soil data. Indoor radon data for Florida were obtained from the report of Florida's State

Table 20. Equivalent uranium (eU), soil permeability, housing construction, and indoor radon data for nine areas sampled during the 1986–87 EPA/State Indoor Radon Survey.

[ppm, parts per million; in/hr, inches per hour; pCi/L, picocuries per liter; N/A not applicable]

				Basement			Nonbasement		
Area	Weighted mean NURE eU (ppm)	Typical soil permeability (in/hr)	Percent of homes with basements	Radon average (pCi/L)	Percent >4 pCi/L	Number of samples	Radon average (pCi/L)	Percent >4 pCi/L	Number of samples
Southern Alabama	1.5	0.2–2.0	1	1.0	0	1	0.8	1	83
Eastern Kentucky	1.8	2.0–5.0	N/A	N/A	N/A	N/A	1.3	3	100
North-central Kentucky .	2.5	.6–2.0	48	6.3	53	64	3.2	27	70
Southeastern Colorado ..	2.8	.6–20.0	24	6.5	62	21	2.5	15	65
Denver metro area	3.2	.02–20.0	83	5.6	60	67	3.0	21	14
Central Michigan	1.2	.6–20.0	86	1.5	8	12	N/A	0	2
Southern Michigan	1.2	.6–20.0	100	3.2	38	16	N/A	N/A	0
N.J. Outer Coastal Plain .	1.0	<.2 to >6.0	64	1.7	7	164	.9	1	90
Florida	1.1	.6–6.0	N/A	N/A	N/A	N/A	.7	3	3,050

indoor radon survey (Geomet Technologies, Inc., 1987). The indoor radon data for New Jersey were obtained from a radon survey conducted by the State of New Jersey (Karl Muessig, personal commun., 1988). Each of the surveys used randomly collected charcoal-based screening measurements.

Average eU values were determined for each area by calculating weighted means from the histogram data in the NURE folios for the appropriate 1°×2° quadrangles. Equivalent uranium values for only those rock types actually exposed in the area were used to determine the average eU values. If the area was covered by more than one 1°×2° quadrangle, the eU data from all quadrangles covering the area were combined to determine the average eU value. The NURE data for the State of Florida were derived from a summary of average eU concentrations listed by county. The New Jersey NURE data were obtained from an unpublished compilation of NURE aerial radiometric data for the mid-Atlantic States (J.S. Duval, written commun., 1988).

A plot of indoor radon and eU data from the nine study areas (fig. 63) shows a positive relation between eU concentrations and average screening indoor radon levels in basement and nonbasement homes. Each data point represents an average radon level for a group of counties and the corresponding NURE eU average. Given the same regional eU concentration, basement construction is associated with higher screening levels than nonbasement construction. The slope of the line for the basement population is steeper than that of the nonbasement population by a factor of approximately two. One part per million eU is associated with a screening measurement of about 1 pCi/L in living areas and about 2 pCi/L in basements. In areas where the surface eU concentration is below about 1 ppm, nonbasement homes should contain little radon. A similar correlation is seen in the Pacific Northwest (Duval and others, 1989). A corresponding minimum level for basement homes may be even lower, but there are few natural soils with radium concentrations lower than 1 ppm.

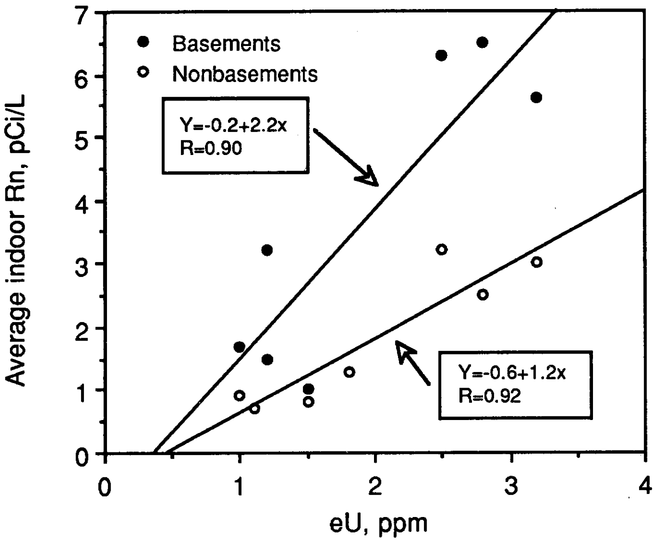

Figure 63. Average equivalent uranium (eU) concentrations versus basement and nonbasement indoor radon averages for nine areas sampled during the 1986–87 EPA/State Indoor Radon Survey. Data from table 20. ppm, parts per million; pCi/L, picocuries per liter.

A CASE STUDY

The EPA/State Indoor Radon Survey of Kentucky provides an illustrative example of characterization of geologic radon potential. Because the initial indoor radon survey in Kentucky has been completed, the geologic data can be compared with preliminary indoor radon data to illustrate the relation between geology and indoor radon concentrations.

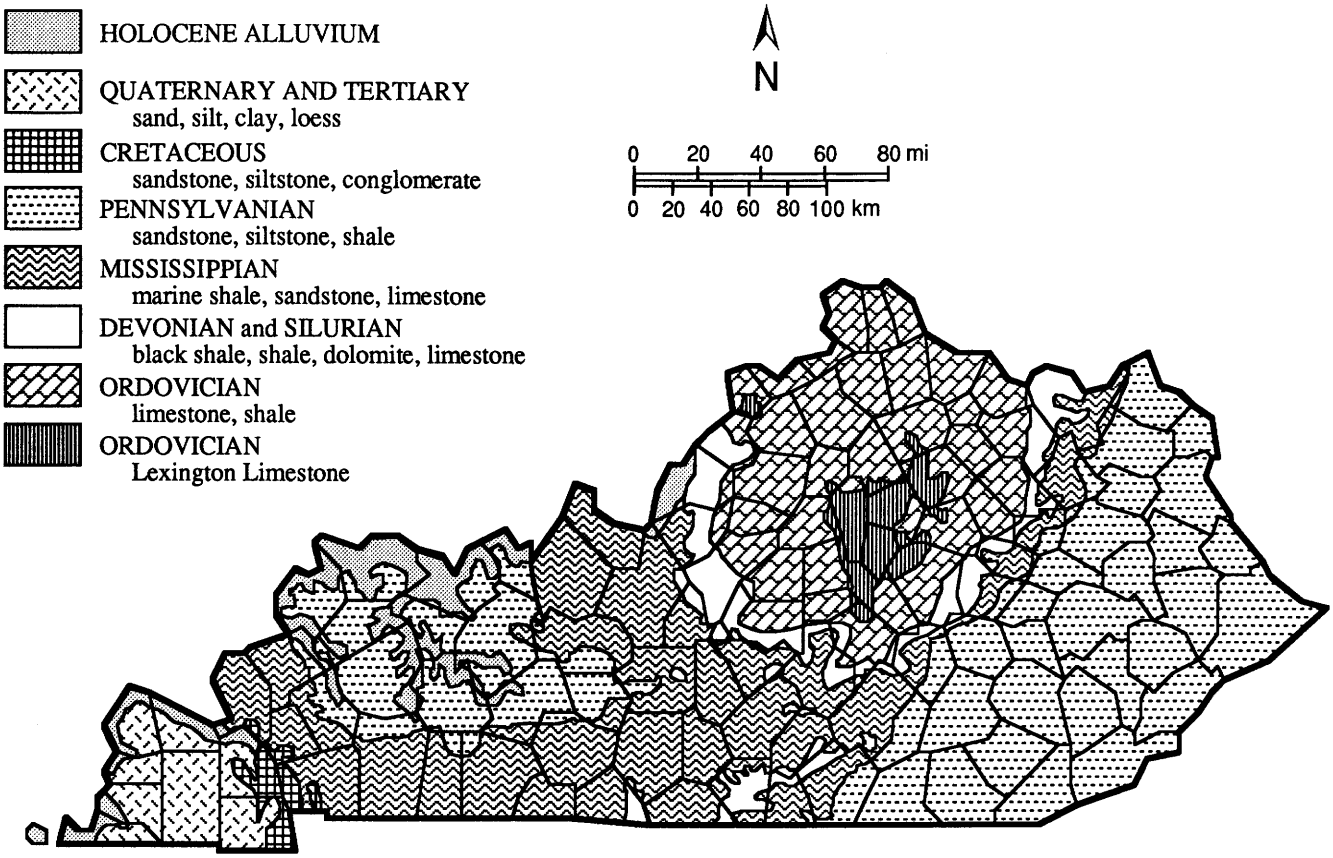

Figure 64. Generalized geologic map of Kentucky (after Winsor and Bailey, 1963).

The geology (fig. 64), physiography (fig. 65), soils (fig. 66), and surface radioactivity (fig. 67) of Kentucky are interrelated, with bedrock geology as the dominant controlling factor. A geologic map of Kentucky (McDowell and others, 1981; fig. 64 is a generalized version) was examined to identify rock types and geologic features suspected of producing elevated radon levels. Three primary areas were identified: (1) areas underlain by Devonian black shales in the Outer Bluegrass region (figs. 64, 65); (2) areas underlain by the Ordovician Lexington Limestone (some of which is phosphatic) in the Inner Bluegrass region (figs. 64, 65); and (3) areas underlain by, or close to, major faults in the Western Coalfield and Inner Bluegrass regions (fig. 65; McDowell and others, 1981). The black shale and limestone areas have associated high surface radioactivity, and the Western Coalfield contains scattered radioactivity anomalies (fig. 67). In addition, generally elevated surface radioactivity is associated with Ordovician limestones and shales in the northern part of the Outer Bluegrass region (figs. 64, 65). The arcuate pattern of radioactivity anomalies along the southern edge of the Outer Bluegrass region (figs. 65, 67) corresponds closely to the outcrop pattern of the Devonian Chattanooga Shale (fig. 64). A large radiometric anomaly along the State's southern border (fig. 67) roughly follows the outcrop pattern of the Upper Mississippian Ste. Genevieve and St. Louis Limestones (McDowell and others, 1981). The clastic sedimentary rocks of the Cumberland Plateau region (figs. 64, 65) are generally characterized by relatively low surface radioactivity (fig. 67).

A generalized soil permeability map (fig. 66) was compiled from a soil association map (Winsor and Bailey, 1963) and county soil surveys published by the SCS. The map shows that most soils in Kentucky are of low or moderate permeability. Some areas underlain by limestones may have increased permeability locally because solution cavities are present. Soils formed on the Lexington Limestone, in the Inner Bluegrass region, have a significantly higher permeability than soils in the surrounding Outer Bluegrass region (figs. 64–66). The Inner Bluegrass region has moderate to high radon potential because it has elevated surface radioactivity and moderate soil permeability. Although high surface radioactivity is also characteristic of the Outer Bluegrass region, it has a low to moderate radon potential because of the low permeability of the soils formed on the shales and limestones (figs. 64, 66, 67). The Cumberland Plateau and Jackson Purchase areas (fig. 65)

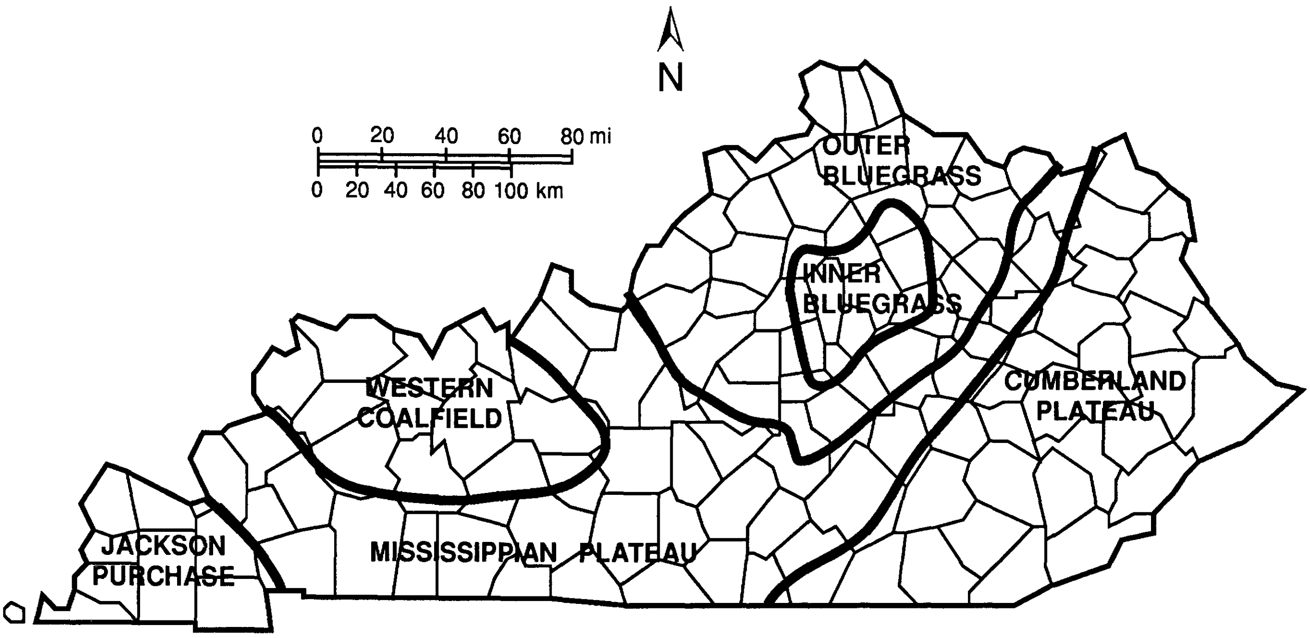

Figure 65. Generalized map of major physiographic provinces of Kentucky (after McDowell and others, 1981).

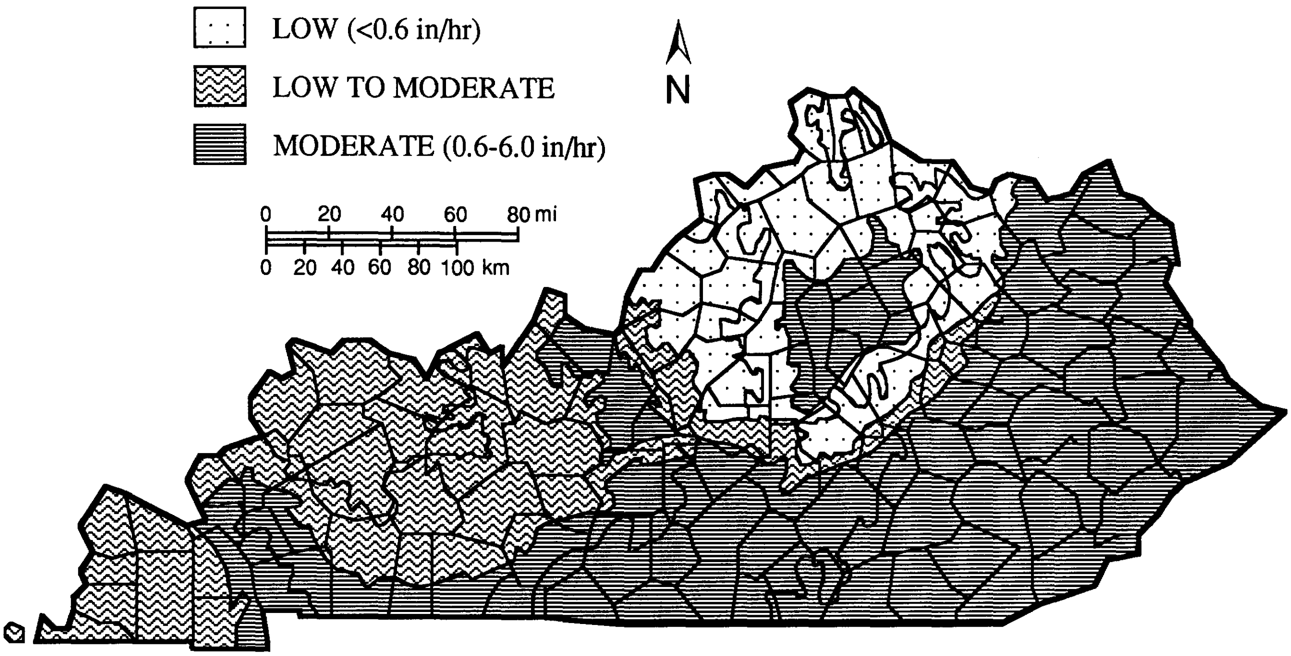

Figure 66. Generalized soil permeability map of Kentucky (after Winsor and Bailey, 1963). "Low to moderate" indicates that the area contains mixed soils, some soils with low permeability and some with moderate permeability.

generally have low radon potential. The Western Kentucky Coalfield (fig. 65) has low to moderate radon potential because few homes are situated directly on faults. In addition, soils in the Western Coalfield are of low to moderate permeability and only scattered radioactivity anomalies are found in this area. The western part of the Mississippian Plateau (fig. 65) has moderate radon potential, whereas the eastern part has generally low radon potential because of the distribution of gamma anomalies and the generally moderate soil permeability.

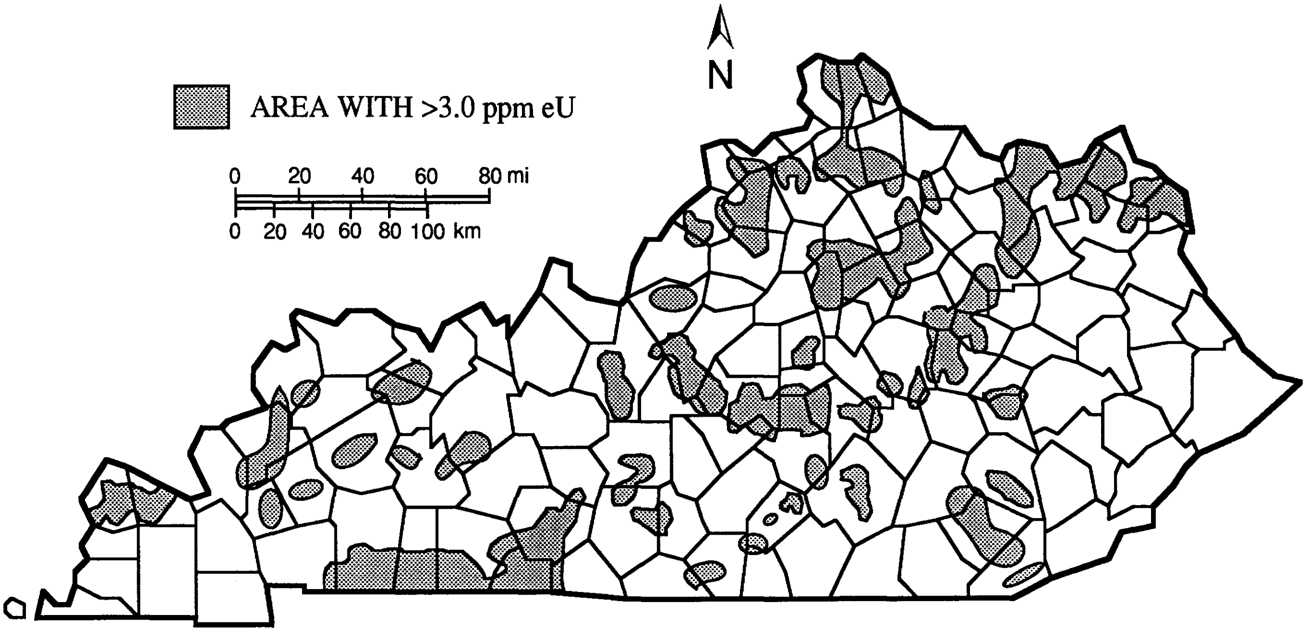

Figure 67. Map showing areas of anomalous surface radioactivity (greater than 3.0 ppm equivalent uranium), compiled from NURE 1°×2° quadrangle reports for Kentucky and adjacent States.

Table 21 presents a partial summary of results of the EPA/State Indoor Radon Survey of Kentucky conducted during the 1986–87 winter season. The areal distribution of these screening indoor radon levels is shown in figure 68. For the purposes of this discussion, only data from those counties in which five or more measurements were made for one floor are presented here. EPA's indoor radon measurement protocol (EPA, 1986) calls for screening measurements to be made in the lowest livable area in the home. Because of this, the data set contains measurements from both basements and main floors of homes that, although effective for estimating "worst-case" radon risk in individual homes, is not well suited for statistical analysis because data from different floors are not directly comparable. To compensate, ratios between average basement and main floor indoor radon levels were calculated for each county containing adequate numbers of measurements from both floors (table 21). Because basement radon levels average approximately twice as high as main-floor levels (table 21), main-floor indoor radon averages were multiplied by two to obtain estimates of average basement indoor radon levels in those counties that had an insufficient number (less than five) of basement measurements. These estimated average basement radon levels are shown in addition to actual measured basement radon averages on the indoor radon map of Kentucky (fig. 68).

The distribution of indoor radon levels (fig. 68) correlates with the underlying geology in Kentucky. Elevated indoor radon levels occur in the Inner Bluegrass region (figs. 65, 68) and in those homes underlain by the Chattanooga Shale, although this conclusion is based only on data from homes in the western part of the black-shale outcrop area. Although some homes with elevated levels were found in the northern Outer Bluegrass region, the number is less than would be expected on the basis of eU (fig. 67), likely because the soil permeability is low (fig. 66). Homes in the western part of the State have generally low indoor radon levels, although the eU data suggest that the indoor radon average should be about 2.5 times the 0.9 pCi/L average for the area. The poor correspondence with surface radioactivity in western Kentucky appears to be due primarily to a combination of low soil permeability and high water tables. Western Kentucky was the only part of the State in which seasonal high water tables were consistently listed in the SCS soil surveys as less than 6 ft, and commonly less than 2 ft. The two counties in the south-central part of the State having average indoor radon levels greater than 10 pCi/L (fig. 68) appear to correspond to the surface radiometric anomaly associated with the outcrop of the Ste. Genevieve and St. Louis Limestones (fig. 67; McDowell and others, 1981). Homes in the Cumberland Plateau region (fig. 65), which has a generally low radiometric signature (fig. 67), have correspondingly low average indoor radon levels (fig. 68).

DISCUSSION

These examples indicate that surface radioactivity and indoor radon levels correlate notably. Soil permeability and moisture conditions can enhance or reduce the radon

Table 21. Average screening indoor radon levels, number of homes tested, and percent greater than 4 picocuries per liter for 40 counties in Kentucky

[Measurements were made during the 1986–87 EPA/State Indoor Radon Survey; only data from those counties in which five or more measurements were made on one floor are listed]

County	Nonbasement			Basement			Ratio of basement/ nonbasement radon
	Average radon	Number of homes	Percent > 4 pCi/L	Average radon	Number of homes	Percent > 4 pCi/L	
Ballard	0.8	8	0				
Barren	1.1	5	0				
Boone				1.4	6	0	
Bourbon				10.1	7	71	
Boyd	.6	5	0	1.7	12	8	2.89
Bullitt				12.6	10	60	
Butler	.7	12	0				
Caldwell	1.3	5	0				
Calloway	.9	6	0				
Campbell				1.6	14	14	
Christian	1.4	15	13				
Daviess	.8	14	0	1.6	6	0	1.92
Fayette	5.0	23	43	7.5	17	65	1.51
Franklin				5.0	11	55	
Graves	.8	9	0				
Grayson				1.1	5	0	
Greenup	2.1	6	17	2.0	5	20	.94
Hardin	1.5	14	7	2.1	12	8	1.38
Harrison	.6	5	0				
Hart				11.6	5	100	
Henderson	1.3	6	0				
Hopkins	.4	6	0				
Jefferson	1.5	33	9	3.7	65	31	2.43
Jessamine	2.0	8	13	6.2	3	33	3.20
Kenton	2.1	9	11	1.1	23	0	.54
Logan	1.1	5	0				
McCracken	.8	13	0				
Marshall	.6	7	0				
Mason				1.4	5	0	
Morgan	.7	5	0				
Nelson	2.2	7	14	6.0	6	67	2.68
Pendleton				5.4	5	60	
Pike	1.6	8	0				
Pulaski				4.1	5	60	
Simpson	1.1	5	0				
Todd	1.3	5	0				
Union	.4	5	0				
Warren	5.8	18	61	16.1	5	80	2.78
Whitley	.9	5	0				
Woodford	3.0	5	20				
Average	1.5	9	7	5.1	11	37	2.03

potential of an area. Because an estimate of geologic radon potential (GRP) is composed of two main components, the concepts discussed here may be summarized by a two-dimensional diagram (fig. 69). Thus, to estimate the GRP of an area, eU should be compared with soil permeability, and the field encompassing the intersection of these values (low, moderate, or high; fig. 69) is the estimated GRP for the area. As was previously discussed, other soil and geologic features such as faults, high water tables, and weathering profiles should also be considered to estimate additional effects on soil-gas radon concentrations.

The GRP yields an estimate of radon source strength based on radiometric, geologic, and soil permeability data, but it will not necessarily provide an estimate of the

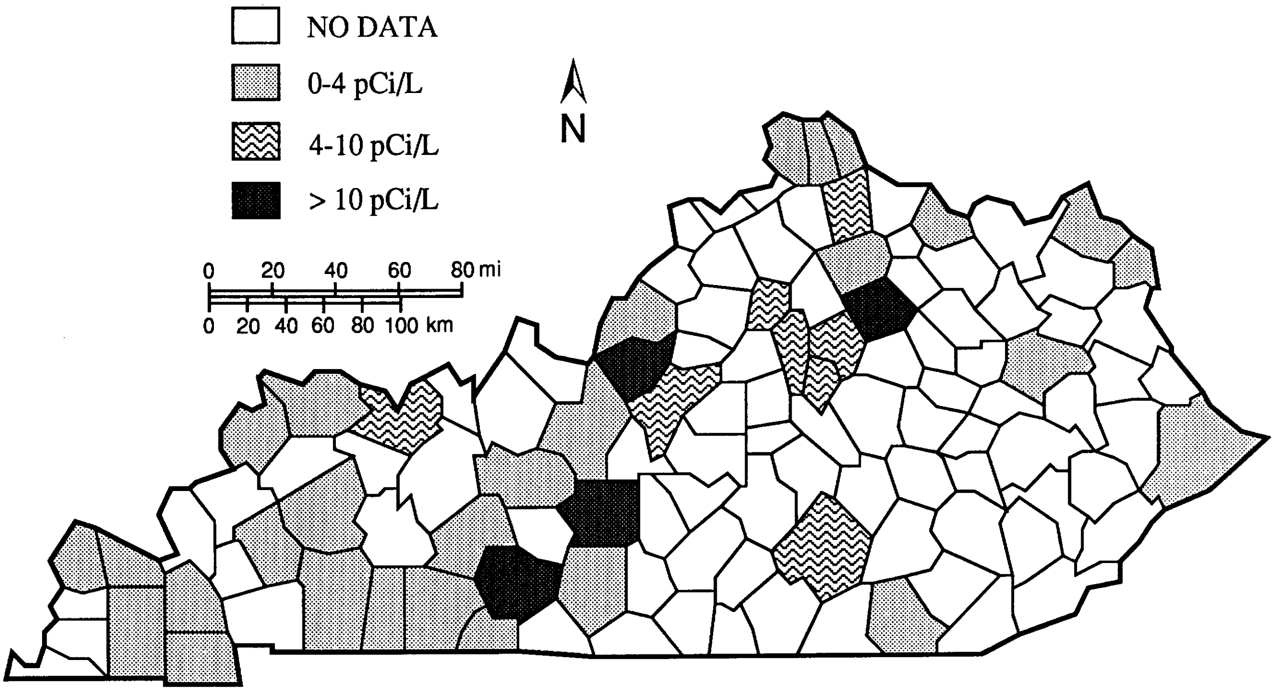

Figure 68. Average basement screening indoor radon levels by county. Data are from the 1986–87 EPA/State Indoor Radon Survey of Kentucky. Only those counties in which five or more indoor radon measurements were taken on one floor (basement or main floor) are shown. Where basement averages were not available, main-floor averages were multiplied by two to provide estimated basement values (see table 21 for a list of county data included on this map).

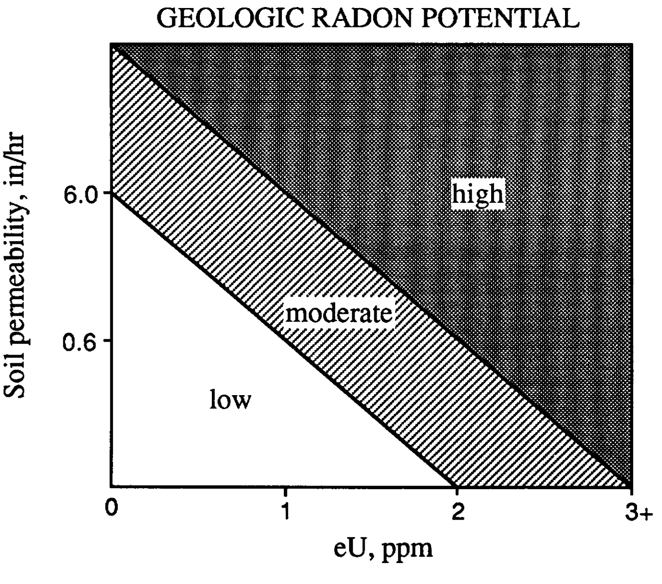

Figure 69. Diagram of geologic radon potential. eU, equivalent uranium; ppm, parts per million; in/hr, inches per hour.

percentage of screening measurements above 4 pCi/L in an area, because the radon concentration in a house is controlled by both geologic and house construction factors: geology indicates the potential for a problem to exist, whereas the house construction determines in part how the geologic potential will be realized in terms of indoor radon levels. As an estimate of expected indoor radon levels (rather than a direct estimate of radon risk), the integrated radon potential (IRP) concept takes the geologic radon potential estimate one step further by incorporating the GRP with house construction characteristics to better estimate the percentage of homes having indoor radon concentrations greater than 4 pCi/L in an area. Because the population of nonbasement homes has an average indoor radon concentration lower than that of the population of basement homes, the IRP for nonbasement homes would necessarily be less than for basement homes in an area that had the same geologic radon potential. An IRP diagram would look similar to the GRP diagram but would plot the previously estimated GRP against the percentage of homes with basements in the area. Although the IRP provides a useful conceptual framework for estimating indoor radon levels, there are currently insufficient data for determining cutoff points for such a diagram. The GRP and IRP concepts are best applied to large areas such as a State or region and may also be applicable to county-sized areas. Many additional details and data are required for radon potential estimates of smaller areas.

SUMMARY AND CONCLUSIONS

The purpose of a regional radon characterization is to identify areas of high radon potential. Existing information can be used to characterize areas quickly and can provide direction to more specific follow-up studies that may be able to identify localized "hot spots." A regional radon characterization serves as the first estimate of potential indoor radon risk within a State or other large region. Once high-risk areas are identified, health officials can allocate resources in areas where the radon risk is the greatest.

Regional geologic, soil, and house construction characteristics appear to strongly influence screening indoor radon frequency distributions. NURE eU concentrations of more than 2.0 ppm indicate high radon potential; however, whether or not a home has a basement has a significant effect on screening indoor radon levels and should be incorporated in analyses of indoor radon and geologic data. In addition, soil permeability and other properties can alter the relationship between eU and indoor radon levels by a factor of two or more; this may be especially significant in areas underlain by glacial soils. Because eU data correlate relatively well with average indoor radon concentrations, and indoor radon averages are correlated with the percentage of homes that have radon levels greater than 4 pCi/L, eU can be used to estimate the percentage of homes expected to contain greater than 4 pCi/L radon.

The concepts of geologic radon potential and integrated radon potential may provide a useful approach to separating and comparing geologic and house construction effects for indoor radon predictions and data analyses. The suggested guidelines are preliminary, but they may improve radon prediction capability as well as understanding of the distribution of indoor radon levels in the United States.

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RECONNAISSANCE APPROACH TO USING GEOLOGY AND SOIL-GAS RADON CONCENTRATIONS FOR MAKING RAPID AND PRELIMINARY ESTIMATES OF INDOOR RADON POTENTIAL

G.M. Reimer, Linda C.S. Gundersen, S.L. Szarzi, and J.M. Been

Abstract

Soil-gas samples collected along a traverse in Prince Georges County, Md., were analyzed for radon. The traverse included principal lithologies and areas for which total count aeroradiometric measurements showed various intensities. Although this region in the Atlantic Coastal Plain might be considered to possess the type of geologic setting or aeroradiometric signature that would not contribute to significant radon potential, the measured soil-gas concentrations of up to 2,500 picocuries per liter indicate that the potential exists for indoor accumulations in excess of the level of 4 picocuries per liter established by the Environmental Protection Agency for remedial action. The higher levels of soil-gas radon were found in some of the Tertiary sediments, and lower concentrations occurred in Cretaceous and Quaternary sediments. Geologic relationships are thought to be the controlling factors for the radon soil-gas concentrations. The few data available from this survey suggest that a reconnaissance approach can be an effective screening method in the evaluation of areas for high radon potential.

INTRODUCTION

Radon, a radioactive gas that is a product of the decay of radium, has been found to accumulate in some homes at concentrations that are considered to pose a concern to public health (Environmental Protection Agency, 1986). Factors causing the gas to accumulate are numerous and include the manner of house construction and the concentration of radium in the soil or bedrock upon which the house is built. Radon, as a mobile gas, does not migrate very far from its radium parent because of its short half-life. Radium itself is derived from the radioactive decay of uranium or thorium, the concentration and distribution of which are strongly controlled by petrologic conditions during rock- and mineral-forming processes. Geology and soil chemistry are then dominant in controlling the surficial, natural distribution of uranium, thorium, and radium (Gabelman, 1977). Estimates of potential indoor radon accumulations would be very useful for assessment purposes in planned or existing residential areas. Preliminary surveys from areas known to have high indoor radon concentrations are providing a data base that can be used to derive the radon potential for an area. This study demonstrates how geology and soil-gas radon measurements can be combined in a reconnaissance survey to establish a data base for estimating the radon potential.

GEOLOGIC SETTING

Prince Georges County, Md., lies in the Atlantic Coastal Plain. In the region of this study, the underlying sediments are deltaic and marine in origin and consist of clay, sand, and gravel of Cretaceous, Tertiary, and Quaternary age. The sediments dip gently to the southeast, and the present-day drainage is toward the south-southeast. In general, the age of the exposed sediments increases toward the west. Figure 70 shows the location of samples on a geologic map for the county. No known faults intercept the surface, but Jacobeen (1972) has interpreted seismic evidence for high-angle reverse faulting at depth.

Clean, silicious, non-organic marine sedimentary rocks have not been considered to be a source for high radon emanation, and, indeed, the natural radioactivity for this area is fairly low and without great contrast (Neuschel, 1965) when compared to areas containing crystalline rocks, such as Montgomery County, located immediately to the west. Very few homes in Prince Georges County have been tested for indoor radon, and some exceed the level of 4 pCi/L (picocuries per liter) established as a guideline for follow-up action by the Environmental Protection Agency (EPA). No indoor concentrations reported by the date of this study exceeded 40 pCi/L.

ANALYTICAL TECHNIQUE

The soil-gas collecting and analytical techniques were as described by Reimer and Bowles (1979) and Reimer (this volume) for making a rapid field assessment of soil-gas radon concentrations. Basically, the method consists of pounding a hollow steel probe into the ground to a depth of 0.75 m (meter) and withdrawing a 10-cm^3 (cubic centimeter) sample with a hypodermic syringe. The sample is then injected into an evacuated, phosphor-coated cell and counted with a portable alpha-particle scintillometer (Reimer, 1977). The analytical unit had been calibrated by using the U.S. Bureau of Mines radon test facility in Denver, Colo. The short-lived radon isotopes were allowed to decay before the sample was introduced to the cell, and only radon-222 (^{222}Rn) concentrations were measured. The accuracy of the radon concentrations are ±50 pCi/L.

Indoor radon concentrations from 21 homes were measured by charcoal canisters exposed in basements for a few days. These data were provided on a voluntary basis by the individual homeowners. The exact locations are not known, only the geologic formations in which the homes

Table 22. Soil-gas radon concentrations for the traverse in Prince Georges County, Md.

[The sample locations are shown in figure 70, and the values are in sequence from west to east with reference to that figure. pCi/L, picocuries per liter]

Sequence number	Radon concentration (pCi/L)
1	1,250
2	150
3	1,100
4	100
5	900
6	500
7	700
8	1,050
9	1,000
10	700
11	1,000
12	500
13	450
14	500
15	1,050
16	950
17	100
18	350
19	850
20	100
21	100
22	500
23	700
24	450
25	400
26	550
27	200
28	650
29	900
30	950
31	150
32	1,700
33	700
34	1,600
35	2,700
36	800
37	1,050
38	2,050
39	250
40	750
41	650
42	100
43	1,300
44	1,450
45	950
46	950
47	150
48	800

were built. No indoor radon concentrations are available from locations along the traverse, so direct correlations between indoor radon and soil-gas radon are not possible.

DATA AND DISCUSSION

Forty-eight soil-gas samples were collected on the approximate 20-km (kilometer) traverse (fig. 70). The soil-gas radon concentrations range from 100 to 2,700 pCi/L, with an arithmetic mean of 750 pCi/L. Table 22 gives the radon concentrations for the individual samples, which are sequenced from west to east. Figure 71 shows frequency histograms of the soil-gas and indoor radon distribution for the individual geologic formations from which the samples were collected. The Tertiary formations show the greatest range in concentrations, although the data are too few to truly characterize the distribution. Figure 72 shows a summary distribution of the radon concentrations for the specific formations, Quaternary, Tertiary, and Cretaceous, but, again, many have few data points. The highest concentrations are associated with the Chesapeake Group (Tc) and the Aquia Formation (Ta). The Chesapeake Group consists of light-colored, fine to very fine marine sand, silt, clay, and thin layers of diatomaceous earth, although these layers have been described as principally occurring south of the traverse location. The Aquia Formation is a greenish-gray clayey sand composed mostly of quartz and glauconite. It is crossbedded and contains layers cemented by iron and shell layers cemented by carbonate. The Upper Member of the Nanjemoy Formation also contains glauconite but was not sampled during this survey. The Marlboro Clay Member of the Nanjemoy Formation (Tmb) is reported to contain glauconite sand also but only in the upper 1-m portion, and there the sand is restricted to filling in tubes or burrows. Higher concentrations of radioactive elements can be concentrated in glauconite and diatomaceous earth (Sverdrup and others, 1942). That this is the case with these particular sediments in Prince Georges County is suggested by the soil-gas radon data. Specific chemical analyses for radium and uranium would reveal if this association is correct.

The soil-gas radon traverse crossed several zones of different radiation intensities revealed in the gamma aeroradioactivity map of Neuschel (1965). The highest gamma radiation along the traverse is associated geologically with a portion of the Tertiary Brightseat Formation and Monmouth Group (TKb), a marine sand and clay also containing glauconite. The mean radon concentration for the four soil-gas samples from this formation is 800 pCi/L, only slightly higher than the mean of 750 pCi/L for the entire data set. No observable correlation existed between the soil-gas radon and the higher gamma-aeroradioactivity signature, but more sampling would have to be performed for a sufficient comparison to be made.

The upper limit of radon soil-gas concentrations observed in this study corresponds to some concentrations found in regions of the Reading Prong in Pennsylvania and New Jersey (Gundersen, unpub. data, 1987; Gundersen and others, 1988a), in Montgomery County, Md. (Gundersen

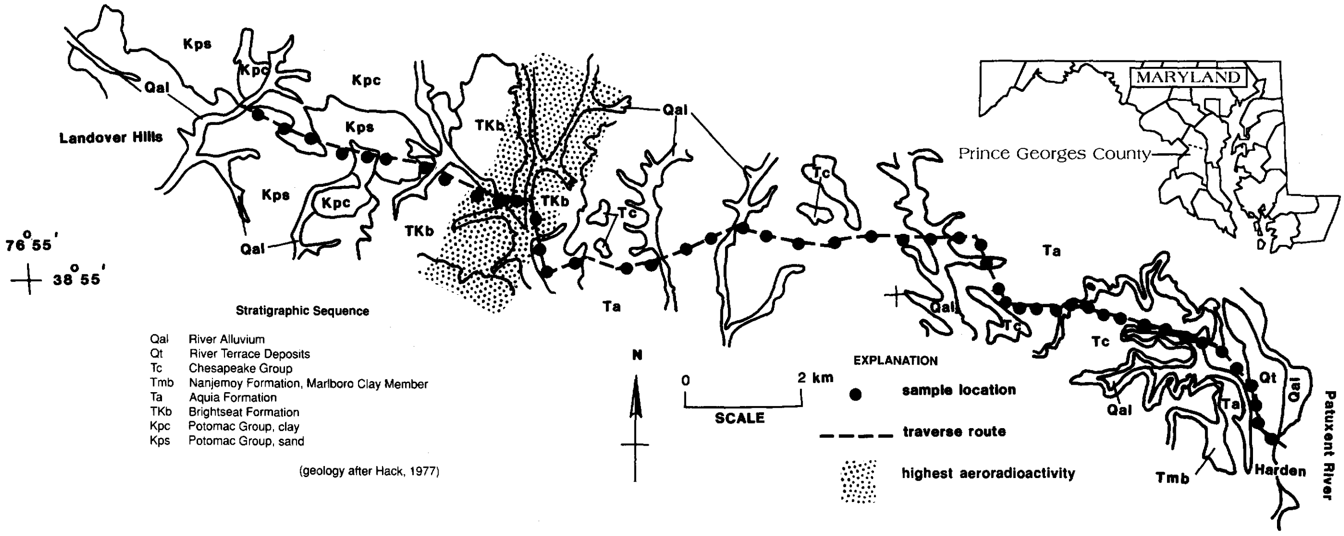

Figure 70. Location of radon soil-gas samples on part of a geologic map of Prince Georges County, Md.

and others, 1988b), and Fairfax County, Va. (Schumann and Owen, 1988), where houses with high indoor radon concentrations are located. Those previous studies indicate a relationship between high indoor radon levels where the soil-gas radon concentration exceeded about 2,000 pCi/L at a depth of 0.75 cm. Those studies also suggest that geology and soil gas were useful indicators of predictability for indoor radon. This appears to be the case with Prince Georges County as well. The radon soil-gas survey shows that soils derived from certain lithologies have the potential of supplying sufficient radon that could create an indoor radon accumulation in excess of the EPA guideline.

CONCLUSIONS

The use of a reconnaissance survey to estimate soil-gas radon potential has been tested in Prince Georges County, Md. It has revealed several Tertiary formations that have radon concentrations in excess of 2,000 pCi/L. Most of Prince Georges County, on the basis of projection from the areal distribution of the various geologic formations, appears to have low concentrations of soil-gas radon. Most tentatively, it appears that when the soil gas exceeds 2,000 pCi/L, the probability is high that the indoor radon concentrations will exceed the EPA guideline; when soil-gas concentrations are less than 300 pCi/L, the probability is low that the indoor concentrations will exceed the guideline. These levels of significance for soil-gas radon are also suggested from measurements in other areas of the United States. For example, when the data from this study, where average soil-gas and indoor radon concentrations are low, are compared to those with higher averages obtained in the Reading Prong of Pennsylvania (Gundersen and others, 1988a), a trend evolves that suggests a correlation between soil gas and indoor concentrations. The variation in home construction methods and soil permeability will probably preclude rigorous values from ever being established, but as more data are collected the limits will be refined.

ACKNOWLEDGMENTS

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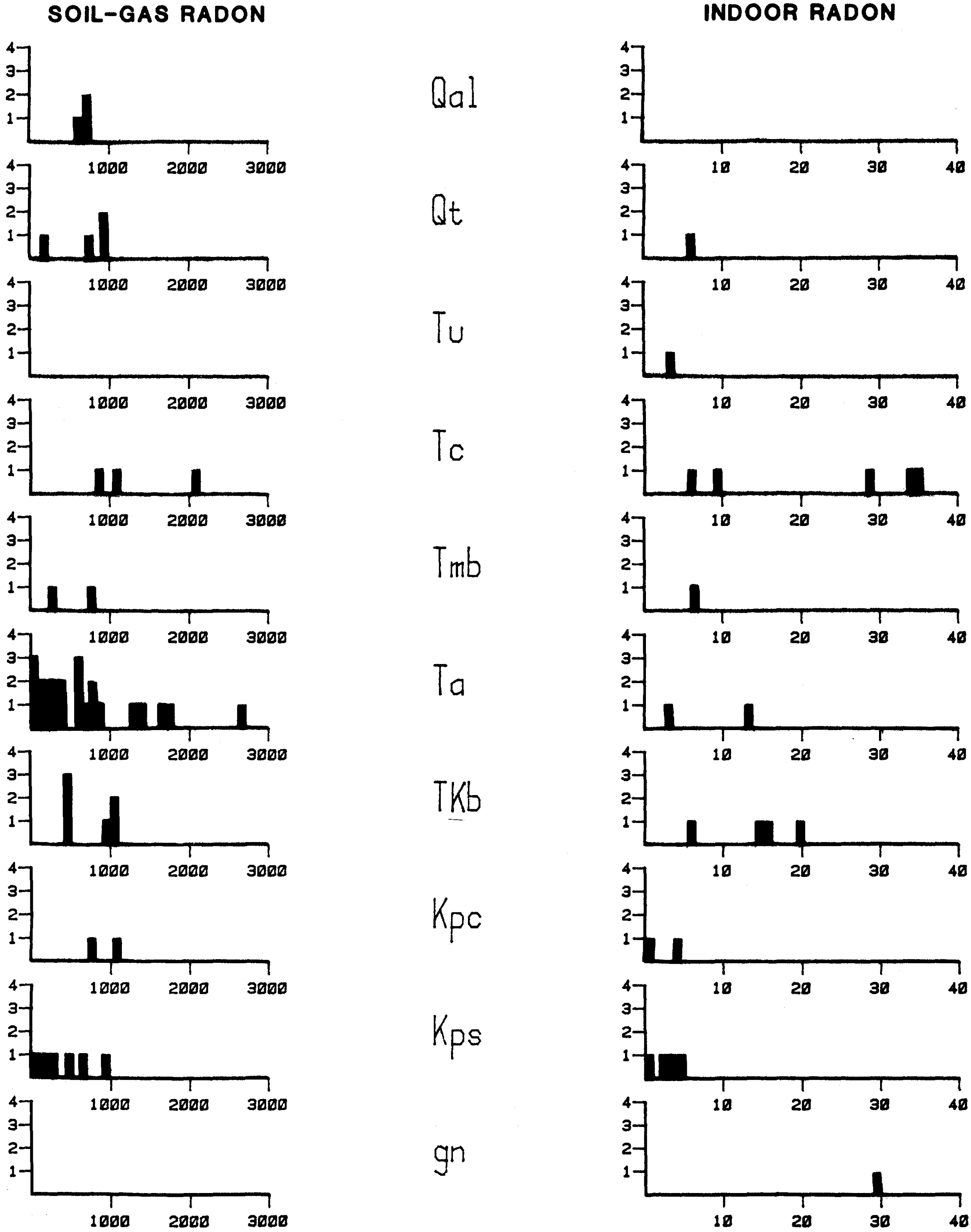

Figure 71. Frequency histograms of the radon distribution plotted by individual stratigraphic unit. The radon concentrations are in picocuries per liter (pCi/L). Tu, upland ancient river deposits; gn, gneiss and schist. No soil-gas samples were collected in those 2 units. See figure 70 for definitions of other rock symbols.

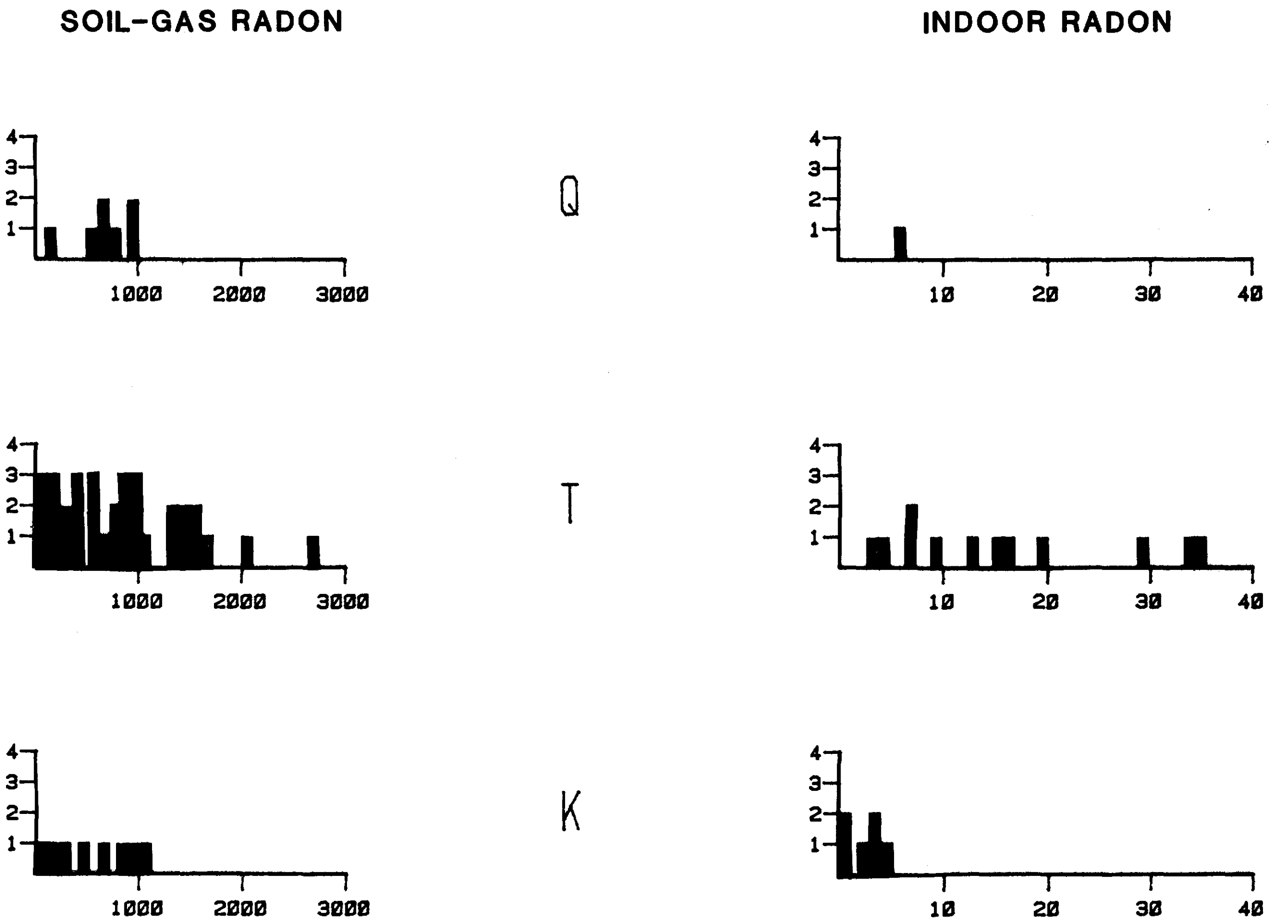

Figure 72. Frequency histograms of radon concentrations for units of Cretaceous (K), Tertiary (T), and Quaternary (Q) ages sampled during this study. The radon concentrations are in picocuries per liter.

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PART 2: STUDIES OF RADON IN WATER

SECTION 1: INTRODUCTION

A REVIEW OF THE CHEMICAL PROCESSES AFFECTING THE MOBILITY OF RADIONUCLIDES IN NATURAL WATERS, WITH APPLICATIONS

Richard B. Wanty and Robert Schoen

Abstract

Increased awareness of possible health problems associated with radon-222 in indoor air in homes has prompted U.S. Geological Survey researchers to turn their attention toward understanding the natural processes that cause radon-222 concentrations in water to be anomalous in certain areas. As part of this effort, the mobility of radon and its parent elements in ground- and surface-water systems is being studied. Many of these studies, represented by papers in this volume, have found that in addition to radon-222 in water, other radionuclides such as uranium, thorium, and radium may be present in amounts that could lead to health risks. For this reason, it is important to understand the mobility of these elements in natural systems as well. This paper is intended to provide a background for understanding the results of the studies presented in this half of the volume by providing a review of the health effects associated with the radioelements, by summarizing the chemistry of the major radioelements, and by discussing the occurrence of radionuclides in various natural environments.

INTRODUCTION

Radon-222 (^{222}Rn) is derived from the radioactive decay of uranium-238 (^{238}U), which occurs to some extent in all rocks. The ^{238}U decay series can be summarized as follows (fig. 73): ^{238}U decays to thorium-234 (^{234}Th), and in turn to protactinium-234 (^{234}Pa), uranium-234 (^{234}U), thorium-230 (^{230}Th), radium-226 (^{226}Ra), and ^{222}Rn. Radon-222 decays by several successive steps, ultimately becoming nonradioactive lead-206 (^{206}Pb). The bulk of the discussion in this paper will be limited to ^{222}Rn and its more abundant precursors, uranium, thorium, and radium. The aqueous chemistry of each of these elements is different, so an understanding of the chemical behavior of each of the elements is prerequisite to an understanding of radon sources in ground-water systems. A discussion of the precursors of radon is important also because of possible negative health effects of uranium and radium in drinking-water supplies. This paper provides brief reviews of the health effects and chemistry of radon and its parent elements and summarizes the results of a few of the studies that have been performed to date. In choosing the studies to be summarized, we have tried to represent several different geologic environments and to contrast those environments with one another. Many more studies that have been performed are not listed here. While this paper is not intended to be an exhaustive review of the current literature, this and the other papers in this volume will provide a suitable introduction to the current literature.

Uranium, thorium, and radium all occur as dissolved ionic species in solution. Uranium has several possible oxidation states, so its mobility in natural environments is directly affected by changes in oxidation potential in addition to pH and other factors such as solution composition, concentration of certain complexing agents, and so forth. Thorium and radium each have only one oxidation state and thus are less affected by oxidation-reduction processes, but both are very sensitive to pH changes and other solution properties. Radon-222, on the other hand, is a noble gas and is not ionized in solution. The aqueous concentration of ^{222}Rn is therefore most strongly affected by temperature and the presence of certain solid surfaces that may adsorb it.

Important distinctions need to be made between secular equilibrium and chemical equilibrium. Discussion in this paper concerning the aqueous geochemistry of radionuclides will focus on chemical equilibria in natural waters. Chemical equilibria are established among aqueous species and solid phases according to the laws of thermodynamics. Chemical reactions occur in response to favorable energy changes between reactant and product species that drive the reaction toward the lower energy state. The tendency for a system to attain chemical equilibrium is a driving force in natural systems. On the other hand, secular equilibrium is a condition that is attained when the rates of radioactive decay of all nuclides in a decay series are equal. The attainment of secular equilibrium in a system (or lack thereof) is merely a condition that is or is not met and depends largely on the definition of the system and whether the defined system is open or closed. For instance, the system could include the ground water only, the rocks only, or both. Once a system is defined, secular equilibrium is attained as a function of time and may be aided, hindered, or prevented by chemical processes. If secular equilibrium is attained in a system, the radioactivity of each element in

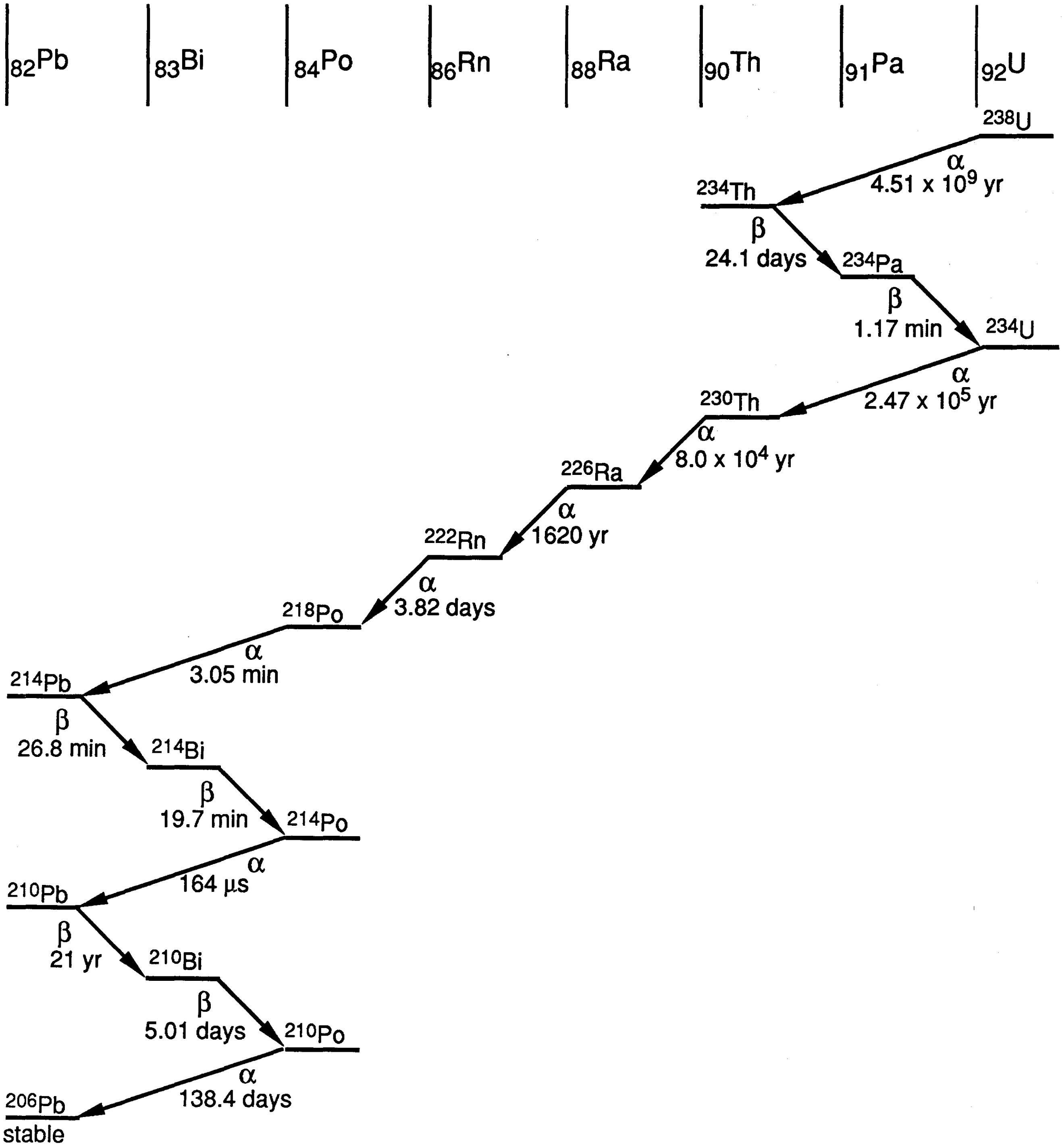

Figure 73. The uranium-238 decay series, showing the half-lives of the radionuclide elements and their modes of decay. Pb, lead; Bi, bismuth; Po, polonium; Rn, radon; Th, thorium; Pa, protactinium; U, uranium; yr, years; min, minutes; µs, microseconds; α denotes alpha decay, β denotes beta decay.

the system is said to be supported by its parents. The radioactivity of an element is unsupported if that of its parents is less. In many cases, secular equilibrium is attained between two or more successive radionuclides in a decay series and not among the entire decay series. Thus, a common comparison is drawn between ^{226}Ra and ^{222}Rn without regard to the other elements in the decay series (fig. 73).

EFFECTS OF NATURAL RADIONUCLIDES ON HEALTH

Although the U.S. Geological Survey does not perform research on the health implications of the natural radionuclides, the subject is appropriate for a brief review here for several reasons. First is the obvious concern of society to control those substances that may provide the most serious threats to human health. A full understanding of the occurrence, transport, and fate of such substances is a necessary first step in their control. Second, a knowledge of the status of toxicological and epidemiological studies permits geoscientists to focus their investigations on those occurrences, pathways, and reactions that appear to be most relevant to the health scientists' needs. Finally, in conducting investigations of elements whose health implications are still being determined, a knowledge of these studies permits geoscientists to exercise a reasonable degree of caution in their studies of these elements.

A highly readable compilation of articles on the health effects of uranium, radium, and radon in water can be found in the American Water Works Association Journal (April 1987). For more information than is contained in this review, readers should consult those articles and their cited references. A more detailed treatment of these questions can be found in the many papers included in the May 1985 volume of the journal Health Physics. Two recent publications, one by the National Research Council (1988) and the other by Nazaroff and Nero (1988), provide summaries of the more recent literature.

Uranium

Natural uranium consists of the three isotopes ^{238}U, ^{235}U, and ^{234}U in the proportion 99.28 percent, 0.72 percent, and 0.005 percent, respectively. Both ^{238}U and ^{235}U have half-lives of billions to hundreds of millions of years. The half-life of ^{234}U is only 2.5×10^5 years, however, so the radioactivities from both ^{238}U and ^{234}U in natural uranium are equivalent (Aieta and others, 1987, p. 145). A concentration of 100 μg/L (micrograms per liter) of natural uranium yields a radioactivity of 67 pCi/L (picocuries per liter) if the ^{238}U and ^{234}U isotopes are in radioactive equilibrium (Wrenn and others, 1985, p. 609). It is known that uranium, when ingested, is a bone seeker similar to radium. There are, however, no unequivocal reports of ingested natural uranium causing cancer as radium has (National Research Council, 1988, p. 298). The health aspects of natural uranium appear to be entirely related to its chemical toxicity as a heavy metal and specifically to its nephrotoxicity (ability to cause kidney damage). The difficulty in demonstrating that natural uranium is radiotoxic is probably related to the fact that the levels of natural uranium that would cause nephrotoxicity provide very low doses of alpha radiation to the bone and bone marrow. Wrenn and others (1985, p. 611) call for further research on the levels of natural uranium in water and in the diet, in different geographic regions, to determine the amounts of uranium normally held in the human body.

Radium

On the basis of the experience of the radium-dial painters, the principal health aspect of ingested radium has long been known to be its induction of bone cancer (National Research Council, 1988, p. 177). Radium, an alkaline-earth cation, readily proxies for calcium in bone structure. As a bone seeker, it might be expected that radium would induce leukemia through its radiation effects on bone marrow; however, this effect appears to be insignificant compared to the number of bone sarcomas radium has caused (Mays and others, 1985, p. 637). Interestingly, ^{226}Ra, unlike ^{228}Ra or ^{224}Ra, seems to cause carcinomas of the paranasal sinuses and the mastoid air cells. It has been suggested that these head carcinomas are related to the decay of ^{226}Ra to ^{222}Rn and the accumulation within the head cavities of ^{222}Rn gas and its progeny (Mays and others, 1985, p. 636), and the National Research Council (1988, p. 238) concurs with this suggestion. Radium-228 decays to ^{224}Ra, but ^{220}Rn, the daughter of ^{224}Ra, has a half-life of only 54.5 seconds. This short half-life may prevent ^{220}Rn from reaching and accumulating in head cavities. On the basis of the ingestion of the current Environmental Protection Agency Maximum Contaminant Level of 5 pCi/L of ^{226}Ra plus ^{228}Ra, the number of excess induced cancers in the lifetimes of 1 million persons would be 9 bone sarcomas and 12 head carcinomas from 5 pCi/L of ^{226}Ra, 22 bone sarcomas and 0 head carcinomas from 5 pCi/L of ^{228}Ra, and 1.6 bone sarcomas and 0 head carcinomas from 5 pCi/L of ^{224}Ra (Mays and others, 1985, p. 644). The National Research Council (1988, p. 238) calls for further research to elucidate the relation between sinus and mastoid carcinomas and the radon released during decay of radium in bone.

Radon

Undoubtedly, radon has been present in the atmospheres of homes since the first humans constructed shelters. This naturally occurring radioactive gas, found in enriched quantities in ground water, natural gas, and soil gas, can enter a house through use of ground water, burning of natural gas, leaks of soil gas into the substructure, and from the building materials themselves.

Although naturally occurring radon has been present in building structures for as long as structures have been built, it is only in the last decade that relatively high values have been discovered in homes not otherwise associated with mining and milling activities. Before this, it was believed that only structures built on or with uranium mill tailings would experience abnormally high levels of radon. It is now known that the normal low levels of uranium, radium, and radon in rocks and soils can, under suitable conditions, cause anomalous levels of radon in a home's atmosphere (Tanner, 1986, p. 5).

On the basis of extrapolation from the experiences of uranium miners who were exposed to high levels of radon for several years, and who subsequently developed lung cancer, the Environmental Protection Agency has estimated the risk of an individual's developing lung cancer from living in a home that has low to moderate levels of radon. These studies have resulted in the Environmental Protection Agency's establishing an annual-average living-space concentration of 4 pCi/L of radon in the living quarters of a house as a guideline above which the risks increase and below which reducing the radon levels may be excessively expensive or impossible (Nazaroff and Nero, 1988, p. 35–36). Radon levels are greatest generally in the winter, when windows and doors are shut and often tightly sealed with weatherstripping. As more outside air leaks into a house, the radon concentration decreases, because ordinary outdoor air has a very low concentration of radon (0.1 to 0.15 pCi/L; National Council on Radiation Protection, 1984, p. 25).

It is often assumed that radon causes lung cancer because, as a gas, it is inhaled into the lungs and while there, undergoes radioactive emission of an alpha particle and a gamma ray that cause carcinogenic damage to lung cells. In fact, most of the radon gas that one inhales is exhaled again within a few breaths. Because radon's half-life is 3.82 days, and because only small numbers of radon atoms are present in natural atmospheres, few if any radioactive disintegrations occur while the radon is in the lungs (Cothern, 1987, p. 154). At very high concentrations of radon, beyond naturally occurring levels, sufficient decay could occur during radon's brief residence in the lungs to make it a health concern.

Radon's deleterious health effects are caused by its continued presence in a confined space, such as a home or an individual room, where the products of its relatively slow, infrequent disintegrations can accumulate (fig. 73). Radon-222 decays into a series of solid daughter radioisotopes. The first four, ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po, have combined half-lives of less than 50 minutes. As solid, charged particles, these metal ions adsorb almost instantly to any surfaces or dust particles in the atmosphere of the home, although a significant fraction can remain unattached. When these dust particles and unattached ions are inhaled, they tend to lodge in the lungs, whose tissues are then subjected to the relatively rapid delivery of radioactive disintegrations during the combined short half-lives.

Because of the accepted relation between lung cancer and radon exposure in the Colorado Plateau uranium miners, the focus of radon research has been on the inhalation route of radon and its progeny. Relatively little work has been done on the ingestion of radon in drinking water, where its concentration can be high. It is generally assumed that the primary health effect of radon-rich water is through its contribution to the atmosphere of the home by degassing.

Studies of the degassing of radon from water into the atmosphere of a home have produced values of the mean transfer factor, f, ranging from 1.0 to 2.5×10^{-4}, in the equation

$$C_a = f\, C_w,$$

where C_a is the radon concentration in air and C_w is the radon concentration in water (Nazaroff and Nero, 1988, p. 132).

This equation demonstrates that the concentration of radon in a home attributable to degassing of radon from the water will be only about 1/10,000 of the water concentration. The few studies that have evaluated this transfer coefficient have found that the major sources of uncertainty lie in the amount of house volume to ascribe to each occupant and in the average air-exchange rate to use. It is also implicitly assumed that the degassed radon and its progeny are distributed uniformly throughout the house volume and not concentrated at the point of degassing. Thus, it can be expected that such wide variations to the "1 in 10,000 rule" may be found in individual homes as to make it impractical for the individual to rely on its accuracy.

With respect to water-borne radon, the National Research Council (1988, p. 80) recommends further research on the possible long-term health effects of ingesting radon-rich water. Nazaroff and Nero (1988, p. 143) note the need to improve the estimate of the parameters that determine the transfer factor, f. Finally, in determining the concentration of radon and its progeny in homes using radon-rich water, it will be useful to measure the spatial distribution with time in order to test the assumption of uniform distribution throughout the house volume.

INORGANIC CHEMISTRY OF RADON AND ITS PARENT ELEMENTS

The geochemical processes that affect the mobility and fixation of the parent elements of ^{222}Rn in the natural environment are summarized in this section. Natural processes that limit the mobility of these elements include precipitation, coprecipitation, adsorption, and ion exchange. In this section, we review the chemistry of uranium, thorium, radium, and radon in the context of these processes.

Uranium

Uranium may be present in natural waters in three formal oxidation states, namely IV, V, and VI. In general,

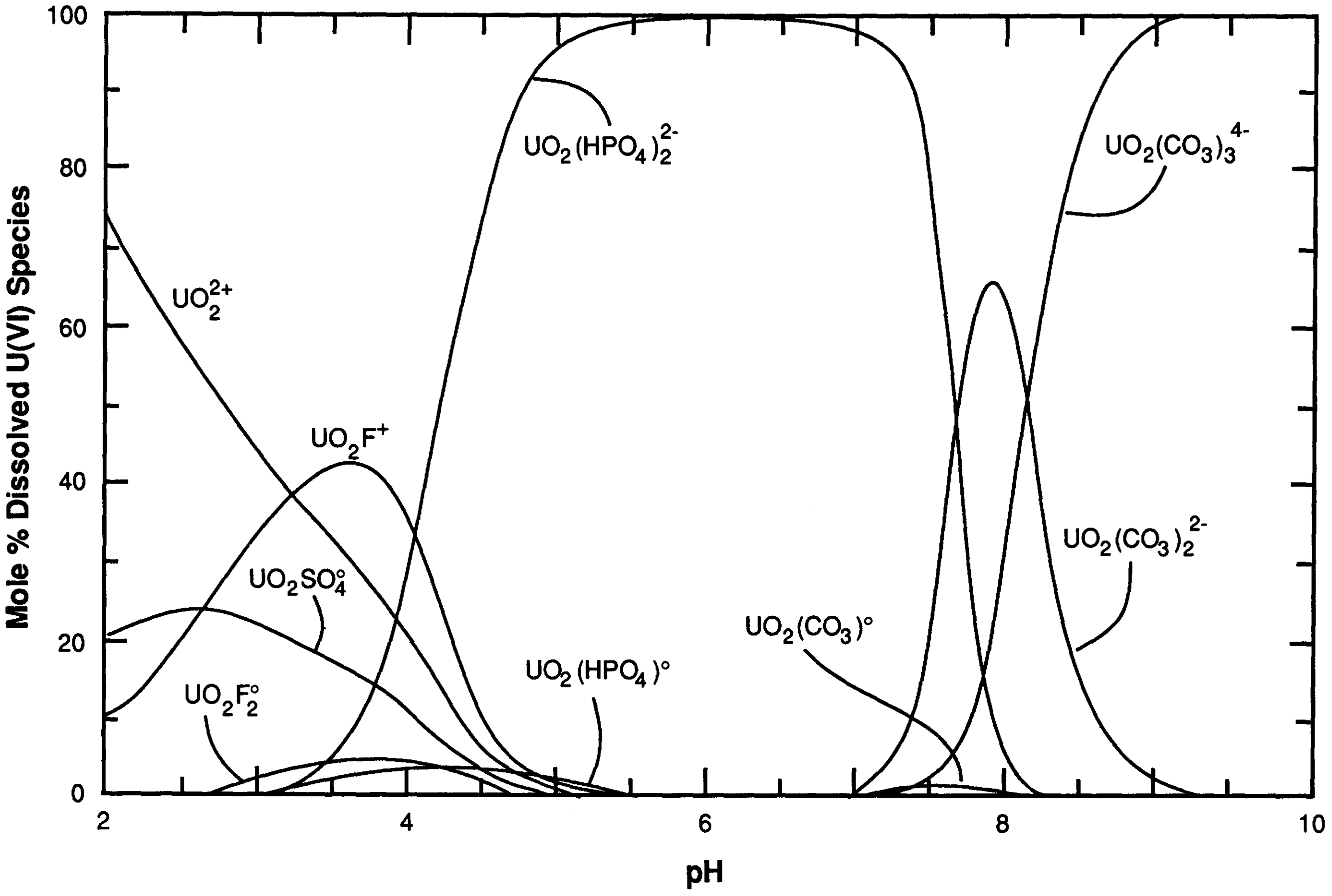

Figure 74. The distribution of inorganic aqueous species of uranium as a function of pH (modified from Langmuir, 1978). In the calculation of the diagram, the following aqueous concentrations were used: $PCO_2 = 10^{-2.5}$ atm; $\Sigma F = 1.6 \times 10^{-5}$ *m*; $\Sigma Cl = 2.8 \times 10^{-4}$ *m*; $\Sigma SO_4 = 1.0 \times 10^{-3}$ *m*; $\Sigma PO_4 = 1.1 \times 10^{-6}$ *m*; $\Sigma SiO_2 = 5.0 \times 10^{-4}$ *m*.

uranium is most soluble in oxidizing water. Uranium(VI) occurs in solution as the uranyl ion, UO_2^{2+}, or some complex thereof. The uranyl ion is the most soluble form of uranium and forms strong complexes with carbonate and biphosphate at pH values greater than approximately 4 (Hostetler and Garrels, 1962; Langmuir, 1978; Dongarra and Langmuir, 1980). Complexes with fluoride or sulfate may be important below pH 5 (fig. 74). There is also evidence that uranyl may be effectively complexed by humic acids at pH values between 3.5 and 7 (Kribek and Podlaha, 1980), although Shanbag and Choppin (1981) present other evidence that the uranyl-humic acid complex may be relatively insoluble and that humic acid in the solid state would effectively scavenge uranyl from solution.

Uranyl minerals are seldom precipitated from aqueous solutions in natural environments. Uranyl hydroxide (schoepite; $UO_2[OH]_2 \cdot H_2O$) is quite soluble in natural waters, but uranyl vanadate minerals such as carnotite ($K_2[UO_2]_2[VO_4]_2$) or tyuyamunite ($Ca[UO_2]_2[VO_4]_2$) may precipitate if the vanadate and potassium or calcium concentrations are high enough (see Langmuir, 1978).

In oxidizing waters, uranyl ions may be adsorbed to varying extents onto solid substrates such as iron oxides (Hsi, 1981; Ames and others, 1983a; Hsi and Langmuir, 1985), organic material (Moore, 1954; Breger and others, 1955; Schmidt-Collerus, 1967; Leventhal and others, 1986), clay minerals (Ames and others, 1983b), and micaceous minerals (Ames and others, 1983c). The adsorption of the uranyl ion onto these substrates is strongest at near-neutral pH and weaker at much higher and lower values. At low pH, hydrogen ions effectively compete for surface sites, while at higher pH, carbonate complexing of uranyl leads to desorption.

The uranyl ion is stable only in oxidizing systems and is reduced to U(V) or U(IV) under progressively reducing conditions. The reduction of U(VI) occurs readily in the presence of H_2S (Mohagheghi, 1985), and the reaction is strongly catalyzed by mineral surfaces. The end product of this reaction is the reduced uranium oxide mineral, uraninite (UO_2), or some amorphous form with a similar stoichiometry (fig. 75). In natural waters that have significant silica concentrations, coffinite ($U[SiO_4]_{4-x}[OH]_{4x}$) may precipi-

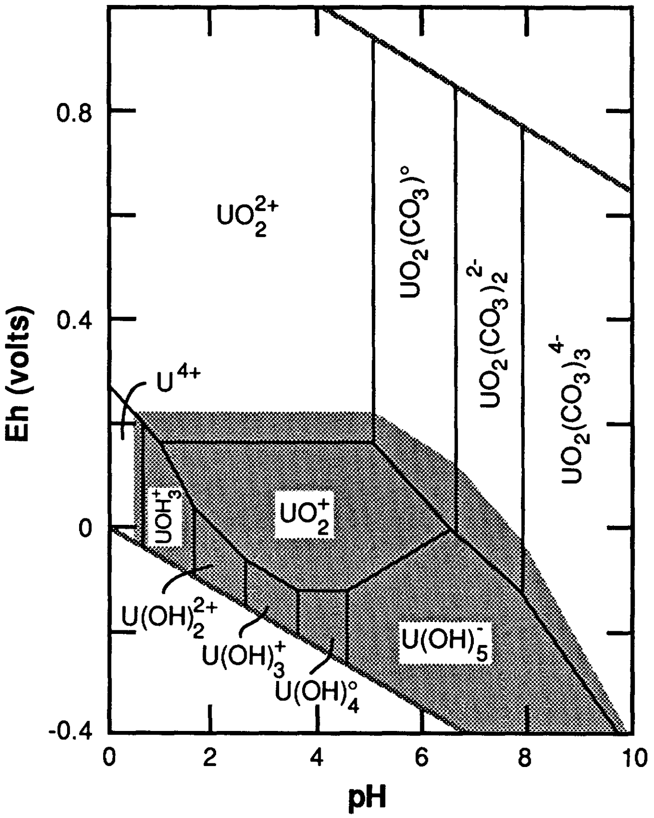

Figure 75. Distribution of inorganic uranium species in Eh-pH space for the system U-O-C-H at 25 °C and $10^{-2.5}$ atm CO_2. The stippled area represents the stability field for uraninite at a total uranium concentration of 10^{-6} M (modified from Langmuir, 1978).

tate instead (Goldhaber and others, 1987). Reduction and precipitation of uranium may lead to a significant accumulation of uranium if there is a sharp transition from oxidizing to reducing conditions in an aquifer. In order to get a significant accumulation, this transition zone must be stable for geologically significant periods of time. Conditions such as these are met in many instances in the Western United States, particularly on the Colorado Plateau, the Wyoming intermontane basins, and the south Texas coastal plain. In many of these cases, uranium may have accumulated to levels of several percent of the rock. These uranium deposits are of particular interest because uranium accumulations in rocks are known to produce anomalous radon concentrations in ground waters (Chatham and others, 1981; Wanty and Gundersen, 1987). Nash and others (1981) summarize many known types of uranium deposits and give descriptions of their origins.

Thorium

Thorium has only one formal oxidation state in nature, namely thorium[IV] (Zebroski and others, 1951). Aqueous thorium(IV) is strongly hydrolyzed in the series $Th(OH)_n^{4-n}$, where $n=0-4$ (Langmuir and Herman, 1980); the species $Th(OH)_4^\circ$ dominates above pH of approximately 4.5. Strong complexes are known to exist between thorium and biphosphate, oxalate, and EDTA at pH values commonly found in nature (Zebroski and others, 1951; Waggener and Stoughton, 1952; and Langmuir and Herman, 1980).

Thorium is generally considered to be insoluble in most natural waters. It occurs in the solid form most commonly as a minor constituent in solid solution in phosphate, oxide, or silicate minerals. It is the major component in the relatively rare minerals thorianite (ThO_2) and thorite ($ThSiO_4$). The most important commercial source of thorium is from monazite ([Ce, La, Nd, Th] PO_4), which is concentrated in stream sediments and reworked beach sands (Rankama and Sahama, 1950). All these minerals are quite insoluble; even in solutions that have very high concentrations of complexing agents, calculated solubilities for thorianite rarely exceed 1 ppb Th above pH 5 (Langmuir and Herman, 1980).

Studies indicate that thorium is adsorbed strongly onto kaolinite (Riese, 1982) and α-quartz (James and Healy, 1972; Riese, 1982) at pH values above about 4. Riese (1982) found that thorium adsorption was inhibited by increasing the sulfate concentration, but even then adsorption was virtually complete at pH values higher than 5.5. Thus, if thorium is not present within the crystal structure of some mineral, it probably is adsorbed onto an oxide or silicate surface; only vanishingly small amounts are present ordinarily in aqueous solution. For more details about the geochemical behavior of thorium, the reader is referred to Langmuir and Herman (1980).

Radium

Like thorium, radium has only one oxidation state in natural systems, in this case, radium(II). But here the similarities end; the chemistry of radium is much different than that of thorium in natural systems because radium has much less of a tendency to hydrolyze. Radium behaves in many respects like other alkaline earth elements (Group IIa). It forms strong complexes with sulfate and carbonate (fig. 76) and weaker complexes with nitrate and chloride (Langmuir and Riese, 1985). Complexing of radium by organic materials is not well characterized, although weak 1:1 complexes with several organic acid species have been detected by Schubert and others (1950).

Radium mobility is limited in natural systems by several processes, chiefly coprecipitation, adsorption, and ion exchange. Although the solids $RaSO_4$ and $RaCO_3$ are sparingly soluble (pK_{sp} values of 8.30 and 10.26, respectively, according to Langmuir and Riese, 1985), they rarely form because radium concentrations are so restricted by

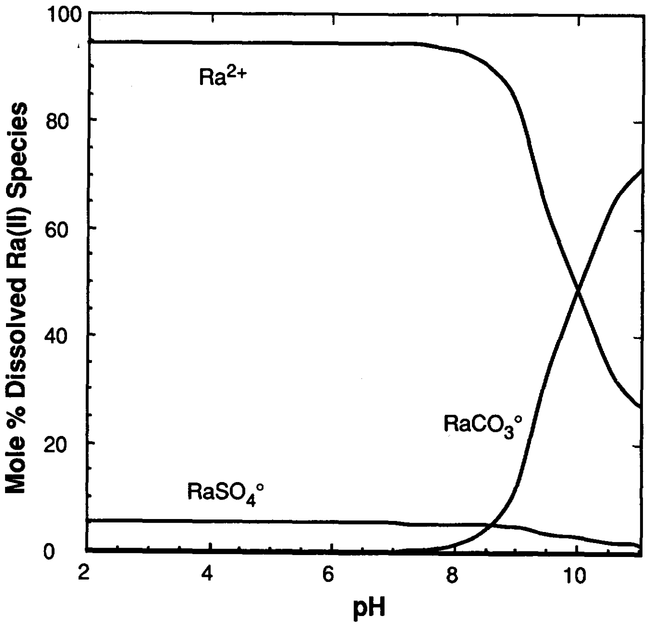

Figure 76. The distribution of inorganic aqueous radium species as a function of pH for the following conditions: $\Sigma CO_3 = 1.0 \times 10^{-2}\ m$; $\Sigma SO_4 = 1.0 \times 10^{-4}\ m$; $\Sigma Cl = 1.0 \times 10^{-4}\ m$. No chloride complexes are sufficiently significant to plot on the diagram.

other processes that natural waters are seldom oversaturated with respect to these phases. If radium precipitates in a solid phase, it usually does so in solid solution in barite (Doerner and Hoskins, 1925), where it substitutes for barium. Radium may coprecipitate in calcite as well (Tanner, 1964a).

Radium is strongly adsorbed by mineral surfaces, including quartz, kaolinite, and other clay minerals (Riese, 1982; Ames and others, 1983c,d). It is also strongly adsorbed on iron(III) oxyhydroxides (Ames and others, 1983a), and Levinson (1980) argues that radium that is *ad*sorbed on a precipitating ferric oxyhydroxide may be *ab*sorbed as the ferric oxyhydroxide continues to grow. For a detailed review of the chemistry of radium, the reader is referred to Weigel (1977).

Radon

Radon mobility in natural waters is affected principally by physical, rather than chemical, processes. Being a noble gas, radon is not ionized in solution, nor does it precipitate in solid phases. It may be adsorbed to a limited extent by organic material, especially charcoal (Lowry and Brandow, 1985), but there is little evidence available to suggest that it is adsorbed to any great degree by other surfaces.

Radon is soluble in water; the solubility is sensitive to temperature. Solubilities for the temperature range 0 to 100 °C are given in table 23. At 10 °C, the solubility of radon is approximately 61 times that of oxygen on a mass basis (Lowry and Brandow, 1985). Because the partial pressure of radon in air is ordinarily very low, radon is readily degassed from water that is exposed to air. Thus, radon concentrations found in surface waters are in general several orders of magnitude lower than those in ground waters. In addition, domestic ground-water supplies that are high in radon may contribute significantly to indoor airborne radon. A thorough review of radon chemistry is provided by Weigel (1978).

Table 23. The solubility of radon in water from 0 to 100 °C

[The data shown are from Weigel (1978). Units are moles of radon per liter with a radon partial pressure of 1 atm]

Temperature (Celsius)	Radon solubility
0	0.51_0
5	$.42_0$
10	$.35_0$
20	$.25_5$
30	$.20_0$
40	$.16_0$
50	$.14_0$
60	$.12_7$
70	$.11_8$
80	$.11_2$
90	$.10_9$
100	$.10_7$

SUMMARY OF SELECTED STUDIES

The mobility, distribution, and chemical form of U, Th, Ra, and Rn in natural surface- and ground-water systems will be determined by chemical processes as described previously and by physical properties of the natural system (for instance, temperature, pressure, flow rate, porosity, permeability). In this section we will describe some previous studies on radionuclides in ground waters in sandstone and crystalline aquifer systems. Despite the differences in application of these studies, the chemical processes that must be considered remain the same. Thus, the studies of broadest interest will be those that examined the overall water chemistry as well as the abundances of the radionuclides.

As seen in the previous section, the mobility of each of the radioactive parent elements of radon is affected to different degrees by different chemical processes. It is through an understanding of the chemical behavior of the radionuclides that the natural systems can be better understood. In the ensuing discussion, radionuclide mobility in specific natural systems will be discussed, and one system will be compared and contrasted to the other. This discussion will be separated according to aquifer rock type, so sedimentary aquifer systems will be divided from fractured-rock aquifer systems. These two types of systems have profoundly different hydrologic regimes. The differences in hydrology are manifested in differences in radionuclide mobility, particularly in the differential mobilities between ^{226}Ra and ^{222}Rn.

Sedimentary Aquifer Systems

Many studies have been performed on radionuclide migration in sedimentary aquifer systems, some related to health aspects, others related to exploration for subsurface uranium deposits. We give here a brief summary of selected studies, stressing the dependence of radionuclide mobility on natural chemical processes active in these aquifer systems.

Many studies have focused on radon distribution in ground waters. The results of these studies have been applied to the investigation of geologic structure, as in the study of Mazor (1962), or to ground water geochemistry, as by Tanner (1964a). Radium-226 and ^{222}Rn distributions in ground waters near Great Salt Lake, Utah, were studied by Tanner (1964a). He found that the waters were for the most part out of secular equilibrium and that ^{222}Rn concentrations were much higher than would be expected, given the aqueous ^{226}Ra concentrations. He proposed that the ^{226}Ra was for the most part locally derived and that it was likely adsorbed on iron-oxide surfaces. In contrast, Rama and Moore (1984) proposed that radon in water could be derived from radium within grains in the rocks and that an extensive network of minuscule pores serves as a migration pathway for the radon thus produced.

Uranium is immobile in reducing systems; thus, organic-rich black shales tend to be more enriched in uranium than do coarse-grained rocks (see Krauskopf, 1967). Uranium may be immobile in reduced systems, but its daughters often are mobile. In a study of radionuclide geochemistry in the Newark basin of New Jersey, Zapecza and Szabo (1987) found high gross alpha-particle activities in ground waters near organic-rich black shales and determined from borehole geophysical tests that the black shales were anomalously radioactive. In a companion study, Szabo and Zapecza (1987) showed that dissolved uranium concentrations were low in the reducing waters associated with the black shales, but dissolved ^{226}Ra concentrations were higher, accounting for most of the gross alpha-particle activity. Further, they found low ^{226}Ra concentrations in sulfate-rich waters, as would be expected if radium sulfate solids formed. In agreement with the findings of Tanner (1964a), the Newark basin studies found that aqueous radon activities were much higher than those of radium, again leading to the conclusion that aqueous radon is derived from radium in the solid phase.

In many cases, ^{226}Ra concentration is low (less than 1 pCi/L) because of adsorption or precipitation processes. An exceptional area was reported by Cecil and others (1987) for the Chickies Formation (Late Proterozoic) in southeastern Pennsylvania. Here, the ^{222}Rn activities are not particularly high (usually less than 10,000 pCi/L), although they are always greater than the ^{226}Ra levels. The exceptional aspect of the Chickies Formation ground waters is the high ^{226}Ra activities and, in most wells, the even higher ^{228}Ra activities. Cecil and others attributed the high total radium levels to low pH values in the ground water and the concomitantly lesser degrees of adsorption of radium onto the aquifer materials. Their research is continuing to determine the source of this radium, and the reason for the anomalously high ^{228}Ra/^{226}Ra ratios (Cecil and others, this volume).

Chatham and others (1981) and Wanty and others (1987) studied ground-water geochemistry in a sandstone aquifer that hosted several uranium deposits in south Texas. They found that dissolved uranium was highest in updip, oxidizing waters near the recharge zone and that it decreased abruptly downdip as dissolved oxygen was depleted in the ground water. This abrupt decrease in dissolved uranium corresponded to the location of the uranium deposits in the area. Ground waters in contact with the uranium deposits were low in sulfate and high in ^{226}Ra and ^{222}Rn, but other ground-water samples from the area contained significantly lower ^{226}Ra and ^{222}Rn activities. Thus, in the south Texas study area, very high uranium levels in rocks (tenths of a percent uranium by weight) were required to produce significant levels of ^{222}Rn (thousands of picocuries per liter) in the associated ground water.

In summarizing the sedimentary aquifer systems that have been studied, we must note that ground-water concentrations of uranium are highest in oxidized waters, usually near the recharge zone. In reduced waters, uranium concentrations are much lower. Thorium in ground water is rarely measured but has been found to be low. Radium-226 activities may be high (several picocuries per liter) in some waters, particularly in the absence of sulfate. Radon-222 activities usually are much higher than would be predicted from the aqueous ^{226}Ra activities, and ^{222}Rn highs in the ground water usually correspond to nearby enrichments of its parent elements in the rocks.

Fractured-Rock Aquifer Systems

There have been many studies of radionuclide concentrations in crystalline rocks, particularly in the New England States, where extremely high ^{222}Rn activities have been found in some ground waters. A summary of natural radioactivity in New England by Hall and others (1987) shows a wide range of uranium contents in the rocks and a complex distribution of that uranium within the rock fabric. Nevertheless, Hall and others (1987) show a map of lithologic groups that have the potential to produce large quantities of radon that could be released to the ground water. In Maine, Brutsaert and others (1981) showed that very high ^{222}Rn concentrations are present in ground waters from granitic rocks and from some high-grade metamorphic rocks. Hall and others (1985) found similar relationships in New Hampshire, where glacial deposits, low-grade metamorphics, and basic intrusive rocks had low ground-water ^{222}Rn activities, and high-grade metamorphics and granites had very high ground-water ^{222}Rn activities.

In a study of ground waters from two-mica granites in Maine and New Hampshire, Wathen (1986) found very high ^{222}Rn concentrations, up to 145,000 pCi/L. Through the use of autoradiographs, he showed that uranium was most strongly concentrated along boundaries of weathered biotite grains, and in sericitized plagioclase grains, the indication being that uranium was adsorbed on these surfaces. Thus, as ^{238}U in these rocks decayed, the daughter elements would be close to the solution-solid interface, and ^{222}Rn would be available to the water. Asikainen and Kahlos (1979) provided isotopic evidence from well waters collected near Helsinki, Finland, that showed that the dissolved ^{222}Rn was derived from parent elements localized along fracture walls, substantiating Wathen's conclusions.

The previous two studies showed a common result in that the ground-water ^{222}Rn levels were unsupported by the parent elements. A study in two different areas of the Reading Prong in eastern Pennsylvania by Wanty and Gundersen (1987; 1988) agrees with these results. In virtually every water sample, the ^{222}Rn activity was almost completely unsupported by the parent elements. In this case, autoradiographs of the rocks showed a concentration of radioactivity near grain margins and fracture boundaries. In addition, analyses of the aquifer rocks (Gundersen and others, 1988) show that the ^{238}U and ^{226}Ra contents of the rocks are high enough to support the ^{222}Rn activity in the water. Thus, although the water itself is not in secular equilibrium, the entire water-rock system is.

Two studies have recently been published from North Carolina (King and others, 1982) and South Carolina (Loomis, 1987). Both of these studies extended across the boundary between crystalline rocks of the Appalachian Piedmont and sediments of the Atlantic Coastal Plain and arrived at substantially similar conclusions. In both States, the metamorphic and igneous rocks of the Appalachian Piedmont had significantly higher ground-water radon (one to three orders of magnitude difference) than did the coastal plain sediments. Similarly in both States, the dissolved ^{222}Rn concentrations were unsupported by dissolved ^{226}Ra, regardless of rock type.

Hydrologic factors probably are of importance in explaining the significant differences found between fractured-rock aquifers and sedimentary aquifers. Significant hydrologic differences exist between these two classifications of aquifers, especially in ground-water flow rates, residence times, the volumetric ratios between water and rock, and the surface area of rock per volume of water. These hydrologic properties define and constrain the physical characteristics of the aquifers. These physical properties may in turn affect the mobility of ^{222}Rn or its parent radionuclides. For instance, Hall and others (1985) found an inverse correlation between dissolved radon activity and well yield. Rumbaugh (1983) noted the same relationship in ground waters of the Reading Prong, Pa., and ascribed this relationship to a simple theoretical model involving aquifer transmissivity and fracture aperture. He found that increased average fracture aperture, and attendant higher well yields, led to lower aqueous ^{222}Rn concentrations. Conversely, smaller average fracture apertures associated with lower well yields had consistently higher dissolved ^{222}Rn levels. Perhaps the best direct comparison between sedimentary and fracture-flow aquifers comes from the results of King and others (1982) and Loomis (1987). Both of these studies showed that average dissolved ^{222}Rn activities were lower in the sedimentary aquifers. The relatively higher water/rock ratio of the porous sedimentary aquifers relative to fractured crystalline ones may be the reason that sedimentary aquifers have generally lower ^{222}Rn concentrations (Wanty and Gundersen, 1987).

SUMMARY AND CONCLUSIONS

The mobility of radionuclides in ground waters is controlled primarily by chemical and physical processes as constrained by the aquifer rock type. In general, dissolved uranium is likely to be found in greater quantity in oxidized ground waters hosted by granitic rocks, or in some cases in high-grade metamorphic rocks. Uranium accumulations in sedimentary rocks are relatively rare but occur at the transition between oxidizing and reducing conditions along a ground-water flow path. In the sedimentary aquifer systems, highest dissolved uranium concentrations usually are found near the outcrop, where oxidizing recharge waters are percolating into the ground. Thorium is strongly hydrolyzed and nearly insoluble in all waters. Radium may be quite soluble in ground waters, particularly in the absence of sulfate. Nevertheless, the mobility of radium is impeded by its relatively strong adsorption onto mineral surfaces such as ferric oxyhydroxides and clays. Radon-222 is extremely soluble in water and not easily adsorbed on most mineral surfaces, so its concentration in ground water is commonly higher than that of its parents, and secular equilibrium between ^{222}Rn and its parents in water is rarely realized. On the other hand, the overall water-rock system may attain secular equilibrium.

A brief review of previous studies of radionuclide migration in a variety of aquifer rock types indicates that ground waters in high-grade metamorphic rocks and granitic igneous rocks generally have the highest dissolved ^{222}Rn activities. Ground waters in low-grade metamorphics and sedimentary rocks are generally lower in ^{222}Rn. One notable exception is the case where uranium has accumulated in sediments at the interface between oxidizing and reducing conditions.

Development of a more complete understanding of the movement of radionuclides in ground water requires more comprehensive studies. Integrated geologic, hydrologic, and geochemical approaches are required to characterize the major variables of the natural system under study. Such studies would be able to answer the greatest range of questions concerning sources and sinks of radionuclides and mechanisms for their mobility in ground-water systems.

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RADIONUCLIDES IN GROUND WATER, ROCK AND SOIL, AND INDOOR AIR OF THE NORTHEASTERN UNITED STATES AND SOUTHEASTERN CANADA—A LITERATURE REVIEW AND SUMMARY OF DATA

Robert T. Paulsen[1]

Abstract

This report briefly summarizes major investigations of the occurrence and concentrations of uranium, radium-226, and radon-222 in ground water, rock and soil, and indoor air in the six New England States, New York, and southern Ontario since the 1950's. It includes tables of concentration data for each State and describes the major results of the 155 studies cited.

The first studies of radionuclides in the Northeast began in the 1950's and 1960's as uranium exploration. Through the 1970's and 1980's, the focus of research changed from uranium exploration to the occurrence and concentration of uranium, radium-226, and radon-222 in the ground water, rock and soil, and indoor air. This change in focus was prompted by the discovery that exposure to alpha-radiation-emitting daughter products from the radioactive decay of uranium was possibly a serious threat to human health. Radon-222 in ground water has been measured in concentrations greater than 1,000,000 picocuries per liter in Maine; the national average is 350 picocuries per liter. Uranium content of bedrock in the Northeast ranges from 0.001 to 0.400 percent by weight. Radium-226, the radioactive parent of radon-222, has been measured in various soils in New York and Ontario; concentrations ranged from 0.2 to 7.0 picocuries per gram. Radon-222 in indoor air has been measured in concentrations greater than 1,200 picocuries per liter in Maine. The Environmental Protection Agency suggests precautionary action be taken at an indoor air concentration of 4.0 picocuries per liter.

INTRODUCTION

The occurrence and distribution of the radionuclides uranium, radium, and radon-222 (^{222}Rn) in the environment have become prominent research topics in the Northeastern United States as well as nationally. Research on ^{222}Rn in the environment has roots in the exploration for uranium, and some of the earliest exploration for, and are research on, the distribution of radionuclides in the ground water and air was done in New England during the early 1960's. The deleterious health effects of inhalation or ingestion of radionuclides, especially ^{222}Rn daughter products, are receiving national attention in the media and arousing concern. Recently, radon has been noted as the most widespread naturally occurring contaminant measured in drinking water, and the health risks from exposure to radon daughter products may be far in excess of any posed by manmade organic compounds that are currently regulated (C.R. Cothern, Office of Drinking Water, Environmental Protection Agency, oral commun., 1987). In response to the estimated 5,000 to 20,000 annual cancer fatalities attributed to exposure to radon daughter products (Environmental Protection Agency, 1986), the public, legislators, and the scientific community are asking (1) which radionuclides they should be concerned with, (2) where they come from and where are they most prevalent, and (3) how they can be removed to lessen the health risks associated with them.

This report addresses the source and prevalence of radionuclides and summarizes the research into their occurrence and distribution in ground water, rocks and soil, and indoor air in New England, New York, and southeastern Ontario (fig. 77). The research summaries are grouped by State, and the section for each State contains tables of reported radionuclide concentrations in ground-water samples and ^{222}Rn concentrations in indoor air. This report summarizes the available research but does not provide interpretations of the cited data.

HISTORICAL PERSPECTIVE

Although research into the occurrence and distribution of ^{222}Rn and other radionuclides in the environment may appear to be a relatively new topic, research on the occurrence and distribution of uranium-238 (^{238}U) that was done in the 1950's included the measurment of ^{222}Rn. The petroleum industry recognized a strong correlation between ^{222}Rn emanations from overburden and petroleum resources in the underlying rocks and has measured for ^{222}Rn as an oil-exploration tool. The U.S. Department of Energy's National Uranium Resource Evaluation program, which ran from 1974 through 1984, was designed to explore the Nation for uranium deposits. The resulting data are now being reevaluated to outline probable ^{222}Rn problem areas. Some researchers in ^{222}Rn investigations are reanalyzing the data they collected decades ago.

National Research Before 1961

The role of ^{222}Rn in the lung cancer of miners in central Europe, although first suspected in 1932, was not generally accepted until the 1960's. In 1949, the U.S. Atomic Energy Commission and the Colorado State Depart-

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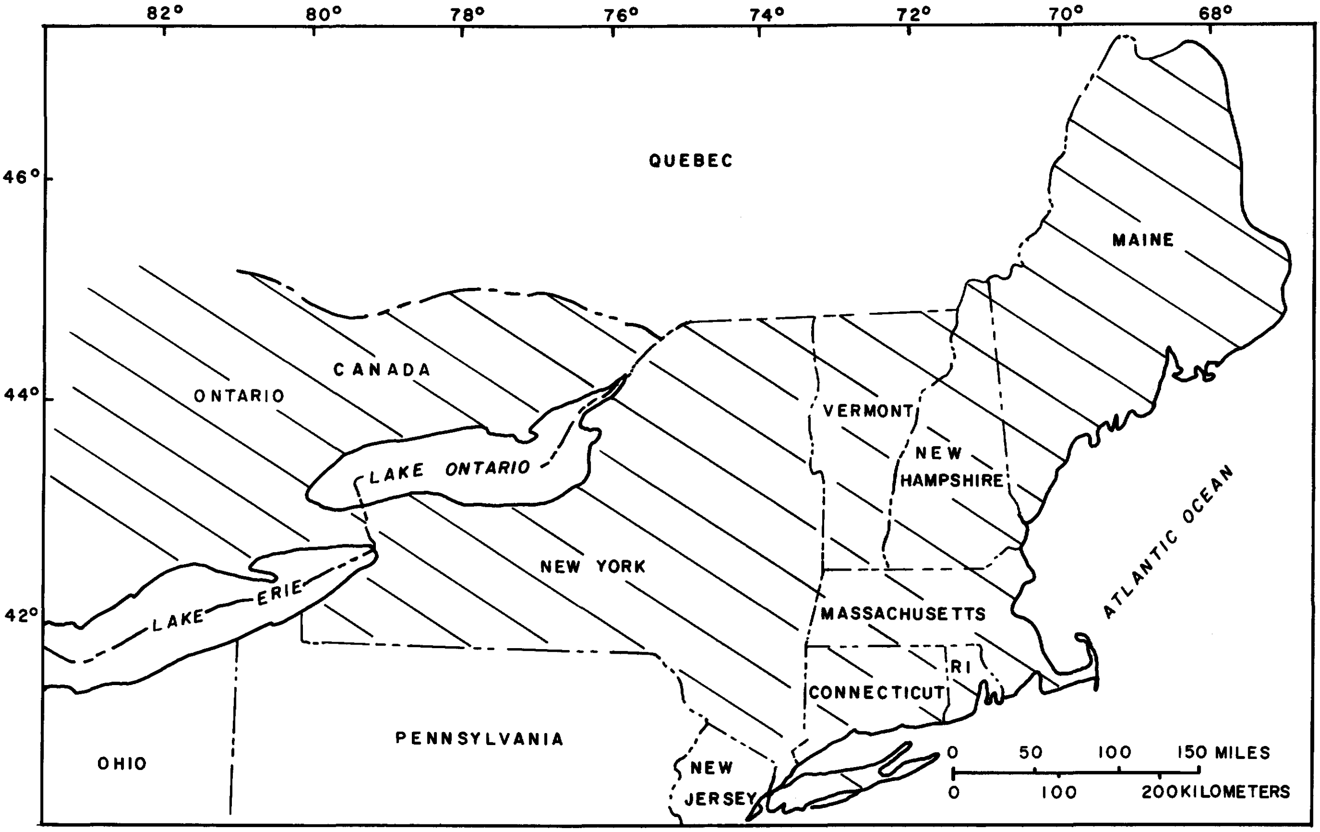


Figure 77. Location of areas represented in this review.

ment of Health, aware of the European experience, obtained data on ^{222}Rn in several mines in the United States. During 1950–52, health physicists suggested that a health hazard existed as a result of the alpha-radiation dose delivered through lung deposition of the short-lived alpha-emitting daughters of ^{222}Rn rather than from ^{222}Rn itself (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988).

From 1947 through 1957, the U.S. Atomic Energy Commission worked with the U.S. Geological Survey in a nationwide exploration for uranium (Warren Finch, U.S. Geological Survey, oral commun., 1988). Much of the information collected was published in U.S. Geological Survey trace-element reports, including those by McKeown (1951), Nelson and Narten (1951), and Johnson (1951), who determined from Geiger-counter surveys by automobile and studies of radioactivity in rocks that the total uranium content of rocks in Vermont, Massachusetts, Rhode Island, Connecticut, and New York ranged from 0.002 to 0.400 percent by weight.

The U.S. Geological Survey published several reports during the middle to late 1950's on the occurrence of uranium throughout the United States. Gott and others (1952) described total uranium occurrences in black shales, lignites, and limestones across the United States, including sites in Vermont and New York. Their findings suggest that uranium was associated with carbonaceous material or occurred as secondary infillings such as vugs. In 1955, the International Commission on Radiological Protection set an occupational exposure limit for uranium miners of 10 pCi/L (picocuries per liter) for ^{222}Rn, and the U.S. Atomic Energy Commission set a limit of 100 pCi/L for ^{222}Rn (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988). McKelvey (1955) described uranium in secondary veins in igneous and metamorphic rocks of New York and distributed throughout Precambrian dolomites in Vermont. McKeown and Klemic (1956) measured uranium and thorium contents of 0.032 and 0.150 percent by weight, respectively, in apatite in the Adirondack Mountains of New York. Chew (1956) described a field study of radioactivity in stream gravels and how the locations of ore deposits could be determined through such field surveys.

Cooper (1958) summarized research prior to 1958 on the occurrence of radionuclides in Connecticut, Delaware, Illinois, Indiana, Maine, Maryland, Massachusetts, Michigan, New Hampshire, New Jersey, New York, Ohio, Pennsylvania, Rhode Island, Vermont, and Wisconsin. The publication includes cross-referenced bibliographic information that relates mostly to radioactivity in rocks.

In 1958, The State of Maine Department of Human Services was notified of the presence of a high concentration of radioactivity in water from a private well in western Maine (Smith and others, 1961) and sent an engineer to investigate. Results confirmed high concentrations of radium-226 (^{226}Ra) and ^{222}Rn (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988). In 1959, the U.S. Public Health Service funded the Georgia Institute of Technology to check 250 wells, drilled in various terranes in Maine and New Hampshire (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988); results indicated that 99.2 percent of the drilled wells tested contained more than 2,000 pCi/L ^{222}Rn and ^{222}Rn daughters (Grune and others, 1960; Smith and others, 1961).

1961–70

When the U.S. Atomic Energy Commission stopped purchases of uranium in the late 1950's, the focus of research shifted from uranium exploration to radioactivity in surface water and ground water. Scott and Barker (1962) published data on the uranium and radium content of the ground waters of Maine, New Hampshire, Massachusetts, Rhode Island, Connecticut, and New York. Barker and Robinson (1963) described methods of determining beta activity in water, including procedures for sample collection, preservation, and analysis with a Geiger-Muller counter. Bell (1963) found uranium by weight percent in marine limestones in Massachusetts, New York, and Vermont (0.0002, 0.0001, and 0.003 to 0.0014 percent by weight, respectively). Lucas (1964) developed and refined analytical techniques for measuring ^{222}Rn content with alpha-counting cells. Barker and Johnson (1964) described the methods of determining ^{222}Rn in water, including procedures for sample collection, preservation, and analysis with a Geiger-Muller counter. Barker and others (1965) described the methods of measuring total uranium in water, including procedures for sample collection, preservation, and analysis by fluorimetry.

Tanner (1964a) described an investigation of ^{222}Rn in the ground water near the Great Salt Lake in Utah and concluded that the ^{222}Rn concentration was dependent mainly on the ^{226}Ra content of the rock or soil near the well. The author suggests that ^{226}Ra coprecipitated with ferric hydroxides near the well but also could be transported through subsurface fractures. Tanner (1964b) produced the first review of ^{222}Rn migration in the ground. He concluded that the principal mechanism by which ^{222}Rn was transported was by radioactive recoil during alpha decay, where ^{222}Rn is ejected from the ^{226}Ra parent into the fluid within the pore space in the rock or soil matrix.

In 1964, the U.S. Public Health Service investigated the wells sampled previously by Grune and others (1960) in Maine. Results confirmed the high ^{222}Rn concentrations but rejected the findings of high ^{226}Ra and concluded that ^{222}Rn, rather than ^{226}Ra , was the major problem in Maine (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988). Hoxie (1966) reviewed methods for analyzing ^{222}Rn in drinking water and developed a technique for analysis by gamma spectrometry. This method called for a 3.5-L (liter) sample, however, which proved to be bulky and difficult to transport. In 1966, the Maine Department of Human Services used an interim drinking-water standard for ^{222}Rn of 3,000 pCi/L in public supplies and 30,000 pCi/L for private water supplies in accordance with results of a literature survey that indicated ingestion to be the critical exposure pathway (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988).

The U.S. Atomic Energy Commission (1969) described the findings of a preliminary reconnaissance for uranium in Connecticut, Maine, Massachusetts, New Hampshire, New Jersey, New York, and Vermont, for the period 1950–59. This document contains hundreds of data sheets that describe field locations, rock descriptions, radioactive content, and references to many outcrops across the Northeast.

1971–79

During the 1970's, several areas of radionuclide research were delineated—uranium-resource evaluation, radioactive-waste disposal, ^{222}Rn as an earthquake predictor, and analytical techniques for measuring radionuclides in the environment.

Uranium-Resource Exploration

In 1974, the National Uranium Resource Evaluation program was started to make a comprehensive assesment of the uranium resources of the United States, in response to concern over the availability of uranium for nuclear-power generation. The evaluation included aeroradioactivity surveys, drilling and gamma-ray spectra logging, and hydrogeochemical surveys (U.S. Department of Energy, 1980).

Tanner (1978), in a supplement to his earlier ^{222}Rn-migration review (Tanner, 1964b), concluded that effective migration of ^{222}Rn was directly related to the water content of the emanating solid and that meteorologic variables affect ^{222}Rn transport in soil gases, where increased barometric pressure and precipitation decrease the exhalation of ^{222}Rn in soil gas. This review also provided detailed references on ^{222}Rn in mines, ^{222}Rn from mill tailings, ^{222}Rn emanation from lunar material, ^{222}Rn in natural gas and petroleum, and geophysical applications of ^{222}Rn measurements in the location of buried geologic structures. Gesell (1975), after reviewing 70 years of measurements, concluded that even though individual ^{222}Rn measurements as high as 1,450 pCi/L have been found, the concentrations in natural gas in the United States average less than 100 pCi/L.

Radioactive-Waste Disposal

Concern about methods of disposal of radioactive waste prompted a large research effort. Ferruccio (1975) proposed that the disposal of radioactive waste by mixing waste with tailings from salt-mine operations and storing them back in salt domes was practical. The topic of radioactive-waste disposal is too broad to address here, however, and is mentioned only for its historical context.

Radon-222 as an Earthquake Indicator

Radon-222 concentrations in ground water and soil gas near seismic zones have been studied to determine whether ^{222}Rn is a reliable predictor of earthquakes. Teng and others (1975) conducted an extensive ground-water survey in California that included weekly measurements of ^{222}Rn in well water near seismic zones and identified a relation between seismic activity and increased ^{222}Rn concentrations in ground water before earthquakes. King (1977) set Track-Etch detectors in shallow-soil auger holes along active faults in California and noted increases in ^{222}Rn concentrations in soil gas several weeks before earthquakes of magnitude 4 to 4.6.

Analytical Techniques

Techniques for measuring radionuclides in water and air interested many scientists and health physicists in the 1970's. Johnson (1971) described methods for measuring radium-228 (^{228}Ra) in water that included procedures for sample collection, preservation, and analysis with a beta-activity counter and found ^{228}Ra concentrations in natural stream water to range from 0.3 to 1.7 pCi/L. Prichard and Gesell (1977) discussed the development of analytical techniques in liquid scintillation that led to relatively easy measurement of ^{222}Rn and gross-alpha radiation in water samples. Hess and others (1973, 1978, 1979) researched ^{222}Rn in the ground water in Maine through liquid scintillation techniques.

Radionuclides in Water

Radionuclides in water and the effects upon human health were studied in the 1970's. Suomela and Kahlos (1972) did one of the few investigations of human elimination of radioactivity after ingestion of ^{222}Rn-rich water. Subjects were given a total dose of ^{222}Rn equal to 60,000 pCi/d (picocuries per day) in two or three portions, and, from this study a ^{222}Rn dose equivalent of 240 to 440 mrem (millirems) per 1,000,000 pCi was derived. From this work, Suomela and Kahlos (1972) suggest that the maximum daily intake of ^{222}Rn in drinking water should be 100,000 pCi/d. Duncan and others (1976), in a paper describing ^{222}Rn in potable water, concluded that the lungs are the critical exposure pathway for ^{222}Rn and suggested that, from the increased lung cancer observed in uranium miners, the ^{222}Rn standard for drinking water should be approximately 500 pCi/L. Partridge and others (1979) began to study the ^{222}Rn release from water and concluded that between 5 and 99 percent of the ^{222}Rn in water could be released during normal household activities. In 1978, the Environmental Protection Agency began to sample water from public supply systems across the Nation and used liquid scintilation techniques developed by Prichard and Gesell (1977) to identify the extent of ^{222}Rn in public drinking-water supplies.

In 1978 the Maine Department of Human Services, with grants made available by the Safe Drinking Water Act, funded the University of Maine to study ^{222}Rn in ground water of Maine (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988). In 1979, the Maine Department of Human Services designed and built a prototype ^{222}Rn removal unit for drinking water. The unit was based on the concept of spraying water several times to allow the release of ^{222}Rn gas, which was vented outdoors (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988).

Radon-222 in Indoor Air

During this time, the occurrence of ^{222}Rn in air began to interest many researchers, and in 1971, preliminary results of studies of lung cancer in underground miners prompted the Federal Radiological Council to reduce the occupational exposure limit for ^{222}Rn to 4 WL (working levels) per year with a maximum airborne concentration not to exceed 1 WL, where 1 WL= 200 pCi/L ^{222}Rn (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988). The Environmental Protection Agency (1975) released preliminary results on ^{222}Rn in homes built over phosphate mine tailings. Archer and others (1976) showed a relation between ^{222}Rn exposure and lung cancer.

1980 to Present

Uranium-Resource Exploration

In the 1980's the price of uranium decreased sharply, and as a result exploration for uranium declined. Duval (1983, 1987) analyzed the National Uranium Resource Evaluation data from the Middle Atlantic States and produced a color-enhanced map of uranium equivalence through a geographic information system. The map outlines some potential ^{222}Rn problem areas, such as those in the Reading Prong in Pennsylvania and the coastal-plain sediments of New Jersey, which typically are low in radioactive content. Otton and others (1985) have continued to study uranium deposits in the surficial environment, including work on the ^{222}Rn hazard associated with uranium deposits.

Radioactive-Waste Disposal

Interest in the disposal of radioactive waste has led to many avenues of research. Dinwiddie and Trask (1987) compiled several papers on all aspects of radioactive-waste disposal. Weeks (1987) investigated field tests for flow and transport in unsaturated media and concluded that gaseous diffusion from depth may be an important process relating to the transport of stored radioactive waste. Cleveland (1987) investigated transuranic elements in ground water and found that ground water from basalt in Washington and alluvial deposits in Arizona could leach plutonium from borosilicate glass. Kipp (1987) modeled radionuclide transport in ground water and concluded that a heat-and-transport model could accurately simulate the transport of radionuclides in ground water.

Radon-222 as an Earthquake Indicator

Research into the use of ^{222}Rn in ground water as a predictor of earthquake activity has continued. Thomas and Koyanagi (1986) investigated the relation between the concentration of ^{222}Rn in soil gas and seismic and volcanic activity of Kilauea Volcano, Hawaii, and found that changes in ^{222}Rn in the ground were related to the monthly eruptive cycle of Kilauea. Fleischer and Mogro-Campero (1985) compared the concentration of ^{222}Rn in soil gas in Albany, N.Y., with ^{222}Rn in soil gas in Alaska during periods of seismic activity and concluded that the concentration of ^{222}Rn sometimes increased before activity and sometimes decreased. King and Wakita (1981) investigated ^{222}Rn concentrations in ground water from an artesian well and related changes in concentration to seismic activity.

Analytical Techniques

Advances into analytical techniques for measuring radioactivity in water and air have continued during the 1980's. Yang (1984, 1987) developed a technique that bubbles helium through a water sample to collect ^{222}Rn in a Lucas or alpha-scintillation counting cell. Cothern and others (1984) investigated the measurement of calibration standards, quality assurance, and data analysis for radionuclides in environmental samples performed by the Environmental Protection Agency and contract laboratories. They concluded that 80 to 90 percent of the laboratories doing water analyses produced results that were within control limits, although they noted some difficulties in determination of ^{228}Ra. Participating laboratories also had difficulty in determining beta radiation in air samples.

Table 24. Natural radioactivity in public water supplies in the United States

Radionuclide	Number of public supplies sampled	Mean concentration (pCi/L)	
		Arithmetic	Geometric
Public water supplies, both surface water and ground water[1]			
Gross alpha radioactivity	2,535	1.8	0.6
Gross beta radioactivity	2,515	3.4	2.1
Total uranium	344	5.1	1.9
Radium-226	386	1.6	.6
Radium-228	100	3.5	2.3
Total radium	91	8.5	7.6
Radon-222	2,520	340	100
Ground-water supplies[2]			
Gross alpha radioactivity	2,510	1.9	.5
Gross beta radioactivity	2,510	3.4	2.0
Uranium-234	362	3.3	1.1
Uranium-238	362	1.7	.4
Total uranium	362	5.1	1.5
Radium-226	403	1.7	.6
Radium-228	107	3.6	2.1
Total radium	97	8.5	7.6
Radon-222	2,457	352.8	86.6

[1] Data from Horton, 1983.
[2] Data from Horton, 1985.

Radionuclides in Water

Research during the 1980's expanded to address the environmental sources of radionuclides, especially ^{222}Rn, and the health effects of both ingestion and inhalation of radionuclides. In 1980, the Maine Department of Human Services, in conjuction with other New England States, reviewed current literature and calculated the exposures that result from ingesting ^{222}Rn with drinking water and suggested a drinking-water standard of about 50,000 pCi/L (D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988). Horton (1983), through an Environmental Protection Agency study begun in 1980, measured gross-alpha and -beta radiation, total uranium, ^{226,228}Ra, and ^{222}Rn in public water systems across the Nation that serve 1,000 people or more; results are summarized in table 24. This work prompted researchers to make a closer examination of radionuclides in private homes and private water supplies. Horton (1985) presented an update of the data collected from the 1980 study that focused on ground-water supplies serving 1,000 people or more; these results also are given in table 24.

A summary of Horton's (1985) data on radionuclides in ground water in New England and New York is presented in table 25. Horton (1983, 1985) showed that ground water from these States generally has lower-than-average concentrations of gross-alpha and gross-beta radiation, total uranium, and ^{226}Ra. Although Horton (1985) gives no data for ^{228}Ra for public ground-water supplies in the Northeast, the national arithmetic means for total radium and ^{228}Ra are 8.5 and 3.6 pCi/L, respectively. Concentrations of ^{222}Rn in

Table 25. Radionuclide concentrations in public ground-water supplies in New England and New York

[Values are in picocuries per liter; —, no data; data from Horton, 1985]

State	Gross alpha			Gross beta			Radium-226			Radon-222		
	Arithmetic mean	Geometric mean	No. of sites	Arithmetic mean	Geometric mean	No. of sites	Arithmetic mean	Geometric mean	No. of sites	Arithmetic mean	Geometric mean	No. of sites
Maine	1.3	0.4	25	2.2	1.7	25	0.6	0.3	2	—	—	0
New Hampshire	.4	.2	26	2.0	1.6	26	.4	.4	1	1,388	944	26
Vermont	.7	.3	11	1.3	1.2	11	1.0	1.0	1	618	556	11
Massachusetts	.3	.1	100	1.9	1.3	100	.2	.2	1	790	507	100
Rhode Island	.4	.2	30*	1.6	1.2	30*	.1	.1	1	1,376	1,140	30*
Connecticut	—	—	0	—	—	0	—	—	0	—	—	0
New York	.3	.2	132	1.7	1.3	132	.1	.1	1	171	43	130
United States total	1.9	.5	2,510	3.4	2.0	2,510	1.7	.6	403	353	87	2,457

* Represents 30 individual wells but only 11 public water supplies.

Table 26. Total uranium and radon-222 concentrations in water supplies in New England and New York

[Values are in picocuries per liter; —, no data; data from Hess and others, 1985b]

State	Total uranium			Radon-222							
				Private well		Public supply[1]		Public ground water		Public surface water	
	Surface water	Ground water	Domestic water[1]	Geometric mean	No. of sites	Geometric mean	No. of sites	Geometric mean	No. of sites	Geometric mean	No. of sites
Maine	0.010	0.035	0.035	7,000	24	990	71	—	0	—	0
New Hampshire	.010	.035	.035	1,400	18	9	12	940	52	—	6
Vermont	.010	.010	.010	210	23	840	4	660	71	13	16
Massachusetts	.010	.010	.010	1,000	8	7	2	500	212	38	2
Rhode Island	.075	.075	.075	6,500	69	5,200	6	2,400	575	—	10
Connecticut	.010	.075	.035	—	0	—	0	—	0	—	0
New York	.010	.035	.035	1,500	4	34	20	52	292	—	1
United States total	—	—	—	920	434	68	224	130	6,298	1	131

[1] May be a blend of surface and ground waters.

ground water are well above the national average except in New York. Hess and others (1985b) compiled nationwide data for total uranium and ^{222}Rn in public surface-water and ground-water supplies and private supplies; table 26 presents their data for New England and New York. Hess and others (1985b) show that, in general, private well water in these States has higher concentrations of total uranium and ^{222}Rn than public ground-water or surface-water supplies.

Radon-222 in Indoor Air

The 1980's were a period of intensive research into the occurrence of ^{222}Rn in outdoor and indoor air. Gesell (1983) concluded that the average outdoor ^{222}Rn concentration across the United States ranged from 0.1 to 0.4 pCi/L and noted that the maximum soil gas concentration occurs at sunrise and the minimum in the afternoon. George and others (1983) compared indoor ^{222}Rn concentrations in solar-energy-efficient homes with those in conventional homes. The indoor ^{222}Rn concentrations in the solar homes in New Hampshire, Connecticut, and New York ranged from 5.0 to 0.3 pCi/L, and the associated ^{226}Ra in the soils ranged from 2.5 to 0.15 pCi/g (picocuries per gram). Their main conclusion, however, was that indoor ^{222}Rn concentrations in solar-energy-efficient homes did not differ from those in conventional homes. Nero and others (1986) concluded from a study of the distribution of ^{222}Rn (ranging from grab samples to 1-year samples) in 1,377 homes across the United States that the arithmetic mean was 1.5 pCi/L, and the mean values for central Maine and New York were 2.97 and 1.57 pCi/L, respectively, both of which are above the national average.

MAINE

Previous Work

The first studies of natural radioactivity in Maine began during the late 1950's and early 1960's as part of a regional uranium-exploration program; results indicated several areas of higher-than-expected background radiation throughout the State. Ground measurements of gross radioactivity by Geiger-counter surveys across Maine and New Hampshire by Nelson and Narten (1951) indicated that the granites in Maine contain 0.002 to 0.003 percent uranium by weight. Grune and others (1960) and Smith and others (1961) began the first study of radioactivity in the ground water and surface water of Maine and found high levels of ^{222}Rn gas in a tank of well water near Raymond (fig. 78). This study involved 500 measurements near Raymond (fig. 78). Lowder and others (1963) made an environmental radiation study in northern New England, including Maine (reported by Brutsaert and others, 1981), and Hoxie (1966) followed up with more specific measurements of ^{222}Rn in Maine's ground water. Hess and others (1973) made a radioisotope investigation near Wiscasset (fig. 78), and Duncan and others (1976) stated that some of the highest ^{222}Rn concentrations in water in the United States were in Maine. Hess and others (1979, p. 4) note that one purpose of their research was to determine whether the high cancer rates in Maine could be correlated with high concentrations of ^{222}Rn. Their research included measurements of ^{222}Rn in water and air, correlation of ^{222}Rn with differing geologic terranes, collection of data on cancers, and statistical testing of the data to determine whether the correlation between ^{222}Rn and cancer was significant. Otton and others (1980) investigated anomalous total uranium and thorium concentrations in the rocks of the Tomah Mountain area in eastern Maine (fig. 78).

Brutsaert and others (1981) measured the concentration of ^{222}Rn in ground water from granites, calcareous rocks, pelitic rocks, and quartz-rich rocks in Maine and attempted to correlate ^{222}Rn concentrations in ground water with more easily measurable characteristics and chemical constituents. Samples from wells completed in granite had the highest ^{222}Rn concentrations, and the concentrations varied inversely with well yield. The concentrations indicated an inverse correlation with sodium, but road salting may have been a factor. Hess and others (1981) investigated ^{222}Rn, ^{226}Ra, and total uranium in the air and the waters of Maine expanding on the work by Hess and others (1979). Weiffenbach (1982) followed up with a report on ^{222}Rn in water and air with emphasis on health risk and control.

Lowry (1983) and Lowry and Brandow (1985) began research into the removal of ^{222}Rn from water by granular-activated carbon filters. Lowry and others (1987) have been applying the technological developments to Maine homes whose private water supplies have extremely high ^{222}Rn concentrations (1.1 million pCi/L). Granular-activated carbon was effective in removing 99 percent of the ^{222}Rn, and uranium was removed with an ion-exchange resin. The removal of ^{222}Rn and uranium from the water led to an almost total removal of ^{222}Rn in indoor air at the home that had a ground-water concentration of 1.1 million pCi/L of ^{222}Rn. Recent investigations (Ground Water Monitor Review, 1988; D.C. Hoxie, Maine Division of Health Engineering, written commun., 1988) suggest that a buildup of radioactive lead-210 (^{210}Pb) in granular-activated carbon filters may be of environmental concern as well as the disposal of the filters, which could be considered low-level radioactive waste.

Hess and others (1985a) investigated the concentration of ^{222}Rn in indoor air with alpha Track-Etch detectors as a function of the energy efficiency or "tightness" of homes in Maine. Hess and others (1986) made a detailed study of the contribution of ^{222}Rn in water to ^{222}Rn in indoor air and concluded that the ^{222}Rn level due to water use could range from 0 to 36 percent of the total ^{222}Rn in indoor air. Lanctot (1985) made a preliminary analysis of data from a cooperative epidemiological study of ^{222}Rn and its relation to cancer in Maine. Lanctot and others (1985) combined the work of Hess and others (1979) with that of Lanctot (1985) and determined that ^{222}Rn concentrations in ground water varied as a function of aquifer hydraulics and uranium geochemistry within the aquifer.

The Cooperative Extension Service of the University of Maine at Orono (U.S. Department of Agriculture, 1986) published a summary of health risks and control measures for radon-222 in water and air that describes the occurrence, distribution, and remediation of ^{222}Rn in Maine.

Distribution and Concentration of Radionuclides in Ground Water

Most of the data from the publications just referred to relate to ^{222}Rn in ground water. Data on ^{222}Rn and other radionuclides in samples collected in Maine during national studies by Hess and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of 500 to more than 10,000 pCi/L for ^{222}Rn and 1 to 5 pCi/L for ^{226}Ra and total uranium in the drinking water from selected counties of Maine. Table 27 includes concentrations of gross-beta radioactivity, total radium, and uranium in water from selected wells in Maine. It is evident from work done in Maine (Hess and others, 1979, 1981) and reported by Lanctot and others (1985) that ground water from most, if not all, of the rock formations in Maine have been sampled at least once. Radon-222 concentrations in water from several types of aquifers are given in table 28.

Ground-Water Availability and Rate of Withdrawal

Data on ground-water withdrawals (Maloney and Cowing, 1985) indicate the percentage of the population that is exposed to potential health risk from radionuclides in the water supply of Maine. The crystalline-bedrock aquifers of Maine are the most widely used for water supply and also have the highest concentrations of ^{222}Rn. Well yields from stratified drift aquifers generally range from less than 1 to

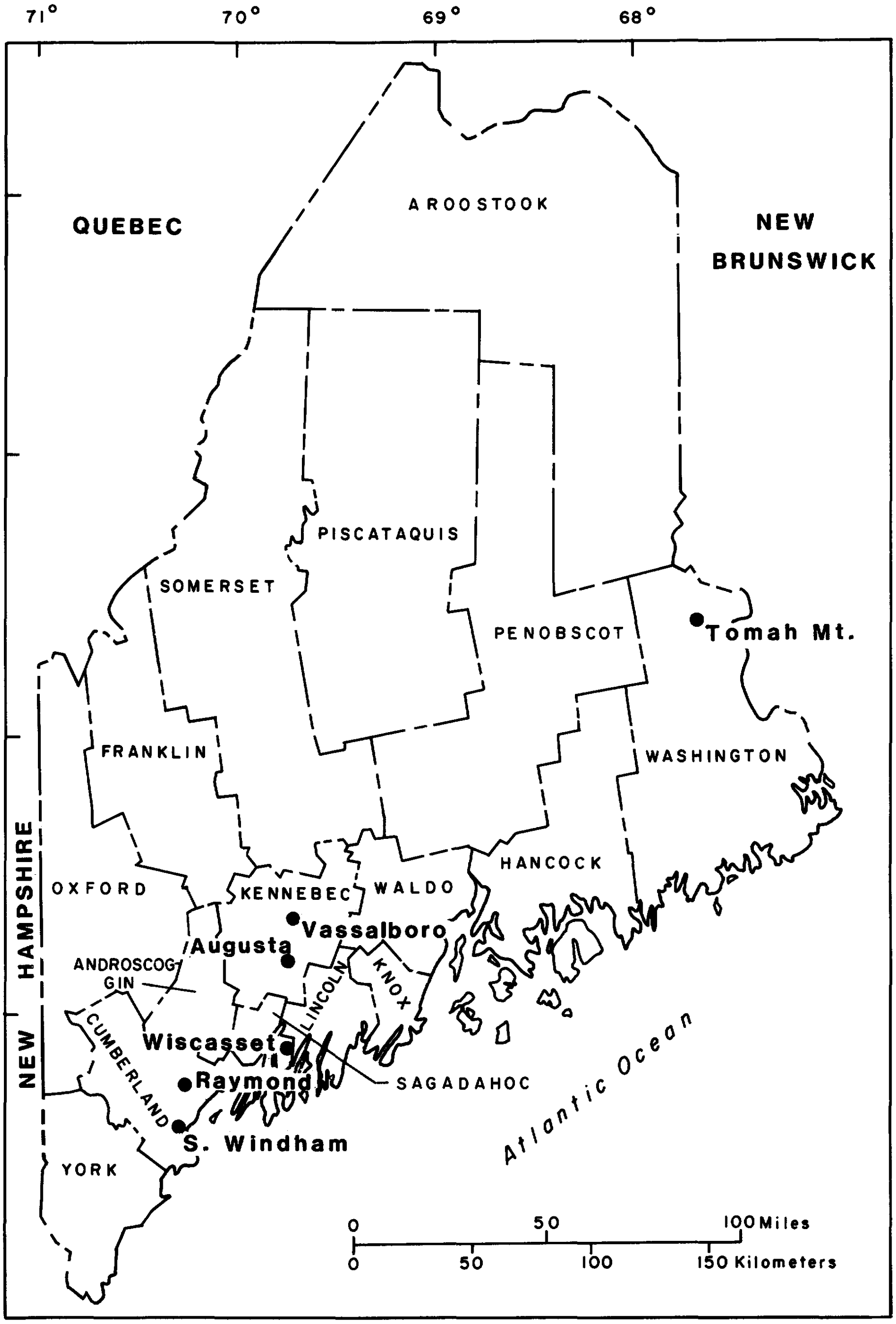


Figure 78. Locations of Maine sites mentioned in text. (Scale 1:3,168,000; base from Adamik, 1984.)

1,000 gal/min (gallons per minute); yields from bedrock aquifers range from 2 to 30 gal/min. The 642,000 residents of Maine that rely on ground water as a drinking supply, 45 percent of which are on small or private systems, withdraw 46 Mgal/d (million gallons per day).

Radon in Indoor Air

Hess and others (1979, p. 80) made a series of 24-hour tests of ^{222}Rn in indoor air in 10 living rooms and 1 school with a Wrenn-Spitz radon detector and obtained an average value of 2.67 pCi/L. Hess and others (1985b) report an average of 3.0 pCi/L for indoor air in Maine. Work by Hess and others (1985a) indicates that measurements need to be made for 1 year to account for seasonal variations in ^{222}Rn concentration in indoor air. Hess and others (1986) note that degassing of ^{222}Rn from tap water resulted in spikes in indoor air concentrations during peak water-use periods.

Table 27. Geohydrologic data and concentrations of radium and uranium in water samples from wells in Maine
[Data from Scott and Barker (1962); locations shown in fig. 78; pCi/L, picocuries per liter; gal/min, gallons per minute]

Location		Geohydrologic data			Concentration (pCi/L)		
Town	County	Water-bearing unit	Well yield (gal/min)	Date of collection	Gross-beta activity	Radium	Uranium[1]
Vassalboro	Kennebec	Schist	8	03–27–57	<5	0.01	0.07
Augusta............	Kennebec	Sand and gravel	1,000	03–27–57	<5	.1	.54
South Windham	Cumberland	Granite	10	03–27–57	7	3.3	7.48

[1] Converted from micrograms per liter (µg/L) by multiplying by 0.68 and assuming that all activity is due to uranium and that uranium is in secular equilibrium.

Table 28. Radon-222 concentrations in Maine ground water, by aquifer type
[Dashes indicate no data; concentrations are in picocuries per liter]

Aquifer material	Number of samples	Radon-222 concentration	
		Arithmetic mean	Maximum
Bedrock			
Igneous[1]			
granite	114	15,008	151,182
volcanics	21	7,395	46,103
Metamorphic[1]			
low-grade	95	2,111	18,115
medium-grade	76	4,093	46,103
high-grade..............	102	7,059	65,656
Sedimentary[1]			
calcareous mixed sandstone..............	99	3,803	36,782
pelites..................	10	6,372	40,069
sulfidic carbonaceous pelites.................	24	4,431	18,115
limestone................	13	8,504	37,424
Interbedded			
sandstone and shale	65	4,998	65,656
limestone and shale	30	1,908	14,243
All granites[2]	136	22,100	—
Metamorphics[2]			
low-grade	56	1,100	—
high-grade..............	35	13,630	—
Granites[3]	84	93,300	—
Metamorphics[3]...........	44	58,200	—
Unconsolidated sediments[4]			
dug wells...............	46	1,432	—
Springs....................	22	3,705	—

[1] Lanctot (1985).

[2] Hess and others (1979), as reported by Hall and others (1985, p. 90).

[3] Smith and others (1961), as reported by Hall and others (1985, p. 90).

[4] Bogdan and others (1987).

A recent study of indoor air in 1,500 homes in Maine by alpha Track-Etch detectors (D.C. Hoxie, Maine Division of Health Engeering, written commun., 1988) obtained an arithmetic mean of 4.87 pCi/L and a geometric mean of 1.80 pCi/L for ^{222}Rn. The highest value recorded was 1,212.46 pCi/L.

NEW HAMPSHIRE

Previous Work

The first studies of natural radioactivity in New Hampshire were begun in the early 1950's as ground measurements of gross radioactivity by McKeown (1951), who found that the uranium content of rocks in the State ranged from 0.002 to 0.003 percent by weight. Smith and others (1961) reported on natural radioactivity in water from selected wells in New Hampshire. Lyons (1964) studied the distribution of uranium and thorium in the Lower Devonian plutonic rocks in New Hampshire and reported content ranging from 0.6 to 39 ppm (parts per million) uranium and 2.8 to 57 ppm thorium. He also reported that, in these rocks, most of the uranium was concentrated on grain boundaries and in interstitial spaces, and not necessarily within accessory minerals. Butler (1975) made a geochemical study of uranium and thorium in rocks of the White Mountain plutonic series (now designated White Mountain Plutonic-Volcanic Suite) through chemical and spectrographic techniques. Boudette (1977) investigated the uranium potential of a two-mica granite. Hoisington (1977) studied the uranium and thorium distribution in the Conway Granite. Bothner (1978) described selected uranium and thorium occurrences in New Hampshire. Page (1980) published a guide to prospecting for uranium and thorium in New Hampshire.

Donahue (1984) investigated the hydrogeologic factors that influence ^{222}Rn in the ground water of southeastern New Hampshire. Hall and others (1985) made a general investigation of ^{222}Rn in the ground waters of New Hampshire from 1982 to 1985; results show that ground water from a two-mica granite has the highest mean concentration of ^{222}Rn in the State (45,000 pCi/L). Observations by Brutsaert and others (1981) that ^{222}Rn concentrations are

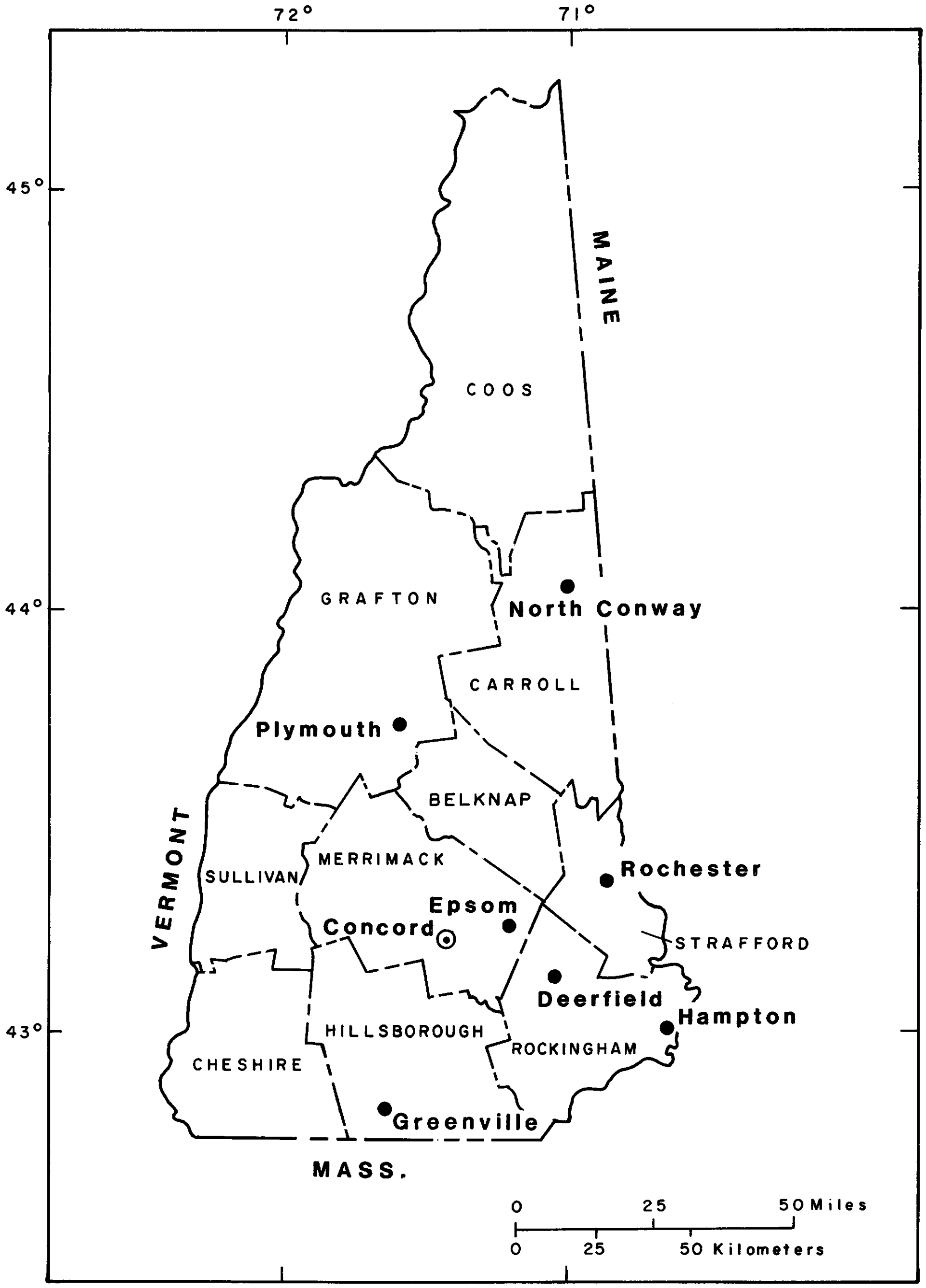


Figure 79. Locations of New Hampshire sites mentioned in text. (Scale 1:2,112,000; base from Cotton and Hammond, 1985a.)

inversely related to sodium, chloride, and iron content were generally upheld by Hall and others (1985), who present data indicating that the inverse proportionality between ^{222}Rn concentration and well yield is not absolute (Hall and others,1985, p. 96).

Campisano and Hall (1986), Campisano (1987), Wathen (1986), and Wathen and Hall (1986) present additional results of studies of ^{222}Rn in ground water near Deerfield, in southeastern New Hampshire (fig. 79), and the two-mica granite. Hall and others (1987) summarize the geologic controls on ^{222}Rn occurrence in New England. A map compiled for the Environmental Protection Agency by Olszewski and Boudette (1986), based on a generalized bedrock geologic map of New England, emphasizes uranium distribution in major rock units as an indicator of ^{222}Rn occurrence. Cameron and others (1986) studied geochemical controls on uranium in selected peat deposits in New Hampshire and Vermont.

Table 29. Geohydrologic data and concentrations of radium and uranium in water samples from wells and springs in New Hampshire

[Data from Scott and Barker (1962); locations shown in fig. 79; <, less then; pCi/L, picocuries per liter; gal/min, gallons per minute]

Location		Geohydrologic data			Concentration (pCi/L)		
Town	County	Water-bearing unit	Well yield (gal/min)	Date of collection	Gross-beta activity	Radium	Uranium[1]
North Conway......	Carroll	Sand and gravel	300	04–02–57	<5	0.1	0.1
Plymouth...........	Grafton	Alluvium	350	10–10–55	<5	.1	.07
Epsom.............	Merrimack	Littleton Formation	15	09–28–54	<5	.1	.2
Rochester..........	Strafford	Littleton Formation	7.5	10–20–55	<5	.5	.3
Greenville..........	Hillsborough	Littleton Formation	2–15	04–03–57	<10	.1	.2
Hampton...........	Rockingham	Sand and gravel	460	09–28–54	<5	.1	.4

[1] Converted from micrograms per liter (μg/L) by multiplying by 0.68 and assuming that all activity is due to uranium and that uranium is in secular equilibrium.

Wathen (1987) studied the siting of uranium and ^{222}Rn activity in ground water from a two-mica granite through autoradiographic techniques and whole-rock analyses for uranium and demonstrated that uranium and ^{226}Ra are found as grain coatings, as suggested by Lyons (1964). Wathen (1987) also found that, although uranium in solution was measured at concentrations as high as 100 μg/L (micrograms per liter), it was not proportional to ^{222}Rn concentration. Wathen (1987, p. 41) suggests that high ^{222}Rn concentrations in water can be correlated with uranium localized on grain borders in the rock near the well.

Distribution and Concentration of Radionuclides in Ground Water

Data on ^{222}Rn and other radionuclides in samples collected in New Hampshire during national studies by Hess and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of 0 to 3 pCi/L for gross-alpha radiation, 1,000 to more than 10,000 pCi/L for ^{222}Rn, and 1 to 5 pCi/L for ^{226}Ra and total uranium in the drinking water from selected counties of New Hampshire. Table 29 includes concentrations of gross-beta radioactivity, total radium, and uranium in water from selected wells in New Hampshire. Hall and others (1985) report that ground water from most, if not all, of the rock formations in New Hampshire have been sampled at least once. Radon-222 concentrations in water from several types of aquifers are given in table 30.

Table 30. Radon-222 concentrations in New Hampshire ground water, by formation

[—, no data; concentrations are in picocuries per liter]

Formation	Number of samples	Radon-222 concentration	
		Arithmetic mean	Range
Kittery[1].............	10	1,900	652–3,880
Eliot[1]................	12	2,520	784–5,470
Berwick[1]............	20	11,900	1,360–29,500
Littleton[1]............	11	10,600	913–44,500
Exeter Diorite[1].......	10	1,800	744–2,800
Quartz diorite[1].......	3	3,570	1,140–8,300
Quartz monzonite[1]....	14	34,000	4,110–105,000
Two-mica granite[1]....	11	45,100	613–258,000
Conway Granite[1].....	13	12,200	342–70,300
Conway Granite[2].....	2	32,540	—
Fitchburg Granite[3]....	8	284,000	—

[1] Hall and others (1985).

[2] Hess and others (1979).

[3] Smith and others (1961).

Ground-Water Availability and Rate of Withdrawal

Data on ground water withdrawals (Cotton and Hammond, 1985a) indicate the percentage of the population that is exposed to potential health risk from radionuclides in the water supply of New Hampshire. The major aquifers in New Hampshire are stratified-drift deposits, which have well yields ranging from 100 to 500 gal/min. The crystalline-bedrock aquifers of New Hampshire are widely used for water supply and also have the highest concentrations of ^{222}Rn. Well yields from bedrock aquifers generally range from 2 to 30 gal/min. The 550,000 residents of New Hampshire who rely on ground water as a drinking supply, 17 percent of which are on small or private systems, withdraw 52 Mgal/d.

Radon in Indoor Air

The focus of radionuclide studies in New Hampshire has been on ^{222}Rn in ground water; research on ^{222}Rn in air has been less extensive. Results from 635 nonrandom, short-term screening tests that used charcoal-canister-type

Table 31. Concentrations of radon-222 and gross-alpha radiation in drinking-water systems in Vermont
[Data from Manning and Ladue (1986); concentrations are in picocuries per liter]

Source (formation)	Number of homes	Radon-222 Range	Radon-222 Arithmetic mean	Gross alpha
Precambrian core of Green Mountains				
Granites, gneiss, andesites, quartzite	18	107–3,662	825	4.0
Metamorphic				
Ottaquechee—greenstone and serpentine	13	26–2,282	931	1.1
Metasedimentary rocks				
Waits River, Gile Mountain, Missisquoi	86	0–5,597	1,196	1.7
Cambrian sedimentary rocks				
Pinnacle, Underhill, Hazen Notch	54	48–6,159	1,127	1.1
Cambrian clastic rocks				
Dalton, Dunham, Monkton, Winooski, Saxe Brook	27	216–2,265	814	4.2
Ordovician black shales				
Stony Point, Iberville, Hathaway	2	111–2,332	1,221	3.5
Carbonate rocks				
Beekmantown Group, Chazy Group	19	115–1,893	635	1.7
Unconsolidated sediment	45	31–2,038	611	.8
Unspecified springs	25	50–1,824	733	.7

detectors indicated an average concentration of 6.7 pCi/L of ^{222}Rn in indoor air of basements (Chris Pirie, New Hampshire Division of Public Health Service, oral commun., 1988). This is not a value from a statistical analysis of the data; rather, it is used to represent an overall "worst-case" value for the State. The New Hampshire Division of Public Health Services began a study of 2,200 randomly selected homes during the winter of 1988 and intends to use the results to establish an average level for ^{222}Rn in indoor air.

VERMONT

Previous Work

The first study of natural radioactivity in Vermont was a series of ground measurements of gross radioactivity by McKeown (1951). Additional research did not begin until the late 1970's, when McHone and Wagner (1980) began general uranium exploration research. Dorsey and Vanecek (1983) and Vanecek and Dorsey (1983a,b), in a series of geologic and radiometric surveys in Vermont, focused primarily on general measurements of radioactivity rather than ^{222}Rn in air or water.

Manning and Ladue (1986) of the Vermont Department of Health measured ^{222}Rn concentrations in 689 samples of drinking water from 366 sites. All samples were analyzed for ^{222}Rn through liquid-scintillation techniques, and data for gross-alpha activity also were collected.

The University of Vermont's Department of Natural Resources has been developing a geographic information system for analyzing radionuclide data. The system entails the digitizing of geographic, geologic, and radionuclide data, which can then be manipulated by computer techniques and graphically displayed.

Distribution and Concentration of Radionuclides in Ground Water

Data on ^{222}Rn and other radionuclides in samples collected in Vermont during national studies by Hess and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of 100 to 1,000 pCi/L for ^{222}Rn and 1 to 5 pCi/L for ^{226}Ra and total uranium in the drinking water from selected counties of Vermont. Manning and Ladue (1986) sampled ground water from most, if not all, of the water-bearing formations in Vermont; results are summarized in table 31. The higest concentration of ^{222}Rn in drinking water was 14,400 pCi/L in a well completed in the Precambrian core rocks of the Green Mountains near Ludlow (fig. 80).

Ground-Water Availability and Rate of Withdrawal

Data on ground-water withdrawals (Cotton and Hammond, 1985b) indicate the percentage of the population that is exposed to potential health risk from radionuclides in the water supply of Vermont. The major aquifers in Vermont are stratified-drift deposits, which generally have well yields of 30 to 400 gal/min. The crystalline-bedrock aquifers of Vermont are widely used for water supply and may have the highest concentrations of ^{222}Rn. The 275,000

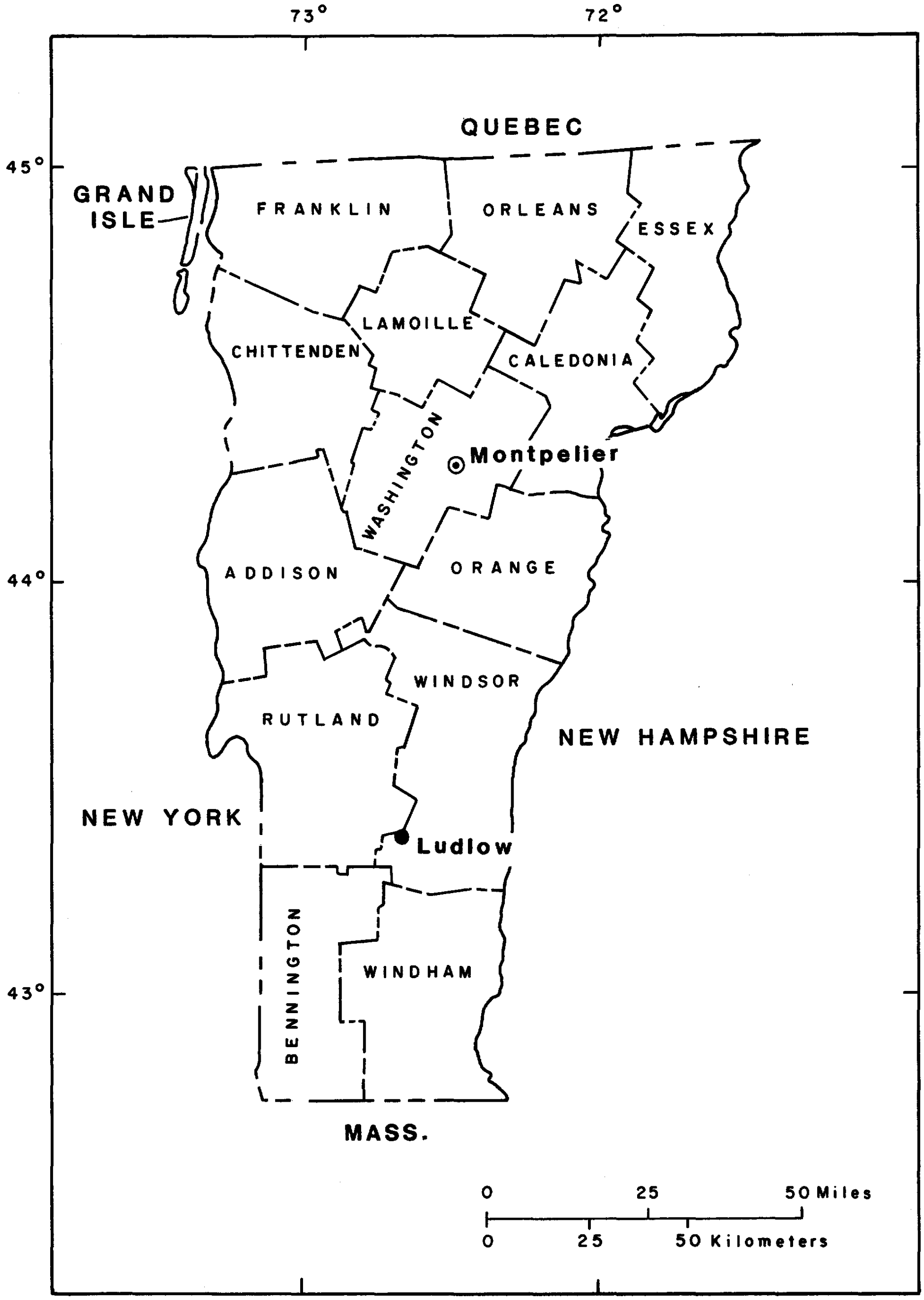


Figure 80. Locations of Vermont sites mentioned in text. (Scale 1:1,900,800; base from Cotton and Hammond, 1985b.)

residents of Vermont that rely on ground water as a drinking supply, 32 percent of which use small or private systems, withdraw 34 Mgal/d.

Radon in Indoor Air

The focus of radionuclide research in Vermont has been on ^{222}Rn in ground water; research on ^{222}Rn in air has been less extensive. The home that had the highest ^{222}Rn concentration in drinking water (14,400 pCi/L) had indoor-air concentrations of 0.6 to 2.5 pCi/L (Manning and Ladue, 1986). Further information on ^{222}Rn in indoor air in Vermont can be obtained through direct inquiry from the Vermont Department of Health, Division of Occupational and Radiological Health.

MASSACHUSETTS

Previous Work

The first studies of natural radioactivity in Massachusetts began in the early 1950's with ground measurements of gross radioactivity by Johnson (1951), who described a region of central Massachusetts as part of the "Central New England Radioactive Province." Popenoe (1966) compared aeroradioactivity surveys with the geology of southwestern Massachusetts. Fisenne and others (1984) reported on abundances of $^{234,238}U$, ^{226}Ra, and ^{210}Pb in sandy soils of North Eastham (fig. 81). These works focused primarily on general measurements of radioactivity rather than ^{222}Rn in water or air.

Davis and others (1987) measured the bulk ^{222}Rn emanation rates of several granites in Massachusetts through high resolution, gamma-ray spectroscopy. They collected and analyzed samples of Andover, Cape Ann, Chelmsford, Milford, Peabody Granite, Sharpners Pond Diorite, the Concord Granite from New Hampshire, and the Rockville Granite from Minnesota. Samples were either cut into slabs of variable thickness or ground into distinct particle-size fractions ranging from less than 0.5 to 20 cm (centimeters) in diameter. The gamma-ray spectrum of each sample was measured and used to calculate both uranium concentration and percent loss of ^{222}Rn for each sample. Results indicate that ^{222}Rn emanation increases with decreasing particle size. Davis and Hon (1988) have expanded that work by applying empirical data on ^{222}Rn emanation to predictive models for unsupported ^{222}Rn in the environment.

Distribution and Concentration of Radionuclides in Ground Water

Data on ^{222}Rn and other radionuclides in samples collected in Massachusetts during national studies by Hess and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of 100 to 10,000 pCi/L for ^{222}Rn and 1 to 5 pCi/L for ^{226}Ra and total uranium in the drinking water from selected counties of Massachusetts. Table 32 includes concentrations of gross-beta radiation, total radium, and uranium in water from selected wells in Massachusetts.

Although recent data on ^{222}Rn and other radionuclides in ground water are scant, ^{222}Rn concentrations in water sampled from five wells in the Ayer Granite range from 2,000 to 5,400 pCi/L, and three other samples from wells in the Ayer Granite, tested by homeowners, had ^{222}Rn concentrations of 93,000, 67,000, and 61,000 pCi/L (James Persky, U.S. Geological Survey, written commun., 1988).

Ground-Water Availability and Rate of Withdrawal

Data on ground water withdrawals (Frimpter, 1985) indicate the percentage of the population that is exposed to potential health risk from radionuclides in the water supply. The major aquifers in Massachusetts are stratified-drift deposits that have well yields generally ranging from 100 to 1,000 gal/min. The crystalline-bedrock aquifers of Massachusetts are widely used for water supply and may have the highest concentrations of ^{222}Rn. Well yields from bedrock aquifers range from 1 to 100 gal/min. The 1,900,000 residents of Massachusetts who rely on ground water as a drinking supply, 7 percent of which rely on small or private systems, withdraw 190 Mgal/d.

Radon in Indoor Air

Information on ^{222}Rn in indoor air in Massachusetts is limited and can be obtained through direct inquiry to the Massachusetts Department of Public Health. Nero (1983) investigated ^{222}Rn in seven single family residences in Boston (fig. 81), and found the average for grab samples in the living area to be 0.07 pCi/L. Although many indoor-air measurements have been made in Massachusetts (William Bell, Massachusetts Department of Public Health, oral commun., 1988), the sites generally have been selected on the basis of socioeconomic factors and do not represent the State in general.

RHODE ISLAND

Previous Work

The first studies of natural radioactivity in Rhode Island began in the early 1950's with ground measurements of gross radioactivity by McKeown (1951). Johnson (1951) described a region of soutwestern Rhode Island as part of the "Central New England Radioactive Province." Popenoe (1966) compared aeroradioactivity surveys with the geology of Rhode Island. Goldsmith and others (1977) correlated aeroradioactivity and geology of the southwestern portion of Rhode Island and found trends similar to those of Popenoe (1966). The references cited here focus primarily on general measurements of radioactivity rather than ^{222}Rn in water or air.

Ryan and Kipp (1985) and Kipp and others (1986) investigated contamination of ground water by radioactive waste near Wood River Junction (fig. 82). The two major radionuclides that they detected were strontium-90 and

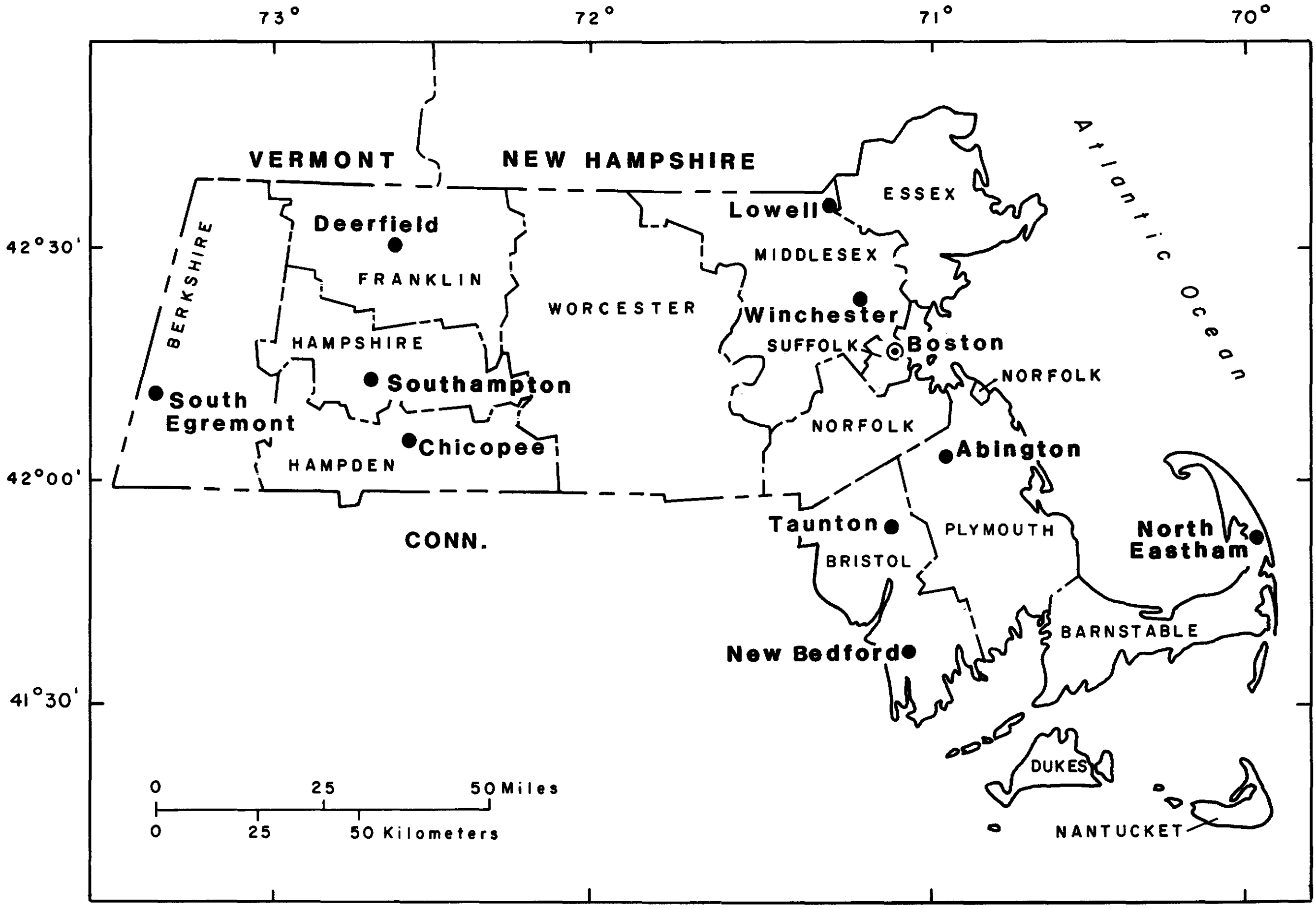


Figure 81. Locations of Massachusetts sites mentioned in text. (Scale 1:1,810,286; base from Delaney and Maevsky, 1980.)

Table 32. Geohydrologic data and concentrations of radium and uranium in water samples from wells and springs in Massachusetts

[Data from Scott and Barker (1962); locations shown in fig. 81; <, less than; pCi/L, picocuries per liter; gal/min, gallons per minute]

Location		Geohydrologic data			Concentration (pCi/L)		
Town	County	Water-bearing unit	Well yield (gal/min)	Date of collection	Gross-beta activity	Radium	Uranium[1]
Deerfield	Franklin	Sugarloaf Formation	2	12–09–54	3	<0.1	0.5
Lowell	Middlesex	Ayer Granite	85	03–27–57	<5	<.1	2.3
Winchester	Middlesex	Newburyport Complex	5	06–16–54	<10	<.1	.3
South Egremont	Berkshire	Stockbridge Formation	22	12–08–54	<20	.1	1.9
Southampton	Hampshire	Sugarloaf Formation	1.5	06–16–54	<5	.1	.2
Chicopee	Hampden	Chicopee Shale[2]	70	12–08–54	<25	.7	1.0
Abington	Plymouth	Wamsutta Formation	75	03–27–57	<5	.4	<.1
Taunton	Bristol	Rhode Island Formation	55	10–28–55	<10	.5	<.1
New Bedford	Bristol	Dedham Granite	100	10–31–55	<10	.3	1.1

[1] Converted from micrograms per liter (µg/L) by multiplying by 0.68 and assuming that all activity is due to uranium and that uranium is in secular equilibrium.

[2] A former usage.

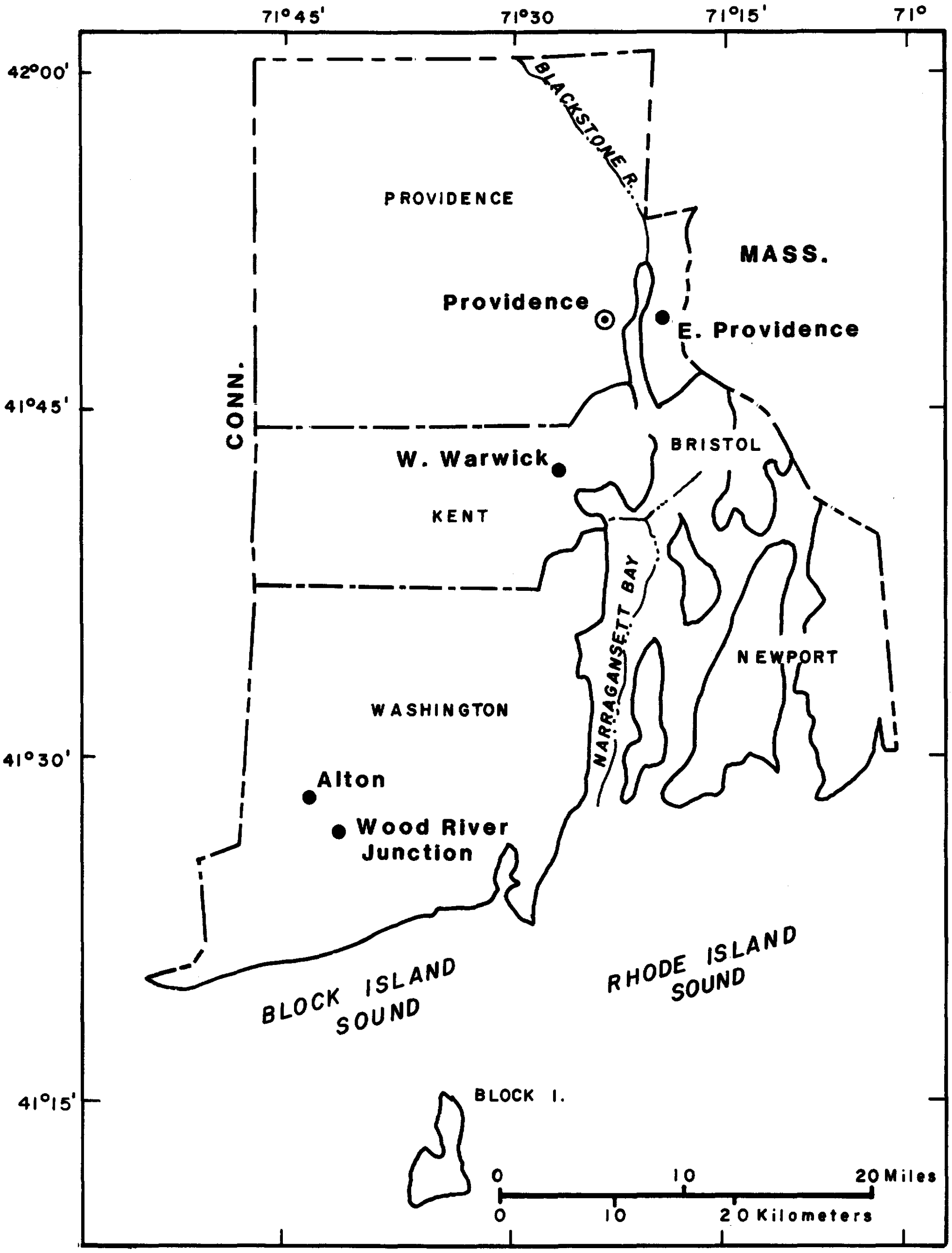


Figure 82. Locations of Rhode Island sites mentioned in text. (Scale 1:633,600; base from Lang, 1961.)

technetium-99, whose combined gross-beta activity ranged from 5 to 500 pCi/L. They also found that both radionuclides were readily transported in the ground water in a sand and gravel aquifer.

The Rhode Island Department of Environmental Management sampled 450 private drinking-water wells for ^{222}Rn in 1986; preliminary results of 260 of those samples suggest an average concentration of ^{222}Rn of 5,220 pCi/L (Margaret Bradley, Rhode Island Department of Environmental Management, written commun., 1988).

Distribution and Concentration of Radionuclides in Ground Water

Data on ^{222}Rn and other radionuclides in samples collected in Rhode Island during national studies by Hess

Table 33. Geohydrologic data and concentrations of radium and uranium in water samples from wells and springs in Rhode Island

[Data from Scott and Barker (1962); locations shown in fig. 82; —, no data; <, less than; pCi/L, picocuries per liter; gal/min, gallons per minute]

Location		Geohydrologic data			Concentration (pCi/L)		
Town	County	Water-bearing unit	Well yield (gal/min)	Date of collection	Gross-beta activity	Radium	Uranium[1]
East Providence.....	Providence	Rhode Island Formation	27	06–21–54	<10	0.4	0.6
East Providence.....	Providence	Outwash	1,200	04–08–54	<5	<.1	.07
West Warwick......	Kent	Cowesset Granite	3	06–26–55	15	.4	.9
Alton	Washington	Outwash	—	11–07–55	<10	1.2	<.07

[1] Converted from micrograms per liter (µg/L) by multiplying by 0.68 and assuming that all activity is due to uranium and that uranium is in secular equilibrium.

and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of 100 to 10,000 pCi/L for ^{222}Rn and 1 to 5 pCi/L for ^{226}Ra and total uranium in the drinking water from selected counties of Rhode Island. Table 33 includes concentrations of gross-beta radiation, total radium, and uranium in water from selected wells in Rhode Island.

Preliminary results of 260 analyses of drinking water from private wells for ^{222}Rn by the Rhode Island Department of Environmental Management show that 13 percent had concentrations greater than 10,000 pCi/L. The concentrations ranged from 140 to 49,080 pCi/L and averaged 5,220 pCi/L (Margaret Bradley, Rhode Island Department of Environmental Management, written commun., 1988).

Ground-Water Availability and Rate of Withdrawal

Data on ground water withdrawals (Johnston, 1985) indicate the percentage of the population that is exposed to potential health risk from radionuclides in the water supply. The major aquifers in Rhode Island consist of stratified-drift deposits that have well yields ranging from 100 to 700 gal/min. The crystalline-bedrock aquifers of Rhode Island are widely used for water supply and may have the highest concentrations of ^{222}Rn; well yields from bedrock aquifers range from 1 to 20 gal/min. The 224,000 residents of Rhode Island that rely on ground water as a drinking supply, 9 percent of which rely on small or private systems, withdraw 24 Mgal/d.

Radon in Indoor Air

Information on ^{222}Rn in indoor air in Rhode Island can be obtained through direct inquiry to the Rhode Island Department of Public Health, Division of Occupational Health. Although some indoor-air measurements have been made in Rhode Island, they are insufficient for statistical analysis.

CONNECTICUT

Previous Work

Research on radioactivity in the environment in Connecticut dates back to the early 1900's, when Bumstead and Wheeler (1903, 1904) investigated radioactive gas in the soil and ground water near New Haven (fig. 83). They concluded that the concentration of ^{222}Rn was not in equilibrium with the concentration of the radioactive parent ^{226}Ra. Systematic studies of natural radioactivity in Connecticut began in the early 1950's as ground measurements of gross radioactivity by McKeown (1951). Johnson (1951) delineated a region of radioactive rocks he called the "Central New England Radioactive Province," which included part of central and eastern Connecticut. Popenoe (1966) compiled a generalized map correlating aeroradioactivity with geology in Connecticut as well as New York, Rhode Island, and Maine.

Goldsmith and others (1977) further investigated radioactivity in the rocks of Connecticut and correlated aeroradioactivity data with geologic data from southeastern Connecticut. They found that the Sterling Plutonic Group, the Narragansett Pier Granite, and the Westerly Granite all have fields of relatively high natural radioactivity. In contrast, the Tatnic Hill and Hebron Formations, as well as the Scotland Schist and Canterbury Gneiss, have fields of relatively moderate natural radioactivity, and the Quinebaug Formation and the Preston Gabbro had relatively low natural radioactivity. Jones (1979) analyzed National Uranium Resource Evaluation data for Connecticut with emphasis on results from the hydrogeochemical and stream-sediment surveys. Krishanaswami and others (1982) investigated radium, thorium, and radioactive lead isotopes in

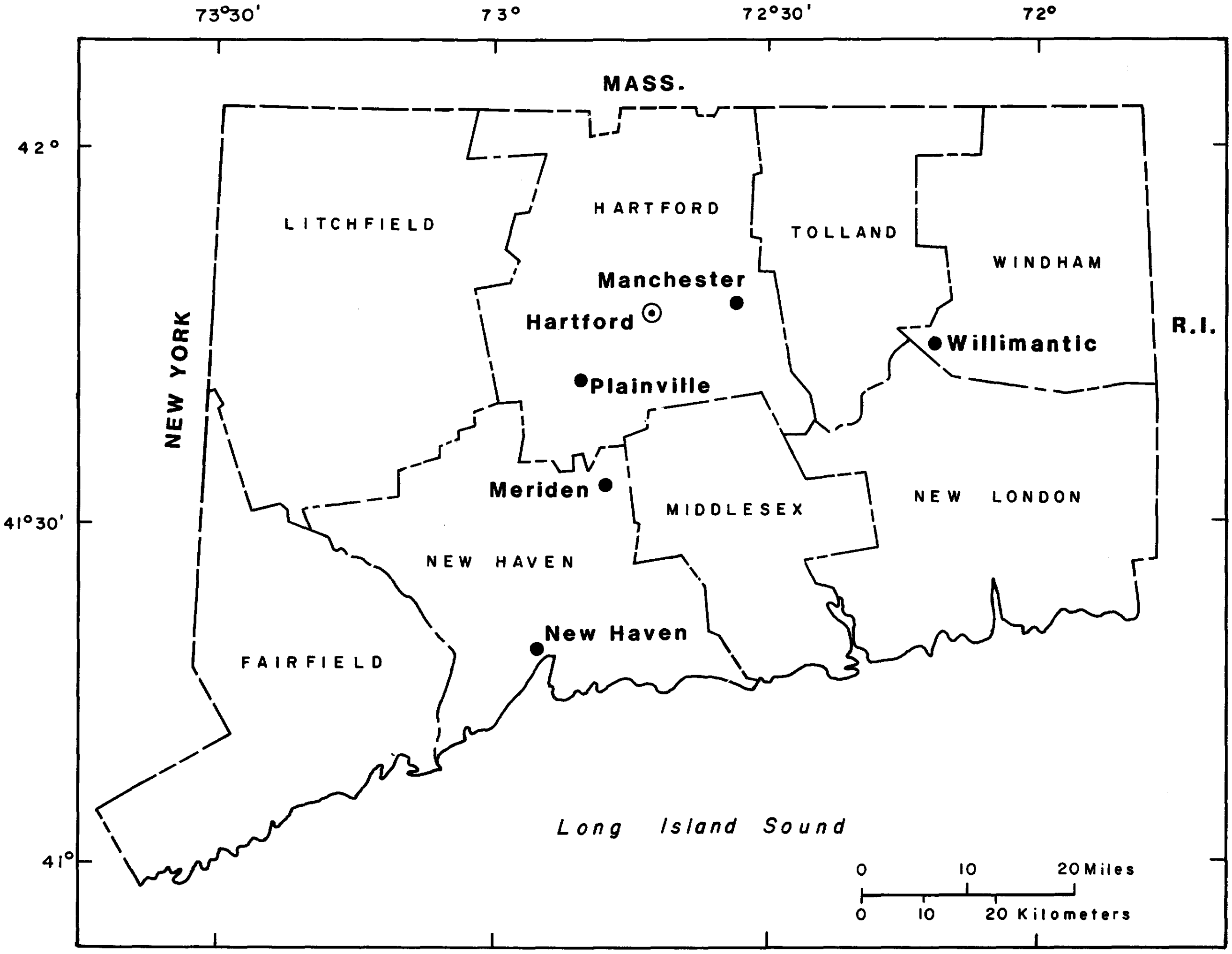


Figure 83. Locations of Connecticut sites mentioned in text. (Scale 1:1,050,000; base from Meade, 1978.)

ground water of Connecticut and concluded that some process was removing ^{226}Ra from the ground water.

In the mid-1980's, the Connecticut Department of Health Services began a study to investigate ^{222}Rn in water and air in Connecticut. In 1985, they sampled 63 domestic bedrock wells for ^{222}Rn by liquid-scintillation techniques (Thomas, 1987) and made winter indoor-air measurements with alpha Track-Etch detectors in homes that had well-water concentrations of ^{222}Rn exceeding 10,000 pCi/L (approximately 25 percent of the original 63 homes). Of the homes tested, 15 percent had ^{222}Rn concentrations exceeding 4.0 pCi/L (Thomas, 1987).

The results of the work done in 1985 indicated that additional investigations were needed. In 1986 the Connecticut Department of Health Services began an expanded survey that included the sampling of 199 domestic wells from 26 different geologic formations totaling 45 rock units (Rothney, 1987a; Thomas, 1987). Additionally, 202 indoor-air measurements for ^{222}Rn were made during three winter months by alpha Track-Etch detectors at each of the homes at which ground water was being tested.

In late 1986 the Connecticut Department of Environmental Protection's Natural Resource Center began a project to measure the background levels of gamma radiation in a ground survey (Thomas, 1987, p. 354) that covered 1,367 mi (miles) across all major geologic terranes in Connecticut. The results correlated well with the ground-water testing program—areas that had high natural gamma-radiation fields tended to have ground water with higher ^{222}Rn concentrations, and areas that had low natural gamma-radiation fields tended to have lower ground water ^{222}Rn concentrations (Thomas, 1987).

Rothney (1987b) summarized the occurrence of ^{222}Rn in domestic wells in Connecticut in relation to geologic and selected hydrologic characteristics and expanded her previous work (Rothney, 1987a) to include the results of the indoor-air survey. In 1988, an expanded indoor-air survey sponsored by the Environmental Protection Agency used charcoal-canister-type detectors in an additional 1,570

Table 34. Geohydrologic data and concentrations of radium and uranium in water samples from wells and a spring in Connecticut

[Data from Scott and Barker (1962); locations shown in fig. 83; <, less than; pCi/L, picocuries per liter; gal/min, gallons per minute]

Location		Geohydrologic data			Concentration (pCi/L)		
Town	County	Water-bearing unit	Well yield (gal/min)	Date of collection	Gross-beta activity	Radium	Uranium[1]
Stratton Brook......	Hartford	Glacial outwash	300	10–26–55	<5	<0.1	0.07
Manchester.........	Hartford	Continental arkose	457	11–04–54	<10	.1	.4
Willimantic.........	Windham	Willimantic Gneiss	10	10–25–54	<10	<.1	.3
Plainville...........	Hartford	Glacial sand and gravel	500	03–06–57	<5	.1	.3
Meriden............	New Haven	Newark Supergroup	1.5 (spring)	06–24–54	<10	<.1	.5

[1] Converted from micrograms per liter (μg/L) by multiplying by 0.68 and assuming that all activity is due to uranium and that uranium is in secular equilibrium.

homes (Margaret Thomas, Connecticut Department of Environemntal Protection, Natural Resource Center, written commun., 1988). The Connecticut Department of Environmental Protection's Natural Resource Center used the department's geographic information system to analyze radionuclide data and develop area ^{222}Rn assessments for the Connecticut Department of Health Services (Thomas, 1988).

Distribution and Concentration of Radionuclides in Ground Water

Data on ^{222}Rn and other radionuclides in samples collected in Connecticut during national studies by Hess and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of 100 to 1,000 pCi/L for ^{222}Rn, 1 to 5 pCi/L for ^{226}Ra, and less than 1 to 5 pCi/L for total uranium in the drinking water from selected counties of Connecticut. Table 34 includes concentrations of gross-beta radiation, total radium, and uranium in water from selected wells in Connecticut.

Hess and others (1985b) note that three public water supplies in Connecticut had total radium concentrations that exceeded 5 pCi/L. During a 1979 sampling of 115 public water-supply wells by the U.S. Geological Survey, 17 samples had gross-alpha concentrations exceeding 15 pCi/L, and 5 had dissolved radium concentrations exceeding 5 pCi/L (Robert Melvin, U.S. Geological Survey, written commun., 1988). Table 35 presents the results of the ^{222}Rn in water survey of 202 domestic wells sampled from 1985 to 1987.

Ground-Water Availability and Rate of Withdrawal

Data on ground water withdrawals (Melvin, 1985) indicate the percentage of the population that is exposed to potential health risk from radionuclides in the water supply in Connecticut. The major aquifers in Connecticut consist of stratified-drift deposits that have well yields ranging from 50 to 500 gal/min. The crystalline-bedrock aquifers of Connecticut are widely used for water supply and also have the highest concentrations of ^{222}Rn. Well yields from bedrock aquifers range from 1 to 50 gal/min. The 984,000 residents of Connecticut that rely on ground water as a drinking supply, 18 percent of which rely on small or private systems, withdraw 118 Mgal/d.

Radon in Indoor Air

Data on ^{222}Rn in indoor air in Connecticut are extensive. During the 1985 pilot study, the concentrations in homes above granitic areas in which ^{222}Rn in ground water exceeded 10,000 pCi/L (Thomas, 1987) ranged from 0.61 to 7.80 pCi/L, with a median of 2.06 pCi/L. In the 1986 study, where 202 alpha Track-Etch detectors were placed in homes across the State, a mean of 1.97 pCi/L was calculated, and concentrations ranged from 0.1 to 24.6 pCi/L, with 12 percent exceeding 4 pCi/L. Table 36 summarizes preliminary results of the 1987 indoor-air survey of 1,570 homes where ^{222}Rn was measured with 2-day exposure charcoal-canister-type detectors.

NEW YORK

Previous Work

The first studies of natural radioactivity in New York were begun in the early 1950's as ground measurements of gross radioactivity by McKeown (1951), Narten and McKeown (1952), and McKeown and Klemic (1953a,b). Klemic and others (1959) have investigated in some detail the Phillips uranium mine in the Camp Smith area in southeastern New York (fig. 84). Swanson (1960) mea-

Table 35. Results of the 1985–87 well-water survey of 202 homes in Connecticut for radon-222

[Data from Rothney (1987b); pCi/L, picocuries per liter]

Geologic terrane[1]	Number of homes	Range (pCi/L) Minimum	Range (pCi/L) Maximum	Geometric mean
Sedimentary basins	44	230	11,620	1,335
Extrusive igneous within sedimentary basins	5	470	2,980	950
Two-mica granites	12	10,720	130,240	28,254
General gneiss	37	180	48,820	3,160
Granitic gneiss	63	420	87,450	7,495
Intermediate to mafic gneiss	11	870	22,300	4,638
Stratified metamorphics	61	370	36,560	2,937
Carbonate within stratified metamorphics	9	200	4,130	746
Quartzite within statified metamorphics	3	100	14,780	4,234
General mixture	16	230	14,470	1,393

[1] Based on classification scheme of Olszewski and Boudette (1986).

Table 36. Summary of radon-222 in indoor air in Connecticut during the winter of 1986–87

[Data from Rothney (1987b, p. 27); pCi/L, picocuries per liter]

Geologic terrane[1]	Number of tests	Percentage of total exceeding 4 pCi/L	Percentage of total exceeding 10 pCi/L	Percentage of total exceeding 20 pCi/L
Merrimack synclinorium	271	26.2	8.1	2.2
Connecticut Valley synclinorium	410	22.9	4.6	1.2
Bronson Hill anticlinorium	107	21.5	2.8	1.9
Avalonian	207	19.3	4.4	.5
Carbonate shelf	51	21.6	5.9	0
Taconic allochthons	46	8.7	4.3	0
Hartford and Pomperaug basins (Newark terranes)	295	9.4	1.7	0
Proterozic massifs	36	13.9	0	0
Total	1,423			

[1] From Rodgers (1985).

sured the uranium content of black shales (probably the Dunkirk member) in southwestern New York. These three measurements show a range of 0.001 to 0.006 percent uranium by weight. Popenoe (1966) compared areoradioactivity surveys with the geology of southeastern New York and noted relatively high background radioactivity in the Hudson Highlands near West Point (fig. 84) and noted above-background radioactivity in the till that forms the terminal moraines on Long Island.

Campbell and others (1979) updated some basic geologic data on the Phillips mine in the Camp Smith area through geophysical techniques including radiometric, magnetic, and very-low-frequency resistivity surveys. Van Tyne and others (1980a–f) mapped the thickness of the radioactive black shales in the Hamilton, Genesee, Sonyea, West Falls, and Java Groups and the Perrysburg Formation of western New York from gamma-ray logs. Nutt (1982) produced a comprehensive review of uranium occurrence in the Precambrian rocks of the Adirondack region in northern New York. Fleischer and others (1982) investigated ^{222}Rn in energy-efficient homes in New York with 6-month, alpha Track-Etch measurements during both winter and summer periods.

Kothari (1983) noted that the National Uranium Resource Evaluation data for Lewiston (fig. 84) suggest a 1.5-ppm uranium equivalence at land surface and that the measured ^{222}Rn in homes in that area ranged from 0.34 to 0.79 pCi/L. Fleischer and Turner (1984a) surveyed an area in Albany and Schenectady Counties with radiometric ^{222}Rn, isotopic, and chemical techniques to investigate a part of the Eastern Overthrust Belt as a possible hydrocarbon source area. Fleischer and Turner (1984b) investigated indoor-air levels of ^{222}Rn in energy-efficient homes in the Albany area for 1 year with alpha Track-Etch detectors. The New York State Department of Health (1985) published a generalized fact sheet on ^{222}Rn in ground water.

The Hudson Highlands of New York (fig. 84) have mineral assemblages and geologic structure that are similar to the rock of the Reading Prong in Pennsylvania (Gundersen, 1986). The U.S. Geological Survey has developed a geologic and metallogenic model for uranium

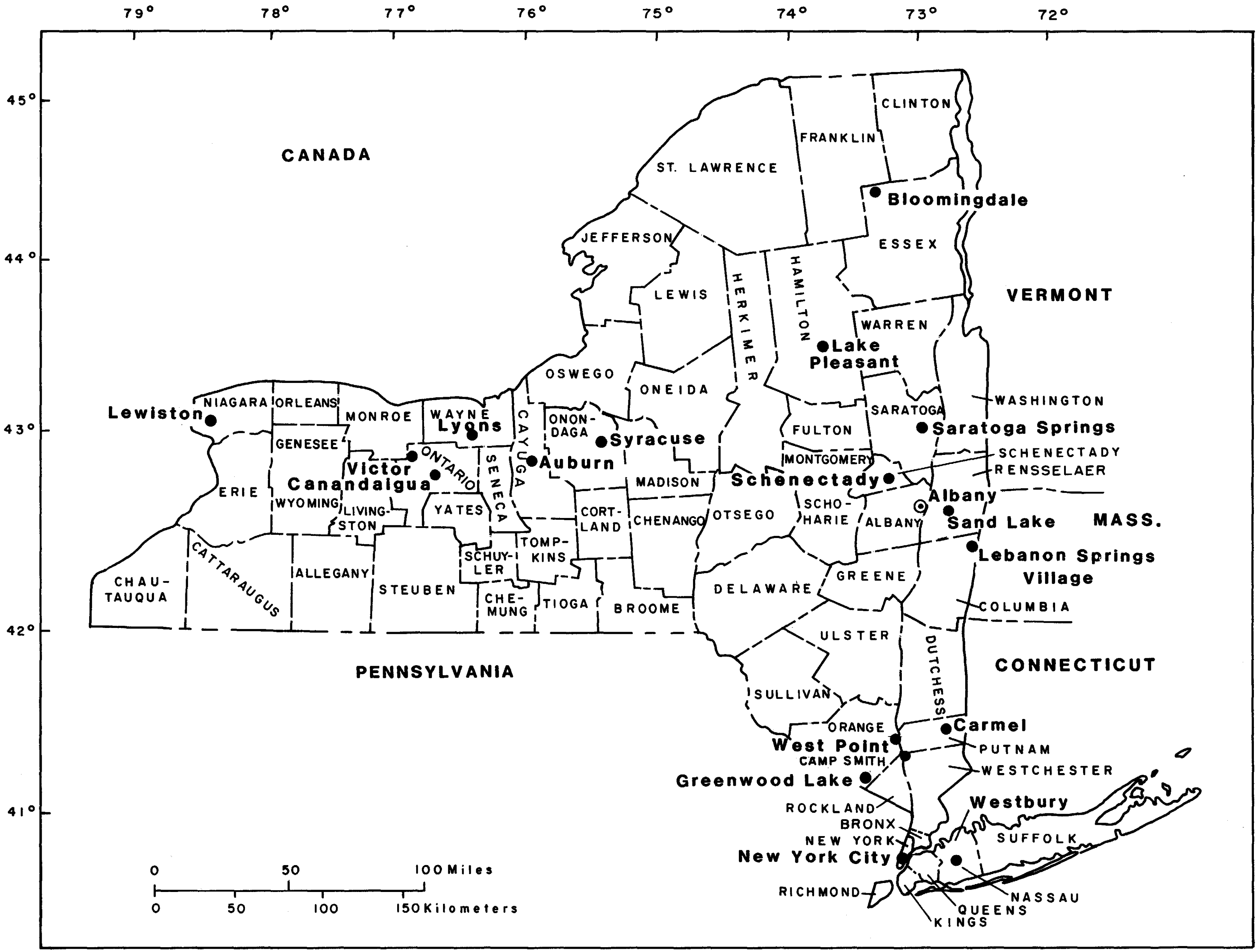


Figure 84. Locations of New York sites mentioned in text. (Scale 1:3,727,059; base from Heath, 1964.)

mineralization in the Hudson Highlands and Reading Prong in New Jersey and Pennsylvania (L.C.S. Gundersen, U.S. Geological Survey, oral commun., 1986).

The New York State Energy Research and Development Authority (1981, 1983, 1984) investigated low-temperature geothermal potentials near Albany, Saratoga Springs, and Auburn (fig. 84). The source of the higher-than-expected geothermal gradient of 22.96 °C per kilometer is not clearly defined, but these geothermal wells are finished in the Precambrian-age igneous and metamorphic basement rock of New York, and heating due to radioactive decay has not been ruled out. The New York State Energy Research and Development Authority (1983, p. 3–3) reports anomalously high helium concentrations in samples from many of these wells and a ^{222}Rn concentration of 350 pCi/L in a sample of spring water from Saratoga Springs.

The New York State Department of Health (Kunz and Kothari, 1986; Lilley and others, 1987) and the New York State Energy Research and Development Authority (American Geological Institute, 1987a) show that the ^{222}Rn concentration in the air in private homes in several counties exceed the recommended 4 pCi/L guideline. Kunz and Kothari (1986) measured ^{222}Rn concentrations in soil gas at a depth of 3 to 4 ft across the State; the values ranged from 100 to 50,000 pCi/L. They also measured the ^{226}Ra content of soils across the State and found average concentrations ranging from 0.46 pCi/g in sandy soils on Long Island to 2.2 pCi/g in soils overlying the Marcellus Shale in western New York. Preliminary results of measurements of ^{222}Rn in drinking water from 132 large public supplies suggest that the concentrations in all sources were lower than 800 pCi/L, although the range of the measurements in wells samples across New York State was 100 to 350,000 pCi/L.

Banikowski and Hand (1987) completed a preliminary study of ^{222}Rn in indoor air in the basements of 210 homes in Onondaga County during a 4-day test with charcoal-canister-type detectors. Results suggest that the Marcellus Shale, a black shale of the Middle Devonian Hamilton Group, is a source rock for ^{222}Rn in indoor air. None of the 100 homes tested that were underlain by the Marcellus Shale had indoor-air levels of ^{222}Rn below 4 pCi/L, and the average indoor-air concentration was 8.8 pCi/L. The New York State Department of Health measured air in basements of 15 homes underlain by Marcellus Shale and found that 4 had levels less than 4 pCi/L (Charles Kunz, New York State Department of Health, written commun., 1988). The U.S. Military Academy at West Point (fig. 84), in the Hudson Highlands near the Phillips mine, began a 10-week indoor-air testing program for buildings in the Academy with charcoal-canister-type detectors (American Geological Institute, 1987b; Frieland and Cherry, 1987). Data for gross-alpha radiation and ^{222}Rn in ground water from the Academy were collected but are unavailable at present.

Bergeron and others (1987) and Yager (1987) report on geohydrologic conditions and ground-water-flow modeling at the nuclear-fuels-reprocessing plant and waste-management facility at the western New York Nuclear Service Center in Cattaraugus County. Chemical analyses for some less common radionuclides were completed, but no analyses were made for ^{238,234}U, ^{226,228}Ra, or ^{222}Rn in ground water.

Nero (1983) found that the average range of ^{222}Rn in 18 single-family residences in New York was 1.0 to 6.4 pCi/L. Results of a national study of ^{222}Rn in indoor air that included some data for New York indicated concentrations of 0.52 to 1.57 pCi/L (Nero and others, 1986). Fisenne and others (1987) reviewed the daily intake of ^{234,235,238}U, ^{228,230,232}Th, and ^{226,228}Ra by New York City residents and found that neither the ingestion of food or water nor the inhalation of air constituted a health threat because the levels did not exceed present standards.

Distribution and Concentration of Radionuclides in Ground Water

Data on ^{222}Rn and other radionuclides in samples collected in New York during national studies by Hess and others (1985b) and Horton (1983, 1985) are included in tables 25 and 26, which also give national averages for comparison. Michel and Jordana (1987) list a range of less than 100 to 1,000 pCi/L for ^{222}Rn, less than 1 to 5 pCi/L for ^{226}Ra, and less than 1 pCi/L for total uranium in the drinking water from selected counties of New York. Table 37 includes concentrations of gross-beta radiation, total radium, and uranium in water from selected wells in New York. The New York State Department of Health (1985) fact sheet on ^{222}Rn in drinking water notes that ^{222}Rn concentrations in drinking water from 132 large public supplies were lower than 800 pCi/L, although one well in Orange County in the Hudson Highlands had a ^{222}Rn concentration of 350,000 pCi/L (Charles Kunz, New York State Department of Health, written commun., 1988).

Ground-Water Availability and Rate of Withdrawal

Data on ground water withdrawals (Waller and others, 1985) indicate what percentage of the population is exposed to potential health risk from radionuclides in the water supply. The 6,133,000 residents of New York that rely on ground water as a drinking supply, 13 percent of which rely on small or private systems, withdraw 670 Mgal/d. About 486 Mgal/d are withdrawn from the aquifers on Long Island, whose aquifers are unconsolidated coastal-

Table 37. Geohydrologic data and concentrations of radium and uranium in water samples from wells and a spring in New York

[Data from Scott and Barker (1962); locations shown in fig. 84; —, no data; <, less than; pCi/L, picocuries per liter; gal/min, gallons per minute]

Location		Geohydrologic data			Concentration (pCi/L)		
Town	County	Water-bearing unit	Well yield (gal/min)	Date of collection	Gross-beta activity	Radium	Uranium[1]
Bloomingdale	Essex	Grenville Complex	—	06–07–54	<10	<0.1	0.7
Lake Pleasant Village	Hamilton	Syenite	10	10–05–54	<5	.1	.4
Lyons	Wayne	Salina Group	150	05–03–56	<1,000	11	4.1
Syracuse	Onondaga	Camillus Shale, Vernon Shale	75	05–03–56	<50	<.1	1.4
Victor	Ontario	Kame deposit	200+ (spring)	05–03–56	<20	<.1	.4
Canandaigua	Ontario	Ludlowville Formation	15	07–12–54	<50	1.0	.7
Sand Lake	Rensselaer	Rensselaer Graywacke	1	09–20–54	<25	.1	3.4
Lebanon Springs Village	Columbia	Stockbridge Limestone	75	09–20–54	<10	.1	.9
Carmel	Putnam	Granitic gneiss	.4	05–12–54	<25	1.8	1.0
Greenwood Lake	Orange	Granitic gneiss	3	09–15–54	<5	.1	.3
Westbury	Nassau	Magothy Formation	1,050	03–26–57	<5	<.1	.2
Westbury	Nassau	Llyod Sand Member of Raritan Formation	1,000	03–26–57	<5	<.1	.7

[1] Converted from micrograms per liter (μg/L) by multiplying by 0.68 and assuming that all activity is due to uranium and that uranium is in secular equilibrium.

plain deposits. Well yields there range from 50 to 1,200 gal/min and are considered to be free of radionuclides. The major upstate aquifers consist of stratified-drift deposits that have well yields ranging from 10 to 1,000 gal/min. The sedimentary and carbonate bedrock aquifers of New York are also widely used for water supply and may have high concentrations of ^{222}Rn. Well yields from crystalline-bedrock aquifers range from 50 to 150 gal/min.

Radon in Indoor Air

The New York State Department of Health and the New York State Energy Research and Development Authority investigated ^{222}Rn in indoor air in 2,400 homes in 1986 (table 38). The statewide mean for ^{222}Rn in indoor air was 1.39 pCi/L, and the range was from 0.06 to 39.8 pCi/L.

Soil-gas measurements of ^{222}Rn have also been made across the State (Charles Kunz, New York Department of Health, oral commun., 1988); the average concentration in samples taken at a depth of between 3 and 4 ft was about 700 pCi/L, and the range was from 100 to 50,000 pCi/L. Data for ^{226}Ra in soil yielded a statewide average of 1.0 pCi/g and a range of 0.2 to 20 pCi/g. Some homes in upstate New York that have high concentrations of ^{222}Rn in indoor air have average ^{226}Ra concentrations in the underlying soils.

Banikowski and Hand (1987) reported that the geometric mean concentration of ^{222}Rn in indoor air in basements in 100 homes over the Marcellus Shale in central New York was 8.8 pCi/L. The mean for homes overlying the Upper Silurian shales and dolostones was 2.5 pCi/L. The mean for homes overlying the upper Middle Devonian sandstones and shales was 1.2 pCi/L.

CANADA—SOUTHERN ONTARIO

Radionuclides in the southern part of the Province of Ontario, Canada (fig. 77), are included in this report for two reasons. First, some of the most detailed data for uranium, ^{226}Ra, and ^{222}Rn in ground water from Cambrian through Devonian marine sedimentary rocks are from Canadian research; these rocks occur in New York State and parts of New England. Canadian researchers also have investigated the Precambrian crystalline rocks of the Canadian Shield for radionuclide content. These sedimentary and crystalline rocks are commonly found in the stratified drift and other glacial deposits that form the most productive aquifers in New England. In addition, many investigators extend their work only to the borders of the United States, so "blank" zones appear on geologic maps in areas just beyond the United States border. Because the geologic terranes continue across national boundaries, information from both sides should be included where possible. This section provides some general information.

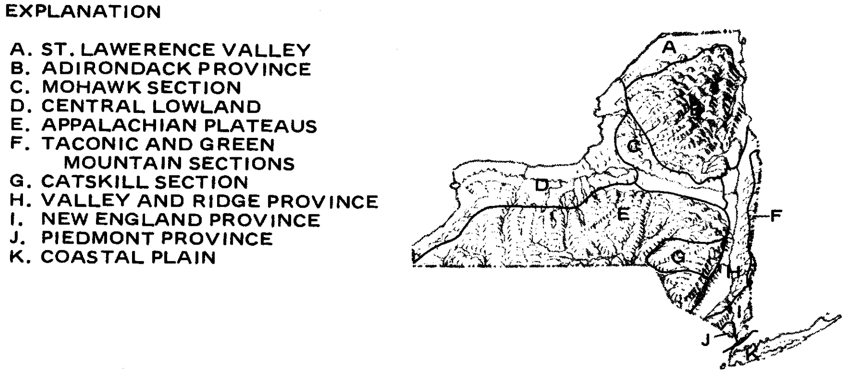
Previous Work

The Joint Federal-Provincial working group on drinking water proposes drinking-water standards, as does the

Table 38. Concentrations of radon-222 in indoor air in New York by region and stratigraphic units

[Data from the New York State Department of Health (1985); locations shown in inset map (from U.S. Geological Survey, 1985); physiographic diagram from Raisz (1954); divisions from Fenneman (1946); pCi/L, picocuries per liter]

Region and geologic units	Number of samples	Concentrations, in pCi/L	
		Mean	Range
Appalachian Plateaus (Devonian shales and sandstones)	346	3.34	0.09–39.8
Central Lowland (Devonian shale, Silurian shale and dolomite, Ordovician limestone)	767	1.58	.07–28.4
Valley and Ridge Province (Cambrian and Ordovician shales, limestones, and sandstones of the Catskill Delta Sequence)	545	1.09	.06–21.6
Adirondack Province (Precambrian igneous and metamorphic rocks)	276	1.82	.08–20.9
Piedmont Province—Staten Island (Precambrian igneous and metamorphic rocks)	51	.75	.19–2.4
Piedmont Province—New York City (Precambrian igneous and metamorphic rocks)	81	.81	.08–2.4
Coastal Plain (Quaternary glacial surficial deposits)	335	0.87	.07–3.4



Environmental Protection Agency in the United States, and the Ontario Ministry of the Environment investigates the occurrence and distribution of radionuclides in drinking water (Beck and Brown, 1987, p. 450). Although some detailed investigations of radionuclides in the Canadian Shield and sedimentary rock environments have been made, no drinking-water standard for ^{222}Rn has been set. Drinking water standards for total uranium and ^{226}Ra have been set at 20 μg/L and 27 pCi/L, respectively (Beck and Brown, 1987, p. 450–451).

McGregor and others (1980) measured ^{222}Rn in indoor air in about 10,000 homes from 14 Canadian cities from June through August of 1977 and 1978. Indoor air was drawn through a Lucas-type cell (alpha-scintillation counting cell) for 5 min at a constant rate of 0.3 L/s (liters per second). Results indicated that generally fewer than 10 percent of the homes tested had indoor air concentrations of ^{222}Rn exceeding 4.0 pCi/L. Work prepared for the Atomic Energy Control Board of Canda (1983) showed that ^{226}Ra in the unconsolidated materials in Canada ranged from 7.0 pCi/g for clays to 0.2 pCi/g for sand at Bancroft (fig. 85). Till from Niagara Falls had a ^{226}Ra concentration of 0.7 pCi/g.

Lemire and Gascoyne (1987) reported on the occurrence and distribution of uranium, ^{226}Ra, and ^{222}Rn in surface waters and ground waters at five nuclear-research laboratories across Canada. These surveys were undertaken to document the natural background levels of radioactivity at the laboratories. Beck and Brown (1987) analyzed 166 ground-water samples (29 from municipal sources and 137 from private supplies) for total uranium, ^{226}Ra, and ^{222}Rn. Two regions were sampled extensively. One, which is near Bancroft, is underlain by the rocks of the Precambrian Canadian Shield, and the other, which is near the eastern shore of Lake Huron, is underlain by limestones and dolomites of the Middle Devonian Detroit River Group.

Distribution and Concentration of Radionuclides in Water

The international system (SI) unit for radiation is the Becquerel (Bq). One Curie is equal to 3.7×10^{10} Bq; 1 pCi is equal to 3.7×10^{-2} Bq (Durrance, 1987, p. 69.). For consistency, all reported concentrations have been converted to picocuries per liter for ^{226}Ra and ^{222}Rn and micrograms per liter for uranium.

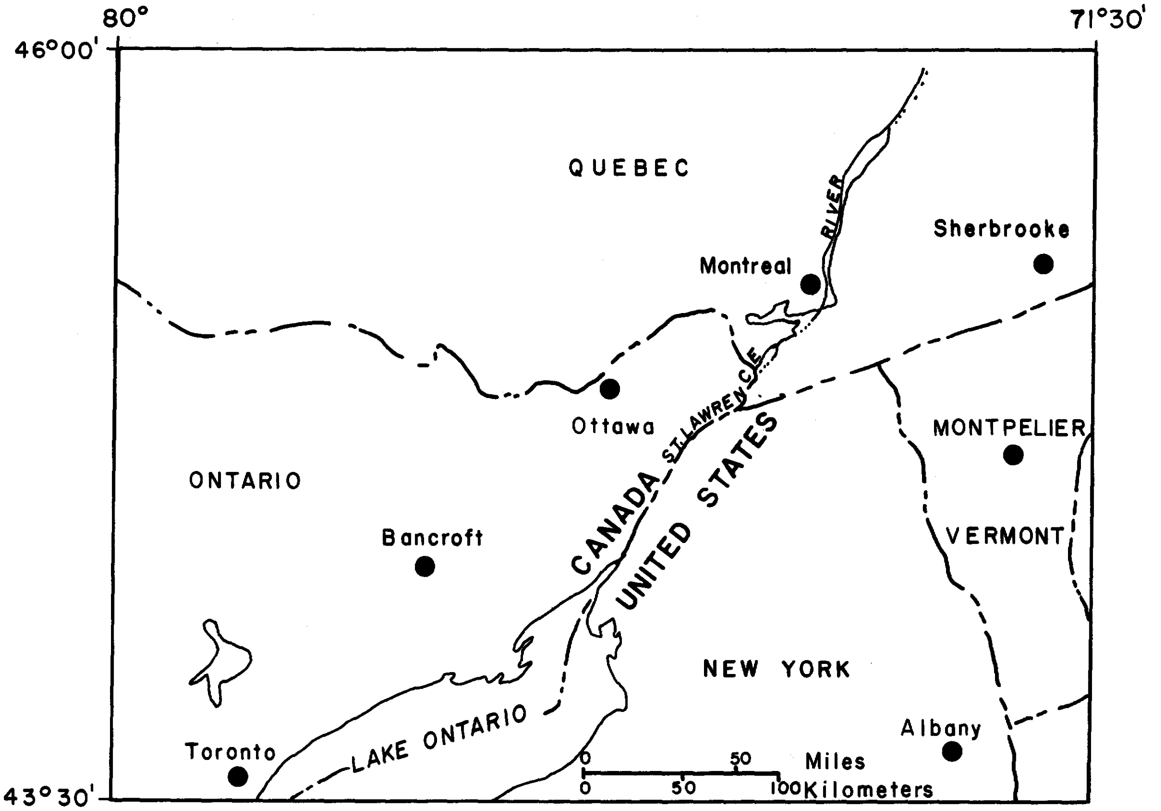


Figure 85. Locations of southern Ontario sites mentioned in text.

Lemire and Gascoyne (1987, p. 108) present ranges of concentrations of uranium, ^{226}Ra, and ^{222}Rn in surface water and ground water. The values for uranium in surface water were generally less than 0.2 μg/L and in ground water less than 0.1 to 10.2 μg/L; ^{226}Ra in surface water ranged from less than 0.5 to 18.9 pCi/L and in ground water less than 0.5 to 1.1 pCi/L; ^{222}Rn in surface water ranged from less than 2.7 to 8.1 pCi/L and in ground water from less than 2.7 to 13,100 pCi/L.

Beck and Brown (1987) reported that uranium concentrations in samples from wells finished in the Canadian Shield rocks near Bancroft ranged from less than 2 to 74.8 pCi/L, ^{226}Ra concentrations ranged from less than 1 to 25 pCi/L, and ^{222}Rn concentrations ranged from 1 to 44,600 pCi/L. They also reported that, for wells finished in the rocks of the Detroit River Group, uranium concentrations ranged from less than 2 to 26.5 pCi/L, ^{226}Ra concentrations from less than 1 to 18.4 pCi/L, and ^{222}Rn concentrations from 140 to 11,000 pCi/L.

Radon in Indoor Air in Toronto, Montreal, Sherbrooke, and Quebec

McGregor and others (1980) presented data on indoor-air concentrations of ^{222}Rn for 14 cities. Four of these—Toronto, Montreal, Sherbrooke, and Quebec—are discussed here because they are close to the U.S. border. The geometric means of ^{222}Rn in indoor air were 0.31 pCi/L in Toronto, 0.29 pCi/L in Montreal, 0.36 pCi/L in Sherbrooke, and 0.28 pCi/L in Quebec. Of the homes measured, 99.9, 98.2, 90.6, and 95.9 percent of the homes in Toronto, Montreal, Sherbrooke, and Quebec, respectively, had indoor-air levels of ^{222}Rn below 4.0 pCi/L. Information on ^{222}Rn in indoor air in Canada can be obtained through direct inquiry to the Department of National Health and Welfare, Ottawa, Ontario.

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PART 2: STUDIES OF RADON IN WATER

SECTION 2: TECHNIQUES FOR MEASURING RADON IN WATER

SAMPLING AND ANALYSIS OF DISSOLVED RADON-222 IN WATER BY THE DE-EMANATION METHOD

In Che Yang

Abstract

Collection of surface- or ground-water samples for determining the concentration of dissolved radon gas is best performed by minimizing aeration of water to be sampled. Radon loss during transportation from the sampling site to the laboratory (within 1 week) was negligible for low-activity samples and was 4 percent for high-activity samples. Radon loss during storage for 1 week is negligible. The direct de-emanation and alpha-scintillation counting method is sensitive (0.2 picocurie per liter for a 60-milliliter sample) and has a better precision than other methods for analysis of most environmental samples.

INTRODUCTION

Radon-222 (^{222}Rn) is a radioactive gas known to occur in ground water. The gas diffuses throughout a ground-water aquifer, and its concentration can be many orders of magnitude larger than the concentration of radium or uranium. Radon-222 is a daughter product of radium-226 (^{226}Ra) and has a short half-life (3.8 days); however, long-lived components of the decay products (those of environmental concern) are lead-210 (^{210}Pb, half-life of 22 years and a weak beta emitter) and Polonium-210 (^{210}Po, half-life of 138.4 days and an alpha emitter). The method by which ^{222}Rn enters the body tissue of a person is not well understood. A maximum contaminant level for ^{222}Rn in drinking water is under consideration by the Environmental Protection Agency (Environmental Protection Agency, 1983).

Sampling and measurement of ^{222}Rn are complicated by the volatility of the gaseous element. For example, ^{222}Rn in water appears to be removed effectively by aeration. Therefore, when water is sampled for ^{222}Rn analysis, aeration of the water needs to be minimized, and transfer of samples through the open air needs to be avoided. Loss of ^{222}Rn during transportation and storage may occur if sampling vessels are permeable to gas, or if they have leaky stopcocks and caps. This paper describes the direct de-emanation and alpha-scintillation count method used for accurate analysis of dissolved radon gas in surface and ground water. In addition, sampling techniques and possible loss of radon during the sample transport are also discussed.

EXPERIMENTAL METHODS

Water Sampling

An all-glass de-emanation bubbler (fig. 86) is used for collecting water samples for analysis of ^{222}Rn. Before sampling, evacuate the bubbler (assembled as shown in fig. 86) to less than 10 mtorr (millitorr) in the laboratory or laboratory trailer through stopcock 1 (stopcock 2 is closed). Then, close stopcock 1. Gently place the evacuated sample bubbler into the surface water until the space between stopcock 1 and the inlet port is filled with water. Open stopcock 1 to draw sample water directly into the main chamber of the bubbler, until the chamber is 60 percent full. Close stopcock 1. Label bubbler with sample number, place of collection, and collection time (day, hour, and minute). Use masking tape to secure stopcocks 1 and 2 in the closed position. Place bubbler inside a foam-packed cardboard box and send by airmail to the laboratory as soon as possible. For ground-water samples, use a peristaltic pump or a submersible pump to supply an uncontaminated, full-column flow of sample water through a short, flexible tubing to the stopcock-1 end of the evacuated bubbler. When the main chamber is about 60 percent full (as shown in fig. 86), close stopcock 1. Label and transport samples to the laboratory in the same manner described for the surface-water samples.

Transportation and Storage

Collect two sets of source waters (one set from a low-activity source, less than 1,000 pCi/L (picocuries per liter), and the other set from a high-activity source, 6,000–7,000 pCi/L) by using three replicates of similar bubblers for this investigation. Analyze one replicate soon after sampling; send a second replicate to University of Texas Health Science Center at Houston, Tex., for return immediately after receipt (about 1 week for a round trip), and keep a third in the laboratory until the shipped replicate is returned. Analyze at the same time the samples that were sent away and the samples that were stored.

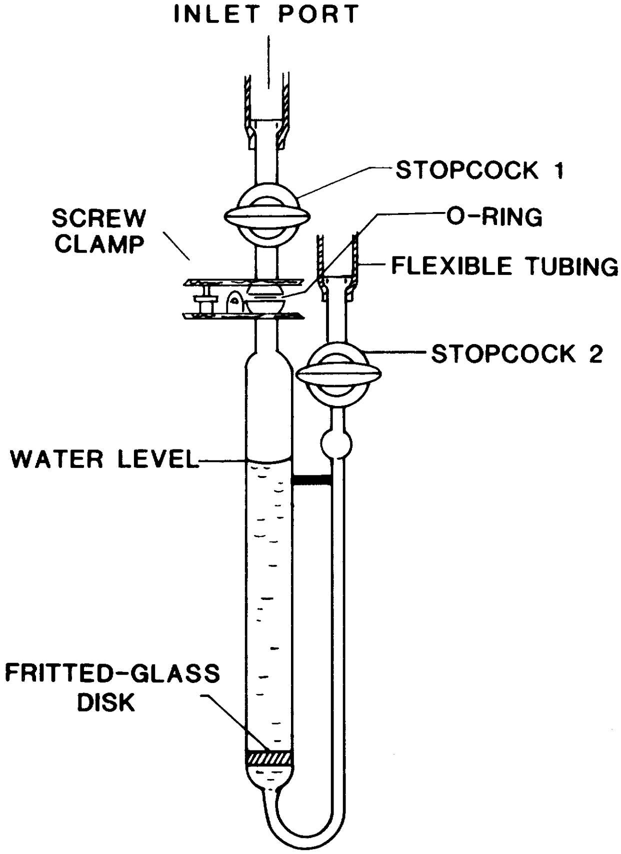

Figure 86. An all-glass de-emanation bubbler used for collecting water samples for analysis of radon-222.

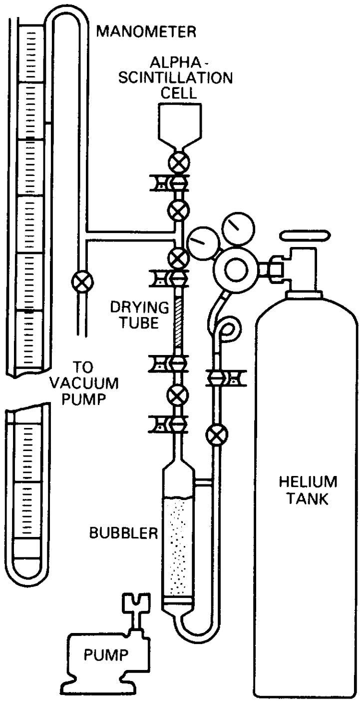

Figure 87. The radon de-emanation system.

Analytical Method

Radon-222 in surface water or lake water derives from two sources: one source is the in situ decay of ^{226}Ra already in water; the other source is external, such as ^{226}Ra in rocks and sediments or inflow of other water containing mainly dissolved ^{222}Rn with little parent ^{226}Ra (commonly called excess or nonsupported ^{222}Rn). To measure the excess or nonsupported ^{222}Rn, the quantity of ^{222}Rn in equilibrium with the ^{226}Ra has to be determined after the total ^{222}Rn measurement (Yang, 1984). The relation among various components of ^{222}Rn can be expressed in the following equation:

$$^{222}\text{Rn (total)} - {}^{222}\text{Rn (supported, in equilibrium with } {}^{226}\text{Ra)} = {}^{222}\text{Rn (nonsupported).}$$

Here the supported ^{222}Rn is assumed to be in equilibrium with the ^{226}Ra; the assumption is reasonable because of the short half-life of ^{222}Rn.

De-emanation

When ^{222}Rn samples are received in the laboratory, they should be analyzed immediately. Clamp the bubbler to the de-emanation system (fig. 87). This de-emanation system consists of the following parts: (1) a bubbler, (2) a glass drying tube packed with drierite (anhydrous magnesium perchlorate) to remove moisture and with ascarite (NaOH on asbestos plus soda lime) to remove CO_2, (3) a helium tank to purge dissolved ^{222}Rn into the cell, (4) an alpha-scintillation cell, (5) a manometer to indicate the pressure inside the system during transfer of ^{222}Rn from the bubbler to the alpha-scintillation cell and also to detect any leak in the system, and (6) a vacuum pump. With the bubbler stopcocks closed, evacuate the system and the alpha-scintillation cell to a vacuum of 10 mtorr. Close the stopcock to the vacuum pump and allow the system to equilibrate for about 2 min. If the manometer meniscus remains stable, connect the helium line (20.68–34.48 kilopascals) to the other side of the bubbler. Purge trapped air in the line between the helium tank and bubbler with helium gas by momentarily opening the O-ring connection on the helium-line inlet a few times. Record the time (day, hour, and minute), then carefully open the bubbler-outlet stopcock to purge ^{222}Rn directly into the alpha-scintillation cell. Allow the vacuum to equilibrate slowly; otherwise, an excessive risk exists of blowing the liquid sample from the bubbler into the drying tube. Continue the ^{222}Rn purging by slowly opening the bubbler-inlet stopcock; check the fritted-glass disk carefully for rising bubbles. Allow the pressure to increase slowly, control the manometer decrease rate, and complete purging in 10 to 15 min. Fill the cell to 4 millimeters of mercury less than the atmospheric pressure and close the cell stopcock. Close all other stopcocks in the

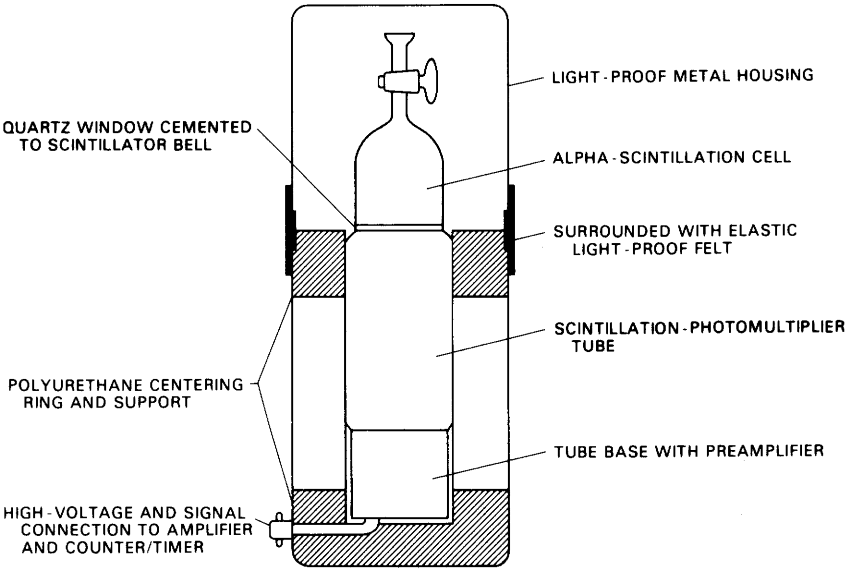

Figure 88. An alpha-scintillation cell and light-proof housing.

de-emanation system. Record the time (day, hour, minute). Quickly remove the bubbler from the assembly and open the outlet stopcock momentarily to release helium pressure. Turn off the room light; remove the alpha-scintillation cell for ^{222}Rn-activity measurement. Measure the water volume inside the bubbler by using a graduated cylinder.

Counting System for Radon-222 Activity

The ^{222}Rn-activity counting system consists of (1) an alpha-scintillation cell internally coated with silver-activated ZnS and having a quartz window 2 inches in diameter, (2) a scintillation-photomultiplier tube 3 inches in diameter, (3) a preamplifier connected to the scintillation-photomultiplier base, (4) an amplifier, (5) a scaler, counter, and timer, and (6) a high-voltage power supply.

Before counting, turn off the room lights; remove the light shield from the alpha-scintillation cell, and place the cell in the light-proof counting chamber (fig. 88). Allow about 20 min for the ^{222}Rn daughters to ingrow, then count the sample until 1,000 counts have been obtained. Flush out the ^{222}Rn from the cell after obtaining the data. The detection limit is about 0.2 pCi/L for a 60-mL (milliliter) sample.

Calibration of Alpha-Scintillation Cell and Equipment

The counting efficiency of each alpha-scintillation cell varies between cells and between counting instruments. Consequently, counting efficiency of each alpha-scintillation cell was determined in each instrument in which it was used by counting ^{222}Rn transferred from a bubbler containing a measured volume of ^{226}Ra standard solution. A minimum of four or five bubblers containing the same standard enables four or five cells to be calibrated simultaneously. Use the following procedures. Pipet 10 mL of 10 pCi/mL ^{226}Ra standard solution directly into each of four to five bubbler tubes and dilute with distilled water to 60 percent full. De-emanate the bubbler to expel all ^{222}Rn; after 1 month, de-emanate the standard again to determine the amount of ^{222}Rn growth, and calculate the instrument and cell efficiency. Barring any spillage or breakage, the standards will last indefinitely and can be de-emanated to provide ^{222}Rn calibration. The counting efficiency of the system used at the U.S. Geological Survey laboratory was in the range of 4.9 to 5.6 counts per minute per 1 pCi of ^{222}Rn, with a background-count rate of 0.03 to 0.09 count per minute.

RESULTS AND DISCUSSION

The method is specific for ^{222}Rn; no direct interferences occur. Radon-220 and ^{219}Rn, if present, will have decayed completely by the time of the analysis because of their short half-lives, which are about 4 s for ^{219}Rn and 56 s for ^{220}Rn. Normally, these constituents will not be supported by any substantial concentration of the parent ^{224}Ra and ^{223}Ra in the sample; thus, they usually are insignificant.

Contamination of the alpha-scintillation cell can be a major problem. Ground water may contain large concentrations of dissolved ^{222}Rn, which, if de-emanated into the alpha-scintillation cells and allowed to remain in them for several hours, will result in considerably increased background count rates. Increased background count rates of more than several tenths of 1 count per minute preclude the reuse of cells for the analysis of most environmental

Table 39. Transportation and storage loss of radon-222

Sample type	Date			^{222}Rn^{1} (picocuries per liter)	^{226}Ra^{1} (picocuries per liter)
	Collected and shipped or stored	Returned	Analyzed		
Original, low activity	1–10–79	—	1–10–79	47.0±2.6	<1
Original, high activity	1–11–79	—	1–11–79	8,180±78	5.41±0.21
Transported, low activity	1–10–79	1–15–79	1–16–79	54.5±1.1	<1
Transported, high activity	1–11–79	1–17–79	1–18–79	7,850±77	5.41±0.21
Stored, low activity	1–10–79	—	1–16–79	52.0±1.1	<1
Stored, high activity	1–11–79	—	1–18–79	8,450±84	5.41±0.21

[1] Counting error is two standard deviations.

Table 40. Precision of radon-222 in water analysis

Sample^{1}	Number of runs	Measured mean concentration (picocuries per liter)	Relative deviation (percent)
1	2	3.12	19
2	3	48	5
3	2	105	4
4	3	756	3
5	3	4,600	2
6	3	8,160	1.5
7	2	24,690	1.0

[1] Samples were prepared by precipitating ^{226}Ra with barium sulfate twice, with minimal stirring, from ^{226}Ra stock solutions stored for 2 months after preparation. The clear solution was decanted as ^{222}Rn water samples.

samples. Therefore, ^{222}Rn in the cells needs to be counted and flushed out immediately after the data are obtained. The contaminated cells can be disassembled, cleaned, and refabricated by using fresh phosphor and a new quartz window if necessary.

Loss of Radon-222

Analytical results for loss of ^{222}Rn during transportation and storage for low- and high-activity samples are listed in table 39. The loss of ^{222}Rn during transportation and storage, after 1 week, is negligible for a low-activity sample (within three standard deviations of counting errors); the loss of ^{222}Rn during transportation and storage, for a high-activity sample after 1 week, indicates a loss of 4 percent during transportation and no loss during storage. The ^{226}Ra concentrations in both high- and low-activity samples also were measured and determined to be less than 1 pCi/L for a low-activity sample and about 5 pCi/L for a high-activity sample. Therefore, contribution of ^{222}Rn from a ^{226}Ra source can be neglected. The data presented here are limited; general conclusions are limited to the same degree.

Precision

Precision of the method may be expressed in terms of the relative deviation at different concentrations, as listed in table 40. The percentage deviation ranges from 19 percent for a sample concentration of less than 5 pCi/L to 1 percent for a 25,000-pCi/L sample concentration.

SUMMARY

Sample collection for determining the concentration of dissolved ^{222}Rn in surface water is performed best by placing one end of an evacuated bubbler into the water and then drawing a water sample directly into the main chamber of the bubbler until the bubbler is about 60 percent full. For collection of ground-water samples, a peristaltic or submersible pump is needed to supply an unaerated water sample through a short, flexible tubing connected to one end of the evacuated bubbler until the main chamber is about 60 percent full. Loss of ^{222}Rn during transportation from the sampling site to the laboratory (within 1 week) was negligible for low-activity samples and was 4 percent for high-activity samples. The direct de-emanation and alpha-scintillation counting method described in this report is sensitive (0.2 pCi/L for a 60-mL sample) and has a better precision than other methods for analysis of most environmental samples.

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A COMPARISON OF TWO TECHNIQUES FOR RADON-222 MEASUREMENT IN WATER SAMPLES

Ann Mullin and Richard B. Wanty

Abstract

As the level of concern over radon-222 in water supplies rises, it will become increasingly important to have established a set of reliable techniques for the measurement of radon-222 in water. Several different techniques are available currently, but for the most part these have not been compared against one another in the field. Rather, most validation techniques involve laboratory calibration against synthetic standards. Although interlaboratory studies are certainly necessary, a field intercomparison is equally important, as it tests all aspects of the measurement, including sample collection, storage, and analysis, as well as correction for the lag time between sampling and analysis. We report here on a field test of two different techniques: liquid scintillation and a technique in which the sample is degassed in the field and then measured in a Lucas cell.

The purpose of this field test was to compare the direct results from the two methods and to test the reproducibility or consistency of the two methods over a period of time after sampling. In the first part of the study, the results were in moderately good agreement, although large relative deviations existed between some samples. A comparison of the samples measured at various lag times between sampling and analysis showed very good consistency for the liquid scintillation method and poor consistency for the degassing Lucas cell method.

INTRODUCTION

Much emphasis is now being placed on radon-222 (^{222}Rn) both in air and water. One reason for this emphasis is the fact that the Environmental Protection Agency (EPA) is expected to issue a maximum contaminant level (MCL) for ^{222}Rn in drinking water soon. Unofficial guideline levels now being used range as high as 10,000 pCi/L (picocuries per liter) in water, but it is likely that the EPA will set the MCL at a lower level. The "action level" (level at which remedial action should be taken) for ^{222}Rn in air is 4 pCi/L.

For ^{222}Rn, most of the danger comes from inhalation, but it is well known that high levels of ^{222}Rn in a water supply can contribute to indoor airborne ^{222}Rn (Nazaroff and others, 1988; Hess and others, 1986, 1990; Lawrence and others, in press). Studies have shown that virtually all of the ^{222}Rn in the air of a home may come from the water supply (Lawrence and others, in press). In natural environments, ^{222}Rn concentrations have been measured as low as $<$10 pCi/L in surface water supplies to as high as 2×10^6 pCi/L in a private well in Maine (Aieta and others, 1987). Cothern (1987) stated "The average concentration of radon in these [U.S. public] water supplies generates a lifetime risk of about 1 in 10,000, the highest level of risk allowed for any contaminant currently regulated under the Safe Drinking Water Act."

Along with considering the health risks, the extent of occurrence, and methods and costs for removal of the contaminant, the EPA must take into consideration the methods and costs for detection of the contaminant before setting an MCL.

Several different analytical techniques are currently in use. The three most common are the bubbler Lucas cell (LC), liquid scintillation (LS), and degassing Lucas cell (DLC) methods. Each has advantages and drawbacks, in terms of accuracy, precision, sample throughput, and cost. The EPA conducted a study with 28 participating laboratories to compare the LC and LS methods (Whittaker and Akridge, 1986). This study was limited to a test of laboratory techniques and did not consider any variability that may arise as a result of sampling techniques in the field. The results of the study are summarized in table 41. Both methods, being considered valid and equivalent, have been approved by the EPA.

Thus far, little attention has been directed at comparing field sampling and analytical techniques. In this study, samples were collected from 17 private domestic wells and two springs in the Colorado Front Range and analyzed by the LS and DLC methods. In addition, a systematic study of the effects of the lag time between sample collection and analysis was performed. These results and a comparison of the two methods are presented in this paper.

METHODS

The LS technique is described in detail here. For a description of the DLC technique, see Reimer (this volume). The two techniques differ greatly in the sampling method as well as in the analytical approach. The LS technique requires a liquid sample that is agitated with a scintillation cocktail and therefore measures the radioactivity of the water sample directly. In the case of the DLC method, the sample is agitated in a closed vessel to extract the ^{222}Rn, and the radioactivity of the gas is measured.

In the LS method, energy from the radioactivity of the sample excites the fluor, which in turn gives off photons that can be measured by a photomultiplier tube. A mineral-oil-based cocktail is used that contains PPO (2,5-Diphenyloxazole) as the primary fluor and bis-MSB (p-bis-lo-Methylstyrylbenzene) as the secondary fluor (New England Nuclear Corp., oral commun., 1988).

Table 41. Comparison of liquid scintillation (LS) and Lucas cell (LC) methods by the EPA, with the cooperation of 28 privately owned laboratories (Whittaker and Akridge, 1986)

[All values are given in picocuries per liter; accuracy and precision comparisons are given at the 95-percent confidence level]

	Sample A	Sample B	Sample C
Known value	1,616±49	16,321±501	66,426±2,034
LS method	1,519±170	15,746±1,100	62,945±6,600
LC method	1,647±261	16,769±2,485	64,516±12,017

	LS Method	LC Method
Average accuracy	95.2±2.0%	100.7±4.6%
Average repeatability (within laboratory precision)	3.6±3.0%	6.4±3.8%
Average reproducibility (combined within and between laboratory precision)	10.2±4.2%	17.6±4.2%

Five mL (milliliters) of cocktail and 10 mL of sample are placed in a 20-mL scintillation vial and capped with a poly seal. Samples are taken in duplicate to check the sampling and counting precision as well as to give an average instead of a single result. Details of the sampling procedure are given in U.S. Geological Survey technical memorandum 88.02 from the Office of Water Quality (L.D. Cecil and I.C. Yang, written commun., 1988) as well as in EPA/EERF Manual 78–1. Samples are held for 3 h (hours) before counting takes place so the ^{222}Rn can equilibrate with its short-lived progeny (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po).

Radium-226, NBS-traceable, is used to prepare the standards. The EPA has shown in their studies (Horton, cited in Environmental Protection Agency, 1983) that ^{226}Ra remains in the aqueous layer and does not contribute significantly to the count rate. The only counts from ^{226}Ra will occur at the interface between the aqueous solution and the mineral-oil cocktail.

A series of standards ranging from 100 pCi/L to 200,000 pCi/L is prepared and held for 21 days to allow for ingrowth of ^{222}Rn. At this time, the ^{226}Ra and ^{222}Rn are considered to be in secular equilibrium. Blanks are prepared with distilled water and, along with the standards, are counted at intervals with the samples collected in the field. A blank or standard is counted after every tenth field sample. Counting time is 50 min (minutes) or more. The three alpha particles and two beta particles emitted from ^{222}Rn and its progeny are all counted. However, this effect is offset because the standards that are counted and used in the calculations all contain the same emitters.

The ^{222}Rn concentration in the sample is expressed in picocuries per liter by the following equation (Environmental Protection Agency, 1983):

$$^{222}\text{Rn}=\frac{\text{Net cpm of samples}\times 1{,}000}{\text{Conversion factor }\times\text{decay factor}\times 10}$$

where

Net cpm (counts per minute) of sample = gross cpm − background cpm; 1,000 converts milliliters to liters;

conversion factor in cpm/pCi = (net cpm of standard)/(^{222}Rn activity of standard);

decay factor = $e^{-\lambda t}$ where λ equals 7.54×10^{-3} h^{-1}, and t is the lag time between sample collection and analysis in hours; and

10 is the volume of sample in milliliters.

RESULTS AND DISCUSSION

For this comparison, 17 wells and 2 springs were sampled from crystalline aquifers along the Front Range west of Denver, Colo. Concentrations of ^{222}Rn ranged from 1,080 pCi/L to 45,000 pCi/L (table 42). The distribution of samples is shown in figure 89. As seen in the figure, most of the samples are less than 5,000 pCi/L; the average of all the samples was approximately 9,000 pCi/L. Figure 90

Table 42. The results of this study comparing the liquid scintillation (LS) and degassing Lucas cell (DLC) methods

[The column labeled "Average" contains the arithmetic average of the values obtained by the two methods; all values are given in picocuries per liter; the percent deviation is calculated from the formula ((DLC−LS)/DLC)×100]

Sample no.	DLC	LS	Average	Percent deviation
88CO–1	3,100	2,200	2,600	30
88CO–2	3,500	2,300	2,900	34
88CO–3	1,700	1,200	1,400	27
88CO–4	2,100	1,200	1,600	43
88CO–5	4,400	2,400	3,400	44
88CO–6	1,100	1,100	1,100	6.3
88CO–7	16,000	19,000	18,000	−16
88CO–8	20,000	16,000	18,000	21
88CO–9	43,000	47,000	45,000	−8.1
88CO–10	13,000	6,600	9,600	47
88CO–11	13,000	14,000	14,000	−6.3
88CO–12	12,000	10,000	11,000	20
88CO–13	3,400	2,300	2,900	32
88CO–14	2,200	2,300	2,200	−2.3
88CO–15	4,200	3,300	3,800	22
88CO–16	4,500	4,200	4,400	7.1
88CO–17	3,600	3,800	3,700	−3.6
88CO–18	19,000	20,000	20,000	−5.2
88CO–19	8,400	6,800	7,600	19

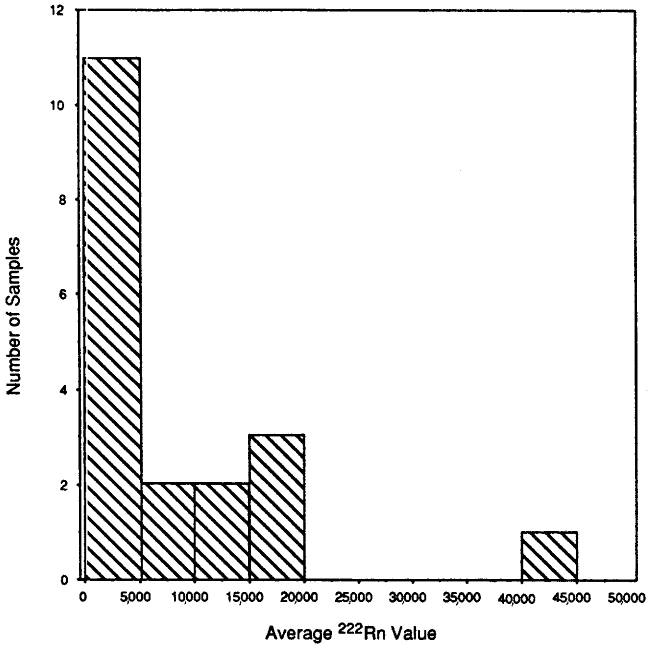

Figure 89. Distribution of radon-222 concentrations found in samples collected from 19 wells and springs in Colorado. The values shown are the average between the liquid scintillation (LS) and degassing Lucas cell (DLC) methods. Values are in picocuries per liter.

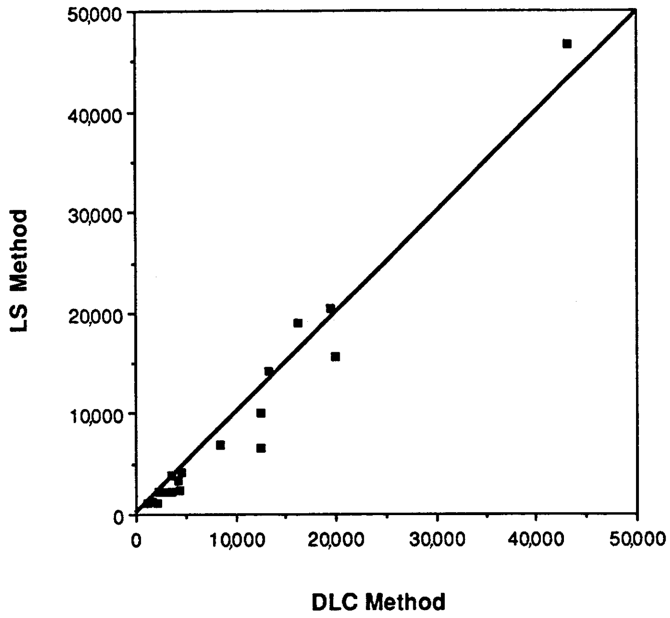

Figure 90. Comparison of analyses for radon-222 by the liquid scintillation (LS, vertical axis) and degassing Lucas cell (DLC, horizontal axis) methods. The line represents the ideal 1:1 correspondence. All values shown are in picocuries per liter.

shows a comparison of the results of the two methods. In some cases, the agreement is very good, but a few samples have significant relative deviations. In 13 of the 19 samples, the DLC method measured higher values than the LS method. For the LS method, all 19 samples were collected and analyzed in duplicate and, except for the first sample, showed very good precision. For the DLC method, duplicates were collected and promptly analyzed at 3 of the 19 sites.

The stability over time of the DLC method was tested by collecting several samples at each of two sites. These two sites were chosen because they represented a wide range of dissolved ^{222}Rn values. For each site, the analysis was repeated after the time intervals specified in table 43. The lag time between sampling and analysis was compensated for by using the exponential formula given earlier in this paper. The results are displayed graphically in figure 91. In both cases, increased lag time led to greater uncertainty and usually lower values, both of which indicate that variable amounts of loss of ^{222}Rn took place from the syringes in which the samples were stored.

The stability over time of the LS method also was tested but in a different way. Unlike the DLC method, in which the sample is lost once it is counted, the samples in the LS method can be recounted over a period of time, provided that adjustments are made for the lag time between sample collection and analysis. These results are shown in table 44 and figure 92. As is indicated, increased lag time from sampling to analysis for periods of up to 7 days shows no loss of ^{222}Rn. This appears to be true for samples that have ^{222}Rn concentrations as low as 1,000 pCi/L. For this study, counting was not extended beyond 7 days.

The LS method is well established, but the DLC method has been used less widely. The principal advantage of the DLC method is that results can be obtained in the field, at each site. Thus, the DLC technique provides for easy, rapid reconnaissance, although it has a larger relative error. The LS method, on the other hand, is probably more reliable but presents logistical problems because the instrumentation cannot be taken into the field.

When the two techniques are examined side by side, several differences are evident. A close look at these procedural differences may point out sources of relative error between the two techniques. In the DLC method, a known volume of water sample is degassed in the field in a closed vessel. The air in the head space of that vessel is collected in a syringe, where it is stored until measurement. Radon-222 in the air sample is measured after the sample is injected into a calibrated Lucas cell. In the LS method, a known volume of water sample is introduced to a scintillation vial that contains the scintillating cocktail. The vial is tightly sealed until it is counted. In either case, excessive degassing of the sample during collection would lead to an erroneously low value. In the DLC method, the syringes used are probably not tight enough to prevent the slow escape of ^{222}Rn, which would lead to a low value. In this

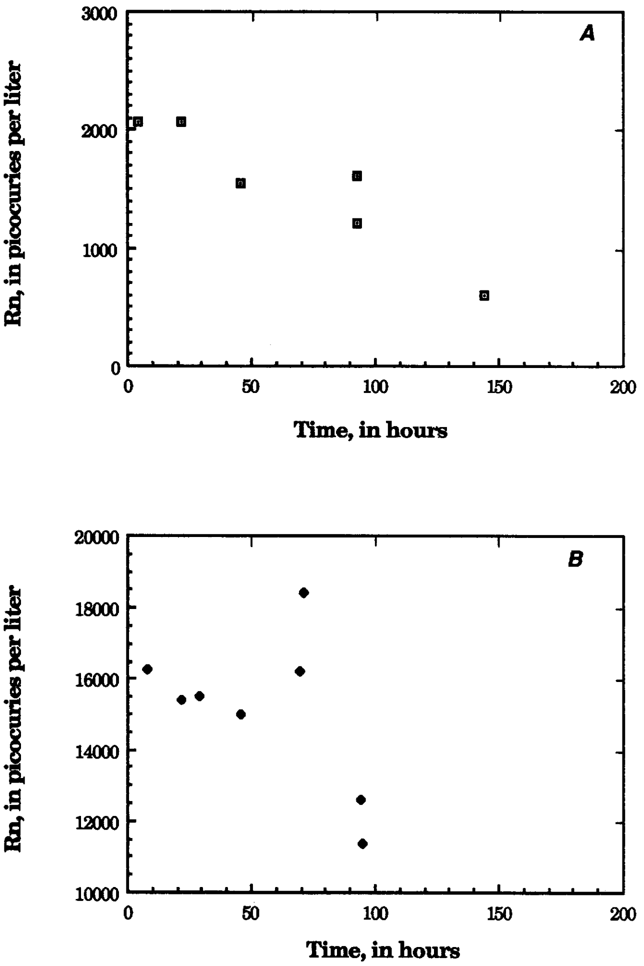

Figure 91. The effect of lag time on the measurement of radon-222 in replicate samples from sites 88CO–7 (*A*) and 88CO–4 (*B*), collected and analyzed by the degassing Lucas cell (DLC) method. The values shown are in picocuries per liter after being corrected for the lag time between sampling and analysis.

sense, the LS method provides for greater sample stability over time because of the fewer number of sample transfers between containers (two for DLC, one for LS). In both techniques, standards are used to calibrate the counting equipment so that a conversion factor from counts per minute to picocuries per liter can be obtained. Nevertheless, the DLC technique leaves more room for error in the calibration phase because the short-lived daughters of ^{222}Rn may not be adequately accounted for. If this is the case, erroneously high values may result from use of the DLC method.

CONCLUSIONS

This study provided a field intercomparison of two methods for measurement of ^{222}Rn in water samples. Although there are significant differences in the sample collection and analytical methods, moderately good agreement was obtained between the two methods when the DLC samples were analyzed after short lag times (<8–12 h).

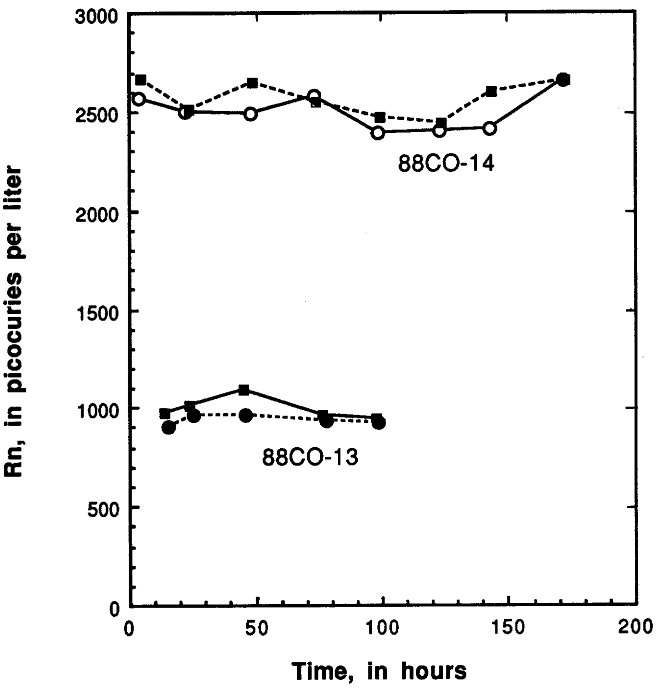

Figure 92. The effect of lag time on the measurement of radon-222 in replicate samples from sites 88CO–13 and 88CO–14, collected and analyzed by the liquid scintillation (LS) method. The values shown are in picocuries per liter after being corrected for the lag time between sampling and analysis.

Table 43. Effect of increasing lag time on the measurement of radon-222 in water samples by the degassing Lucas cell (DLC) method

[For each sample, the first measurement is taken to be the most reliable by this method]

Lag time (in hours)	^{222}Rn (in picocuries per liter)
Sample 88CO–4	
4	2,100
21.5	2,100
45.5	1,600
93	1,200
93	1,600
144	590
Sample 88CO–7	
8.3	16,000
21.7	15,000
29	16,000
45.8	15,000
69.8	16,000
71	18,000
94	13,000
94.8	11,000

Table 44. Effect of increasing lag time on the measurement of radon-222 in paired water samples by the liquid scintillation (LS) method

Lag time (in hours)	^{222}Rn (in picocuries per liter)
Sample 88CO-14	
3.5, 4.5	2,600, 2,700
22.5, 23.5	2,500, 2,500
47.75, 48.75	2,500, 2,600
73.0, 74.0	2,600, 2,600
98.25, 99.25	2,400, 2,500
123, 124.	2,400, 2,400
142.75, 143.75	2,400, 2,600
171.5, 172.5	2,700, 2,700
Sample 88CO-13	
13.75, 15.0	970, 900
24.0, 25.0	1,000, 1,000
45.0, 46.0	1,100, 960
76.5, 77.5	970, 930
97.5, 98.5	950, 930

Large relative deviations were encountered when the lag time for the DLC method was greater than 12 h.

The principle advantage of the DLC method is that results can be obtained in the field at each site. On the other hand, the DLC method is seen as a reconnaissance technique. If, for instance, the samples are not immediately analyzed in the field, problems develop. First, the syringes may not be tight enough to prevent the slow escape of ^{222}Rn from the gas sample. Second, the short-lived progeny of ^{222}Rn, specifically ^{218}Po, closely approaches secular equilibrium with ^{222}Rn in about 10 min. The activity of this daughter may not be accounted for adequately in the calibration scheme. Further, because the DLC analysis is performed at a time when the net alpha activity of ^{218}Po is rapidly building, large random errors may result.

Although the LS method does not give immediate onsite results, the results of this study indicate that the LS method is more accurate. First, there are fewer sample transfers between containers (two for DLC, one for LS), and the vials are more tightly sealed between collection and analysis. Second, the standards used in the calibration phase of the LS method contain ^{222}Rn in equilibrium with its progeny; the same is true for the samples. Thus, the activity of these daughters is adequately accounted for.

Experiments in which large lag times between sample collection and analysis were used showed that the LS method is much more stable than the DLC method. This observation further supports the conclusion that the syringes used to hold the samples in the DLC method are not completely impervious to ^{222}Rn escape. The most effective use of the two techniques is as follows: the DLC method is useful for reconnaissance sampling, where interim results may help to design a sampling grid, but the LS method is preferred for providing more reliable results.

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PART 2: STUDIES OF RADON IN WATER

SECTION 3: FIELD STUDIES OF RADON IN SURFACE AND GROUND WATERS

USE OF RADON MEASUREMENTS IN CARTERS CREEK, MAURY COUNTY, TENNESSEE, TO DETERMINE LOCATION AND MAGNITUDE OF GROUND-WATER SEEPAGE

Roger W. Lee and Este F. Hollyday

Abstract

The location and magnitude of ground-water seepage to streams can be determined by measuring the activity of radon-222 in streamflow. Radon activity in ground water may be 2 to 4 orders of magnitude greater than in surface water. Thus, ground-water seepage to a stream usually results in increases in localized radon-222 activity in the streamflow. Downstream of ground-water seepage, radon-222 activity decreases in the stream as radon escapes to the atmosphere, particularly in turbulent reaches of the stream. The relation between ground-water and surface-water flows can be determined by mass balance if it is assumed that no other radon-222 sources exist and no significant gas loss from the mix of surface water and ground water takes place at the sampling point.

Measurements of radon-222 activity in water from a 0.75-mile reach of Carters Creek, a small bedrock-channel stream in Maury County, Tenn., ranged from 32 to 196 disintegrations per minute per liter. A sample of ground water from an adjacent spring contained radon-222 activity of 489 disintegrations per minute per liter. On the basis of radon-222 activities down the sampled reach of the stream, during low base-flow conditions 36 percent of flow leaving the reach was ground-water seepage at a point 0.5 mile downstream from the upper sampling boundary. Measurements of temperature in the water of the stream verified the point location of ground-water seepage. Subsequent measurements of radon-222 activities and streamflow in Carters Creek at higher base-flow conditions indicated two gaining sections of stream, which were receiving about 0.3 cubic foot per second ground-water seepage in the upper reach and about 0.2 cubic foot per second ground-water seepage in the lower reach.

INTRODUCTION

Measurements of radon-222 (^{222}Rn) activity in water from stream-aquifer hydrologic systems have been successfully used to characterize ground-water seepage to rivers and streams (Rogers, 1958; Jacoby and others, 1979). If measurements of streamflow are combined with measurements of ^{222}Rn activities in surface water and ground water, quantitative analysis of ground-water seepage to a stream is possible. Ellins and others (1985, 1990) have shown the applicability of this approach to stream-carbonate aquifer interactions in Jamaica and Puerto Rico.

This report describes the results of a study to apply the method in the reconnaissance of small gaining streams in the Ordovician limestone areas of middle Tennessee. Determination of points of discharge and quantity of ground-water seepage could be useful for locating potential ground-water supplies as well as for quantifying base-flow components of streams. Two surface-water flow measurements and 11 measurements of ^{222}Rn activities were taken in Carters Creek, Maury County, Tenn., during periods of base flow in the summer of 1986 (fig. 93). Additionally, six surface-water flow measurements and six ^{222}Rn activities were measured in Carters Creek during a higher low-flow condition in February 1987 (fig. 94).

RADON CYCLING IN GROUND-WATER AND SURFACE-WATER SYSTEMS

Radon-222 occurs naturally in the Earth's crust as a daughter of the radioactive decay of radium-226 (^{226}Ra). The concentration of ^{222}Rn in ground water is primarily related to the amount of ^{226}Ra present in the aquifer matrix, especially near grain boundaries or in coatings on grains or fractures, although hydrology and structural geology may exert some influences. Activities may range from 100 dpm/L (disintegrations per minute per liter), typical of water in clastic sedimentary rocks, to greater than 10,000 dpm/L, typical of water in igneous or metamorphic rocks. The activity of ^{222}Rn in the atmosphere is almost nil. Thus, as ground water containing ^{222}Rn seeps into surface-water bodies, ^{222}Rn escapes to the atmosphere across the air-water interface, and the concentration of ^{222}Rn in surface water decreases rapidly. Activities of ^{222}Rn in water from an aquifer may be 2 to 4 orders of magnitude greater than activities in an adjacent surface-water body (King and others, 1982). Radon-222 is therefore suitable for determining the ground-water discharge to surface-water systems.

Several factors that control ^{222}Rn activity in water should be considered for this determination:

1. The difference in activity of ^{222}Rn between ground water and adjacent stream water.

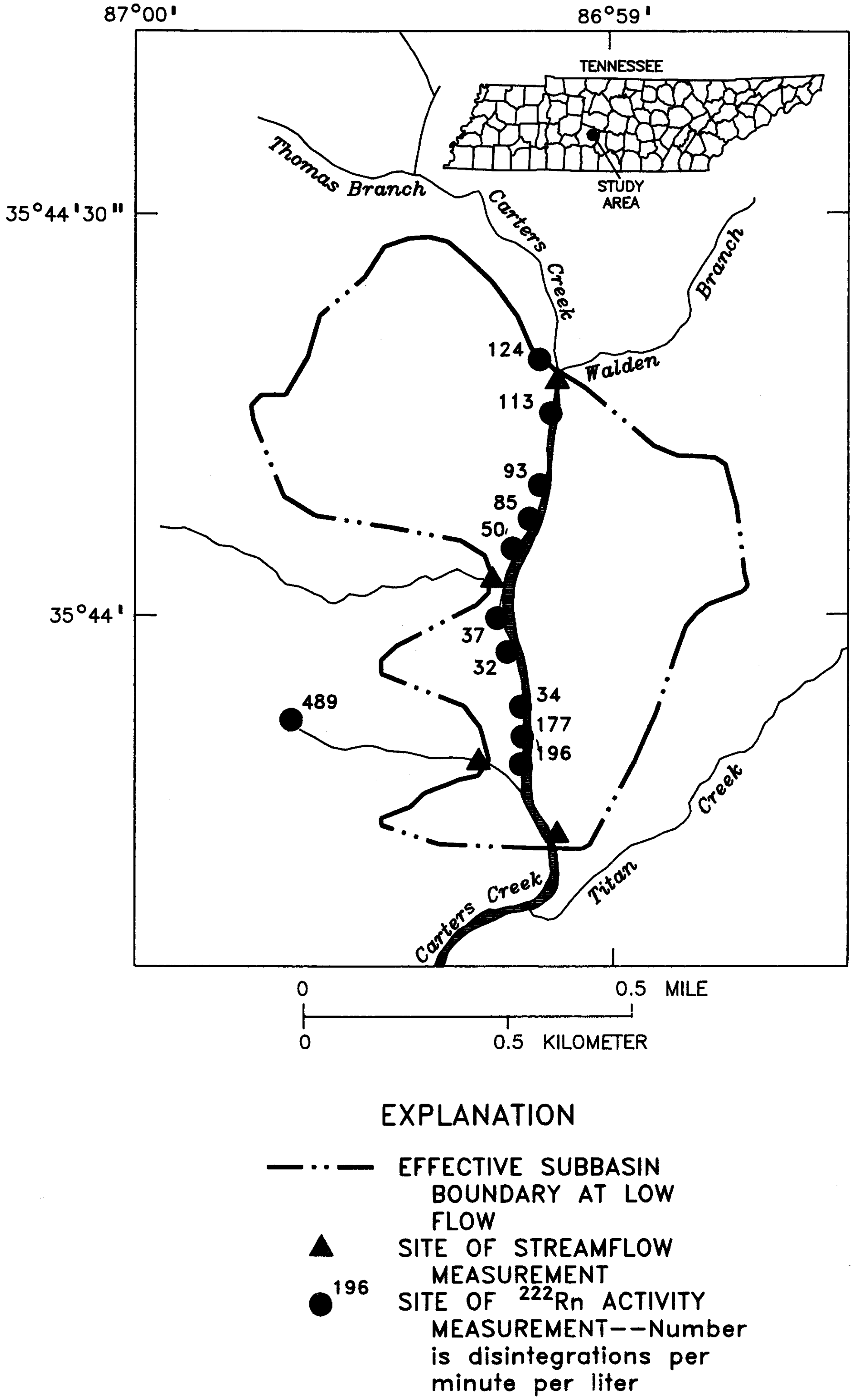

Figure 93. Locations of measurements of surface-water discharge and radon-222 activities for September 16–17, 1986, in Carters Creek.

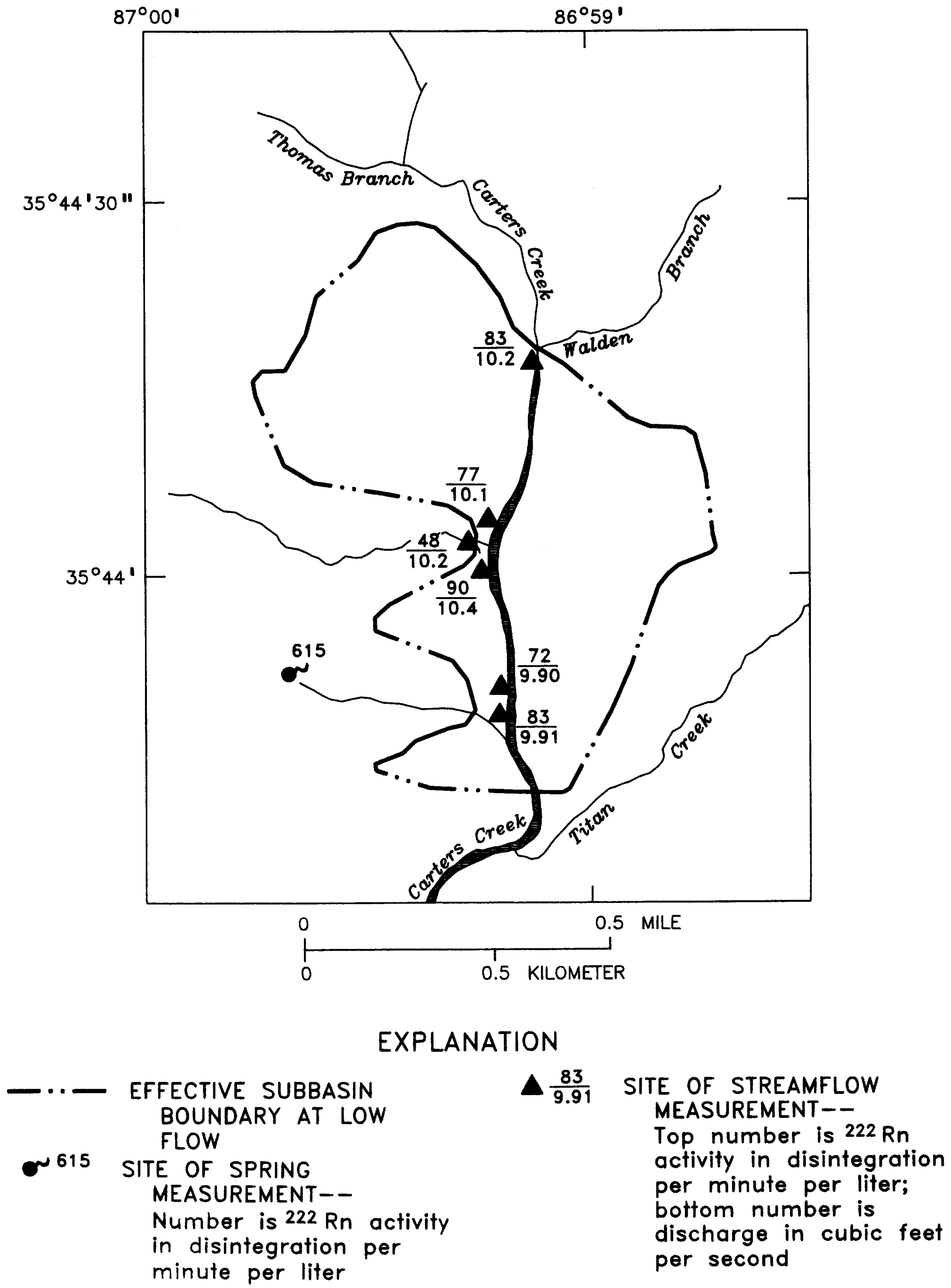

Figure 94. Locations of measurements of surface-water discharge and radon-222 activities for February 12, 1987, in Carters Creek.

2. The magnitude of a ground-water seep in relation to total flow along a reach of stream.
3. The rate of gas loss in the stream (gas-exchange rate) based on temperature, diffusion, turbulence, and surface area of air-water interface relative to stream volume (Elsinger and Moore, 1983).
4. The length of the stream reach measured and the number of ground-water discharge points.
5. The contribution of flow and ^{222}Rn from tributaries to the main channel.
6. The characteristics of ground-water seepage (discrete or dispersed discharge). Factors 1 and 2 are most significant in determining suitability of this method to ground-water and surface-water systems. Factors 3 through 6 are determined subsequently by application of the method to the specific system.

CHARACTERIZATION OF BASE FLOW

Ground-water seepage may occur as a dispersed discharge into the stream channel along a given length of channel. This type of seepage is typical of channels flowing on granular material. Seepage also may be discrete, which means it contributes flow at well-defined points along the stream. Discrete seepage is typical of channels that flow on soluble bedrock, fractured bedrock, or faulted and fractured rock.

The relation between ground-water and surface-water flows for a reach of stream as determined by use of ^{222}Rn activities is defined by the mass balance mixing equation

$$Q_s \times A_s + Q_{gw} \times A_{gw} = Q_m \times A_m, \tag{1}$$

where

Q_s=the rate of streamflow, in cubic feet per second;

A_s=the activity of ^{222}Rn in streamflow at upstream point, in disintegrations per minute per liter;

Q_{gw}=the rate of ground-water seepage, in cubic feet per second;

A_{gw}=the activity of ^{222}Rn in ground water, in disintegrations per minute per liter;

Q_m=the rate of mixed streamflow and ground-water flow downstream, in cubic feet per second;

A_m=the activity of ^{222}Rn in mixed water downstream, in disintegrations per minute per liter; and

$$Q_s + Q_{gw} = Q_m. \tag{2}$$

Equation 1 assumes no other source of ^{222}Rn other than that conveyed to the stream from ground water and also assumes no significant loss of ^{222}Rn from the mix of surface and ground water at the sampling point. Thus, measurements of streamflow and ^{222}Rn activity in the stream and in ground-water sources allow calculation of the magnitude of ground-water seepage.

In the absence of streamflow measurements, a proportional value of ground-water seepage may be determined. Substitution of equation 2 into equation 1 for Q_s yields

$$\frac{Q_{gw}}{Q_m} = \frac{A_m - A_s}{A_{gw} - A_s}. \tag{3}$$

This form of the equation permits use of ^{222}Rn activities to determine the ratio of ground-water seepage to the total streamflow at the measured point. Equation 3 is subject to the same limitations discussed for equation 1.

APPLICATION TO CARTERS CREEK, TENNESSEE

Carters Creek is a small stream that has a low base flow of about 1.0 ft^3/s (cubic foot per second) and an average annual flow of about 17 ft^3/s. The stream is incised through regolith, and much of the streambed is on thin-bedded, clay-rich limestone of Ordovician age. At low base-flow conditions, the streamflow is characterized by long, shallow pools and short riffles. The stream channel of the sampled reach is relatively straight, and, during higher flows (10 ft^3/s or greater), the riffles become submerged and flow is more uniform.

A base-flow survey conducted June 26, 1986, in Maury County, Tenn., indicated that a substantial increase in flow in the uppermost reach of Carters Creek was from ground-water seepage (Lowery and others, 1987). Subsequently, a reconnaissance sampling of ^{222}Rn activities along this reach of Carters Creek under low base-flow conditions (1 ft^3/s) was conducted on September 16, 1986 (fig. 93). No significant surface-water inflow from tributaries was observed along the sampled reach of stream.

A second base-flow survey of ^{222}Rn activities was conducted on February 12, 1987, during high base-flow conditions (10 ft^3/s) in order to compare the two base-flow extremes. Streamflow measurements also were made, along with the ^{222}Rn measurements, to better quantify ground-water discharges under the prevailing hydrologic conditions.

METHODS

Water samples of approximately 200 mL (milliliters) were collected from surface-water sites in Carters Creek and one adjacent spring (fig. 93). The samples were drawn into evacuated samplers where radon gas was vacuum stripped from the water into calibrated Lucas cells. The activity of ^{222}Rn was determined by alpha spectrometry by use of commercially available equipment. Sufficient length of counting period and repetition of counting events produced an analytical error of less than 3 percent for all samples. Samples were collected in midstream, about 6 to 12 in (inches) below the water surface, where there would be

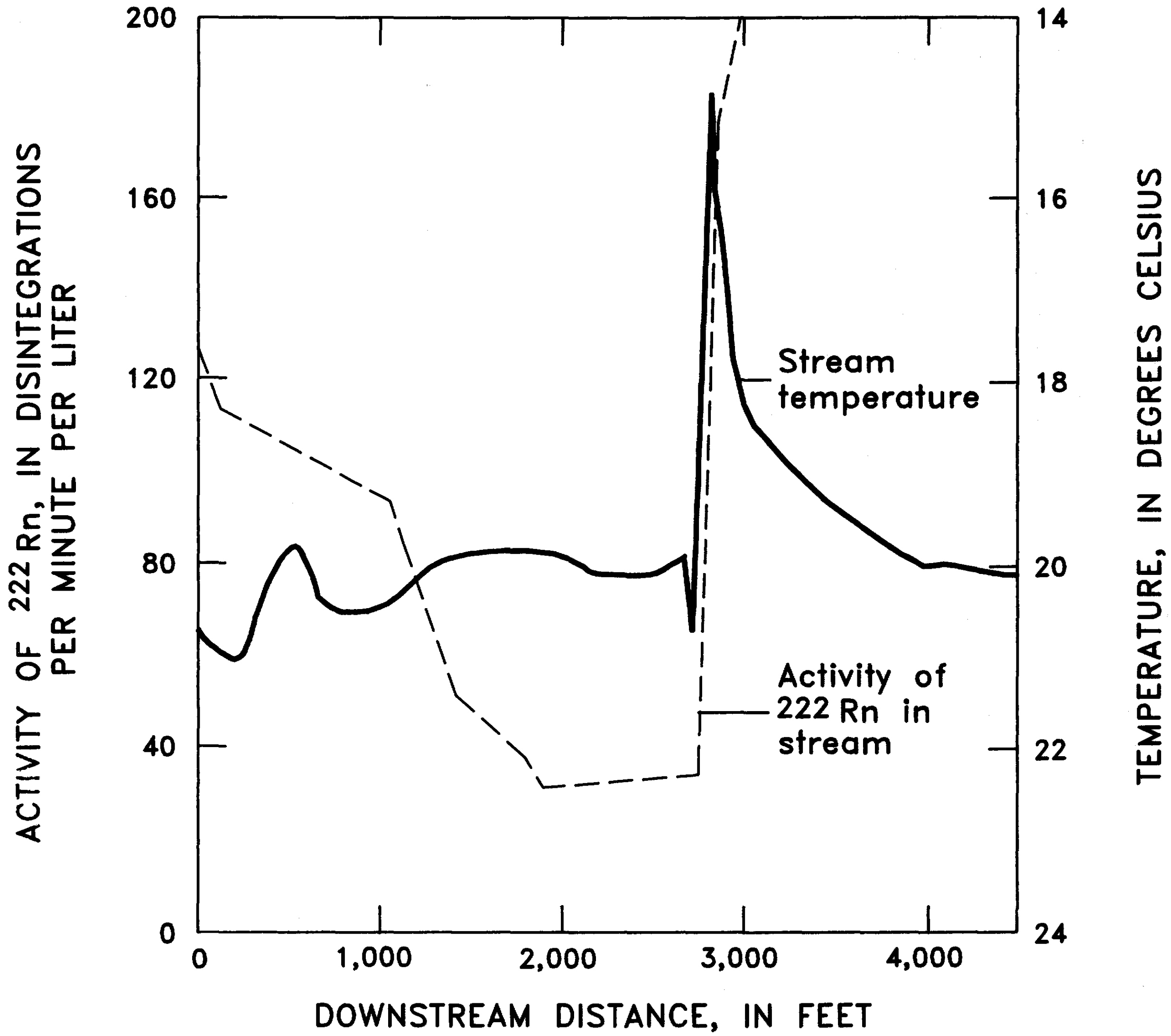

Figure 95. Stream temperatures and radon-222 activities in a 0.75-mile reach of Carters Creek for September 16–17, 1986.

optimum mixing of ground and surface waters. Collection points were usually just upstream of riffles, at the end of a pool or flow channel, and just downstream of riffles, at the head of a pool or flow channel. Occasionally, samples were collected in the middle of a pool to determine intermediate ^{222}Rn activities in the pool.

RESULTS AND DISCUSSION

In the initial sampling of Carters Creek in September 1986, the activities of ^{222}Rn ranged from 32 to 196 dpm/L (fig. 93). An adjacent spring produced water that contained 489 dpm/L, or, for the only reach of stream increasing in ^{222}Rn activity,

$$A_s=34 \text{ dpm/L},$$
$$A_{gw}=489 \text{ dpm/L},$$
$$A_m=196 \text{ dpm/L}.$$

Substitution into equation 3 yields

$$\frac{Q_{gw}}{Q_m}=0.36,$$

meaning that about 36 percent of the total flow downstream of the lowermost pool entered the stream at that pool. Stream temperatures and an intermediate ^{222}Rn measurement were used to verify the seepage of ground water into the pool (fig. 95). Ground-water discharge to the pool was estimated to be about 0.6 ft ^{3}/s (Lee and Hollyday, 1987). The second sampling of Carters Creek, on February 12, 1987 (fig. 94), produced lower ^{222}Rn activities and a narrow range of values (72 to 90 dpm/L in the main channel). Streamflow measurements showed only one possible gaining section (0.3 ft^3/s), which was in the middle of the reach of stream sampled. Radon activities from this sampling indicate two middle-reach gains from ground-water seepage and some seepage of ground water into the same lower pool that had seepage during the September 1986 sampling. A second measurement of the adjacent spring indicated water with a ^{222}Rn activity of 615 dpm/L, compared to the previous value of 489 dpm/L. This variance cannot be explained at present; 615 dpm/L will be used for A_{gw} for the computations to follow.

Solution of equation 3 for the middle reach shows that about 2.4 percent of the streamflow was from ground-water seepage along the middle reach. For Q_m of 10.4 ft^3/s,

$$Q_{gw}=0.252 \text{ ft}^3/\text{s}.$$

This value is very close to the 0.3 ft^3/s gain shown by streamflow measurements.

Further refinement can be obtained by considering the input of tributary flow and its ^{222}Rn activity in a modification of equation 1 to

$$Q_s \times A_s + Q_{gw} \times A_{gw} + Q_t \times A_t = Q_m \times A_m \qquad (4)$$

where

Q_t = the rate of tributary flow, in cubic feet per second, and

A_t = the activity of ^{222}Rn in tributary above confluence, in disintegrations per minute per liter.

In solving for Q_{gw},

$$Q_{gw} = 0.256 \text{ ft}^3/\text{s},$$

or a difference of 0.004 ft^3/s, which is a negligible quantity, considering the errors of the streamflow measurements and the radon-measuring procedures as applied here.

However, at the lower reach, ^{222}Rn activity increased from 72 to 83 dpm/L, and this increase indicates ground-water seepage to the stream, although no significant gain in flow is apparent (9.90 to 9.91 ft^3/s). From equation 3, 2.07 percent of total streamflow is gained in this reach, or about 0.2 ft^3/s. This figure indicates that streamflow measurements are not sensitive enough to detect this small inflow or that streamflow also is lost to an aquifer as ground water is gained in the stream.

These results are based on the premise that all data used are without error, although streamflow measurements may contain up to 5 percent error. Data collected on both ^{222}Rn and streamflow can be refined statistically and with more sophisticated measuring techniques. The use of three significant figures for ^{222}Rn activities measured in this investigation may be somewhat questionable. However, these data do demonstrate variabilities of ^{222}Rn activities in the stream and how measurements of streamflow and ^{222}Rn activity can be used to locate and quantify points of ground-water seepage to streams.

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GEOLOGIC AND GEOCHEMICAL FACTORS CONTROLLING URANIUM, RADIUM-226, AND RADON-222 IN GROUND WATER, NEWARK BASIN, NEW JERSEY

Zoltan Szabo and Otto S. Zapecza

Prepared in cooperation with the New Jersey Department of Environmental Protection

Abstract

Elevated concentrations of naturally occurring radionuclides are found in ground-water samples collected from the Newark basin in central New Jersey and are associated with uranium enrichment in the bedrock formations. Elevated radionuclide concentrations are present primarily in the lower part of the Passaic Formation but also are common in the underlying Lockatong Formation and, locally, in the Stockton Formation. These formations are exposed along the southern and eastern margins of the Newark basin and in the Flemington and Hopewell fault blocks, where the formations are stratigraphically repeated. Other formations in the Newark basin contain significantly lower concentrations of radionuclides.

Borehole geophysical testing, analysis of lithologic cores, and autoradiographs were used to determine that the source of the natural radioactivity in the lower part of the Passaic Formation and the upper part of the Lockatong Formation is thin, laterally extensive, uranium-enriched black mudstones that locally contain carbonate-rich laminae. Radioactive zones are found dispersed along horizontal bedding planes throughout the matrix and in arcuate bands that crosscut bedding. Autoradiographs from the Stockton Formation show that the radioactive sources are dispersed but are not conclusive as to whether the source of the radioactivity is in the sandstone matrix or in the fracture-fill materials.

Results of radiochemical analyses of ground-water samples show that gross alpha-particle activities range from less than 1 to 124 picocuries per liter, uranium concentrations range from less than 0.4 to 60 micrograms per liter, radium-226 concentrations range from less than 0.6 to 22.5 picocuries per liter, radon-222 concentrations range from 71 to 15,900 picocuries per liter, and gross beta-particle activities range from less than 1 to 49.7 picocuries per liter.

The local ground-water chemistry determines the distribution of dissolved uranium and radium. Elevated uranium concentrations are associated with high gross alpha-particle activity in oxidizing ground water that is low in iron and manganese and commonly high in sulfate and bicarbonate. In contrast, elevated concentrations of radium-226 are associated with high gross alpha-particle activity in reducing ground water that is high in iron, manganese, and sometimes barium and low in sulfate. High concentrations of uranium and radium typically are not found concurrently in the same sample. Radon-222 concentrations are weakly correlated with radium-226 concentrations but not with uranium concentrations or the concentrations of any other dissolved chemical constituent.

Chemical processes that may govern the distribution of the radionuclides in the ground water of the Newark basin are postulated on the basis of the chemical composition of the ground water and published laboratory experiments. Uranium concentrations are controlled by the solubility of the U^{+6} ion and by carbonate complexing. Radium-226 concentrations are restricted by adsorption onto iron and manganese hydroxides, adsorption onto or ion-exchange with secondary clay minerals, and coprecipitation with sulfate minerals. These reactions, in turn, are limited by the Eh (oxidation-reduction potential) and the pH of the ground water. Radon-222 concentrations are governed by the abundance and distribution of radium-226 in the rock matrix, the emanation efficiency of radon-222 from the rock matrix, and the physical characteristics that determine ground-water flow in the rock.

INTRODUCTION

In 1984, the U.S. Geological Survey, in cooperation with the New Jersey Department of Environmental Protection (NJDEP), Division of Water Resources, began a study of naturally occurring radionuclides in ground water in the Newark basin, New Jersey. The principal objectives of the study are to (1) determine the occurrence and distribution of naturally occurring radionuclides in ground water and identify areas, aquifers, or parts of aquifers where elevated radioactivity may be expected; (2) identify nonradioactive constituents that might serve as indicators of radionuclides in ground water; and (3) define geological and geochemical factors that control radionuclide distribution and concentrations in ground water. The Newark basin (fig. 96) was chosen for this study because of previous evidence of uranium mineralization, known radioactive ground-water anomalies, locally high surficial radioactivity as mapped by aeroradioactivity studies, and zones of high gamma activity in the subsurface as recorded by borehole geophysical logs.

Purpose and Scope

This paper presents significant findings on the geological and geochemical factors controlling the distribution of naturally occurring radionuclides in ground water of the Newark basin, New Jersey. Two hundred and sixty ground-water samples were collected from the study area and analyzed for gross alpha- and beta-particle activity. Alkalinity, dissolved oxygen, Eh (oxidation-reduction potential), pH, specific conductance, and water temperature were measured on site. Selected samples also were analyzed for uranium, radium-226 (^{226}Ra), radon-222 (^{222}Rn), major ions, and trace metals. In conjunction with the ground-water sampling, borehole geophysics and rock coring were used to identify specific zones of radioactivity in subsurface rocks.

Study Area

The Newark basin in New Jersey (fig. 96) coincides with the Piedmont physiographic province and comprises a 10-county area of approximately 2,000 mi^2. The basin is exposed at land surface in a band 16 to 32 mi wide that trends southwestward from the New York-New Jersey border to the Delaware River. On the northwest, it is bordered by the Border Fault, which separates it from the Precambrian and Paleozoic crystalline and sedimentary rocks of the New Jersey Highlands, also known as the Reading Prong. On the southeast it is bordered by the Fall Line, the unconformable contact with the overlying unconsolidated deposits of the Atlantic Coastal Plain. The Newark basin is a highly populated area in the northeastern corridor of the United States where significant quantities of ground-water are withdrawn from fractured-rock aquifers. In the New Jersey part of the Newark basin, an estimated 143 million gallons of ground water per day is pumped, 78 percent of which is used for public supply (U.S. Geological Survey, 1990).

Geologic Setting

The Newark basin is one of many fault-block basins in the Eastern United States that were formed in Triassic and Jurassic time during the initial rifting of the continents (Manspeizer and others, 1978). These basins were filled with thick sequences of continental sediments along with interbedded lava flows and intrusives. Newark basin formations are estimated to attain combined maximum thicknesses of 16,000 to 20,000 ft in New Jersey (Van Houten, 1969). The source of sedimentation in the basin was erosion of Precambrian and Paleozoic highlands that surrounded the basin. Sediment dispersal was from all directions during basin filling (Abdel-Monem and Kulp, 1968); however, the principal source of sediment was the crystalline-rock highlands to the southeast (Van Houten, 1969).

A generalized geologic map of the New Jersey part of the Newark basin is shown in figure 97. Progressively younger rocks are exposed toward the northwest because of the northwestward-tilting structure of the Newark basin. In the southwestern part of the study area, postdepositional faulting has caused part of the stratigraphic sequence to be repeated in outcrops northwest of the faults in the Hopewell and Flemington fault blocks (fig. 97).

The three major sedimentary units in the Newark basin are the Stockton Formation, the Lockatong Formation, and the Passaic Formation of Olsen (1980a). In figure 97, the Jurassic sedimentary units that are interbedded with, and overlie, the basalt flows in the northwestern part of the basin have been termed "undifferentiated rocks." The Passaic Formation and the undifferentiated rock units were formerly called the Brunswick Formation.

The Triassic Stockton Formation crops out along the southeastern part of the study area and in the Hopewell and Flemington fault blocks. Its dominant lithology is gray- and buff-colored arkose and arkosic conglomerate, red arkosic sandstone, and red and gray siltstone. The Stockton Formation, which is the oldest and most widespread unit in the subsurface, unconformably overlies Precambrian and Paleozoic basement rocks. Along the southeastern edge of the study area, the coarse fraction of the formation was deposited by steep-gradient streams (Olsen, 1980b). Conglomerate and coarse sandstone generally grade into finer grained sandstone and siltstone toward the center of the basin and may represent a facies change to a marginal lacustrine deposit (Turner-Peterson, 1977).

The Triassic Lockatong Formation conformably overlies and interfingers with the Stockton Formation over most of the Newark basin. The Lockatong Formation is composed primarily of alternating sedimentary cycles of gray, green, and black mudstones, siltstones, and minor carbonates that reflect the expansion and waning of an extensive lake, often referred to as Lockatong Lake (Van Houten, 1969). In the northeastern end of the basin, where the unit is thinnest, the Lockatong Formation consists of darker colored mudstones that interfinger with beds of buff arkosic sandstone that are virtually indistinguishable from the sediments of the Stockton Formation (Olsen, 1980b). Wider fluctuations of the Lockatong Lake are noted in the upper part of the Lockatong Formation and continue upsection into the lower part of the Passaic Formation. In this part of the stratigraphic section, alternating deposits of nearshore red mudstones with offshore gray and black mudstones indicate extensive interfingering of both oxidizing and reducing environments. The interfingering of these oxidizing and reducing environments plays an important role in the formation of uranium-enriched zones.

The boundary between the Passaic Formation and the Lockatong Formation is so gradual that it is defined both horizontally and vertically on the proportion of red mudstone beds to gray and black mudstone beds (Olsen, 1980b). The lower part of the Passaic Formation contains many black mudstone beds interbedded with the predominantly red mudstone beds. Overlying this transitional zone, the upper part of the Passaic Formation is composed predominantly of reddish-brown to maroon mudstone, siltstone, and sandstone. It is the most widespread unit cropping out in the basin. The depositional environment of the Passaic Forma-

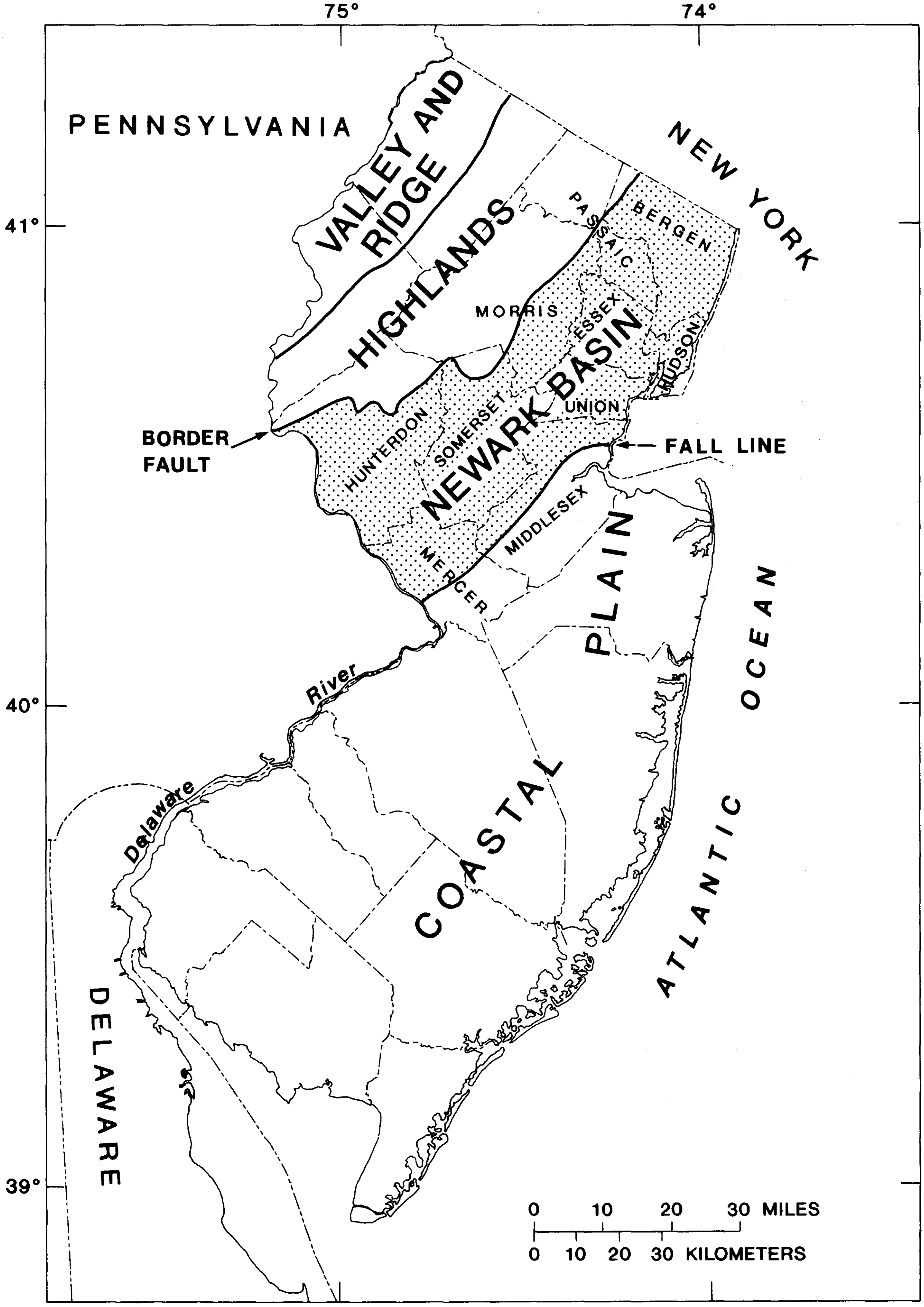


Figure 96. Location of study area (stippled), Newark basin, New Jersey.

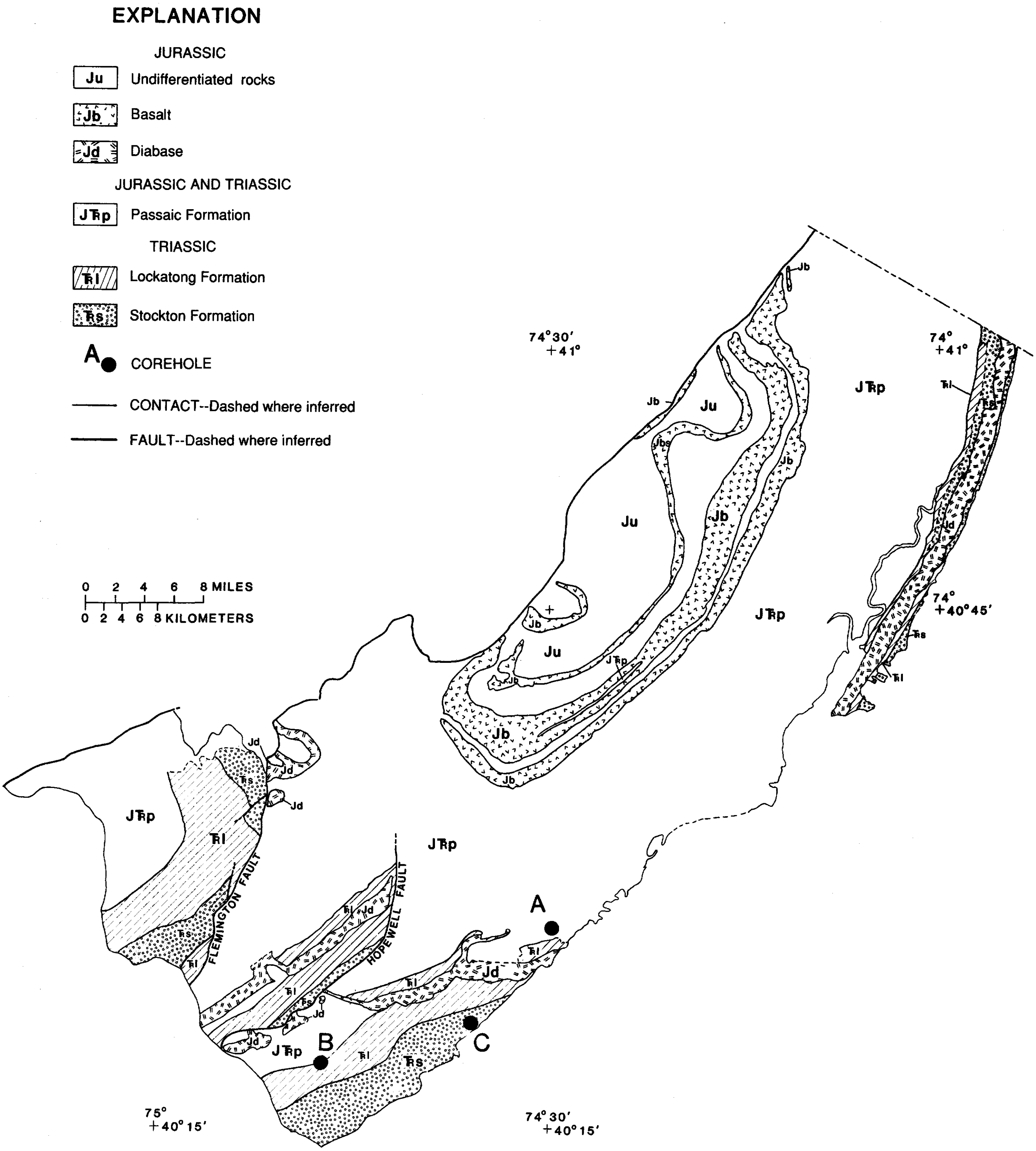


Figure 97. Generalized geologic map of the Newark basin, New Jersey, and location of coreholes. Scale 1:546,456. Geologic map modified from Lyttle and Epstein (1987).

tion was originally lacustrine; subsequently, it gave way to broad, weakly drained mud flats and playas (Van Houten, 1969).

During Early Jurassic time, continued crustal extension was accompanied by emplacement of diabase intrusives along the eastern and southwestern part of the study area, and basaltic lava flows were extruded over the northwestern part (Manspeizer, 1980). Jurassic sedimentary deposits that are interbedded with and overlie the basalt flows record a return to cyclic lacustrine-nearshore deposits. Although these deposits are shown in figure 97 as being undifferentiated, Olsen (1980a,b) has divided these deposits into formal units based on lithology and biostratigraphy.

SOURCES OF URANIUM

The crystalline rocks that flank the Newark basin are rich in uranium and are the source of sediments delivered to the basin during Triassic and Jurassic time. According to Grauch (1976), the crystalline rocks adjacent to the Newark basin in the New Jersey Highlands Province (Reading Prong) and in eastern Pennsylvania form one of four uranium-rich provinces of crystalline rocks in the Eastern United States. Uranium from these source rocks was delivered along with clastic sediments to the Newark basin by sediment-laden streams and was redistributed by circulating ground water (Turner-Peterson, 1980).

Numerous uranium-enriched zones have been reported in formations of the Newark basin (McKeown and others, 1954; Turner-Peterson, 1980; Popper and Martin, 1982; Bell, 1983; Turner-Peterson and others, 1985). Most of the reported uranium-enriched zones are in outcrops of the Stockton and Lockatong Formations in the Flemington fault block. Turner-Peterson (1980) states that concentrations of 0.01 to 0.02 percent uranium oxide (reported as U_3O_8) are the usual grade for mineralized mudstones of the Lockatong Formation. Uranium concentrations as high as 1.28 percent uranium oxide (reported as U_3O_8) have been measured for the Stockton Formation (Turner-Peterson, 1977).

Turner-Peterson (1977, 1980) has provided a lacustrine-humate model for uranium mineralization in the Newark basin for the offshore lacustrine black mudstone of the Lockatong Formation and the marginal lacustrine sandstone of the Stockton Formation. According to the model, uranium mineralization in the Lockatong Formation occurred during deposition. Humic matter and aqueous sulfide, capable of fixing uranium by reducing it from the U^{+6} to the U^{+4} valence state, were forming in offshore black muds at the time of deposition. Bacterial anaerobic respiration reduced sulfate from the lake water to bisulfide within the bottom sediments. Uranium, dissolved in river water entering the lake, precipitates from solution in reducing environments and therefore was fixed near the sediment-water interface.

Mineralization in the sandstone beds of the Stockton Formation occurred shortly after deposition. Reducing agents (humic acids and aqueous sulfide) generated from the black lake-bottom muds are believed to have been expelled during compaction into adjacent marginal lacustrine sands. Solubilization of humic acids and aqueous sulfide was favored in the pore fluids of the muds because the environment was alkaline. Uranium-bearing ground water passed through the sands as it moved upward into the lake, and the reductants from the mud caused precipitation of uranium. In addition to uranium mineralization during deposition, or shortly thereafter, it is possible that some zones of uranium enrichment formed more recently along secondarily permeable zones during migration of intrastratal solutions.

SAMPLING AND ANALYSIS

Well Construction

Water samples were collected by the U.S. Geological Survey (USGS) from 260 wells distributed throughout the Newark basin (fig. 98). Information about well construction for each site was obtained from the files of the USGS and the NJDEP.

Wells finished in the rocks of the Newark basin usually are cased from the land surface, through the weathered regolith zone, into unweathered rock below. The remainder of the well is left uncased, commonly open to several hundred feet of fresh rock, so water from the fractures intersected by the borehole is allowed to drain freely into the well. Individual fracture zones within the same well may yield water of somewhat different chemical compositions (Edmunds and others, 1987). Therefore, these open-hole wells can be visualized as penetrating a thick aquifer in which water quality is vertically nonhomogeneous. Water from these wells has a gross chemistry that is a composite of the different water chemistries contributed to the well by the individual fracture zones.

Sampling Methods

All samples consisted of raw (untreated) water collected from a tap near the well head or at the base of a pressure tank or small holding tank that was drained before sampling. A minimum of one well-casing volume of water was pumped from the well, and three well-casing volumes of water were removed whenever possible (Claassen, 1982). Alkalinity, dissolved oxygen, Eh, pH, specific conductance, and water temperature were measured on site according to the guidelines of Wood (1976). Dissolved-oxygen concentration and Eh were measured in water passing through a flow-through chamber. Stabilization of specific conductance, pH, and water temperature was necessary before a water sample was collected (Claassen, 1982). Samples were filtered through a 0.45-μm (micrometer) filter and preserved with nitric acid following USGS procedures (Thatcher and others, 1977; Fishman and Friedman, 1985). Dissolved ^{222}Rn gas samples were collected into liquid-scintillation vials containing premeasured amounts of mineral-oil-based scintillation cocktail according to Environmental Protection Agency (EPA) approved methods (Environmental Protection Agency, 1978).

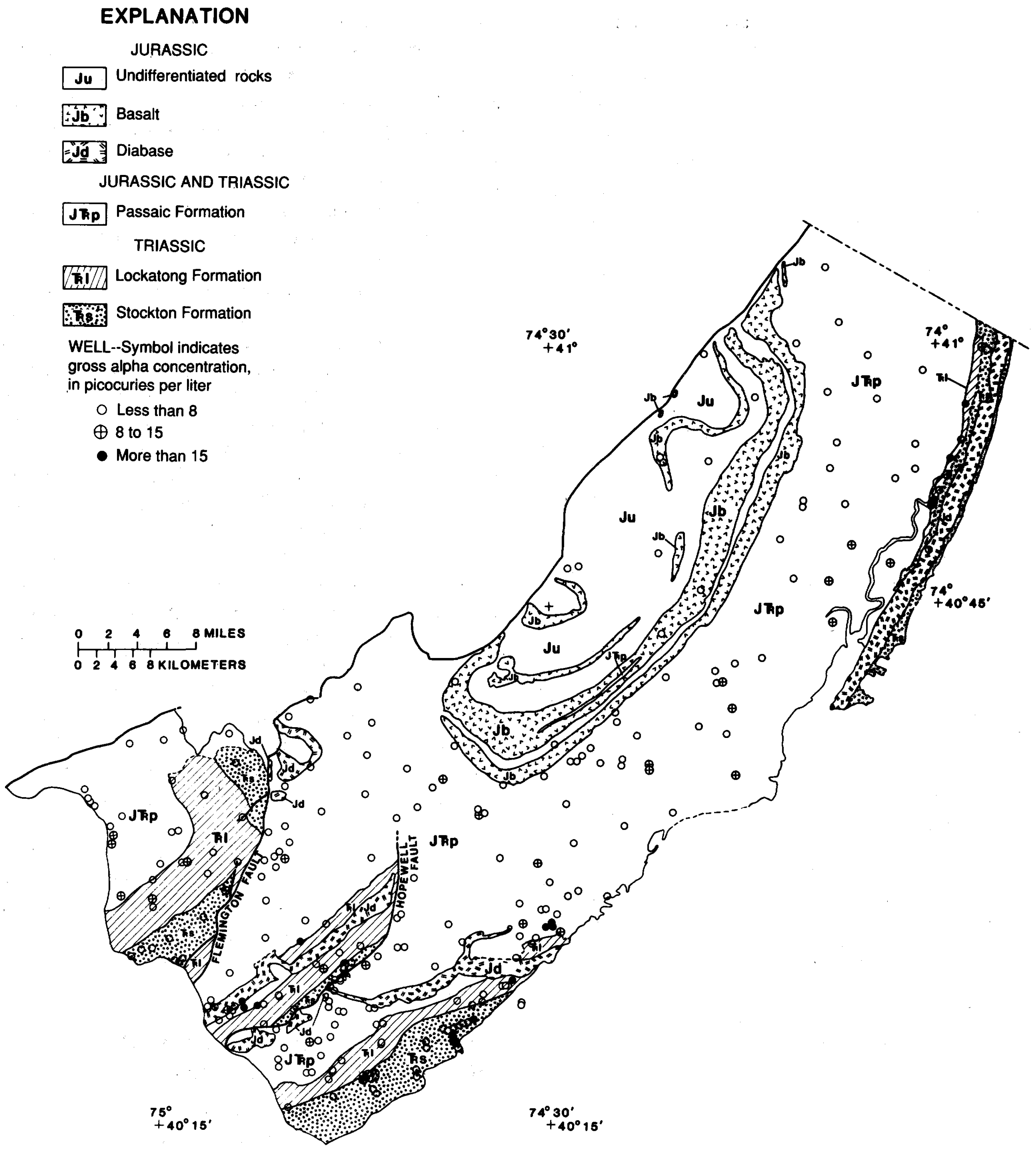


Figure 98. Well-sampling sites and gross alpha-particle activities in ground water from the Newark basin, New Jersey. Scale 1:546,456. Geologic map modified from Lyttle and Epstein (1987).

Analytical Methods

Gross alpha-particle, gross beta-particle, and ^{226}Ra activities were determined by the NJDEP Radiation Protection Laboratory, which used planchet-counting methods in a low-background alpha-beta counter following EPA procedures (Krieger and Whittaker, 1980). Radium-226 was removed from solution by coprecipitation with barium sulfate, and the alpha-particle activity due to the ^{226}Ra was determined by a low-background alpha-beta counter (Krieger and Whittaker, 1980). Radon-222 activity was determined by liquid scintillation (Prichard and Gesell, 1977). The radioactivity (or activity concentration) of these constituents of the water samples is reported in picocuries per liter (pCi/L), where 1 pCi represents 2.2 radioactive disintegrations per minute. If the radioactivity of a sample is so low that the measured activity cannot be clearly distinguished from normal background radiation, the activity of the sample is said to be below the minimum detection limit (MDL). The MDL for gross alpha- and beta-particle activities was 1 pCi/L and was 0.6 pCi/L for ^{226}Ra activity. The measured activity of a sample was reported by the laboratory even if the activity was below the MDL. The measured activities reported by the laboratory are plotted, even if they are below the MDL, to enhance the clarity of the graphics. However, for the purpose of graphical representation, a minimum value of 0.1 pCi/L was set for all samples that had measured activities below this value so that logarithmic graphs could be made.

Uranium concentration was determined by a contractor for the USGS National Water Quality Laboratory by the laser-induced phosphorescence method (Bushaw, 1983) and was reported in micrograms per liter (μg/L). The MDL for uranium concentrations was 0.4 μg/L. The laboratory reported no value for the uranium concentration if the uranium concentration was below the MDL. For the purpose of graphical representation, a value of 0.2 μg/L was set for the uranium concentration for samples for which the uranium concentration was below the MDL.

Concentrations of major cations, anions, and trace metals were determined by standard methods in the USGS National Water Quality Laboratory (Fishman and Friedman, 1985). Blanks, standards, and duplicate samples were submitted to each laboratory for each type of constituent analyzed so that analytical quality could be monitored (Thatcher and others, 1977).

All water samples were analyzed for gross alpha-particle and gross beta-particle activity. If gross alpha radiation exceeded 5 pCi/L, concentrations of uranium, ^{226}Ra, major ions, and trace metals were determined. In addition, concentrations of uranium and ^{226}Ra were determined for selected samples collected from throughout the Newark basin to ensure representative areal coverage. Radon-222 concentrations were determined for selected samples from areas where uranium or ^{226}Ra concentrations of the ground water were known to be elevated.

Statistical Methods

Radionuclide and inorganic chemical constituent concentrations for samples of ground water from the Newark basin do not fit a normal distribution; therefore, nonparametric statistical techniques were used to analyze the data. Descriptive statistical summaries, including the 5th, 25th, 50th (median), 75th, and 95th percentiles were calculated for radionuclide and inorganic chemical constituent concentrations in the ground water. Radionuclide concentrations in excess of the 95th-percentile concentration value (approximately two standard deviations about the mean value) were considered, for the purposes of this report, to be anomalously elevated when compared to the rest of the sample population. An anomaly threshold that includes only the top 5 percent of the data has been used for defining uranium anomalies in other regional studies (Hoffman, 1983). The use of the top 5 percent of the data (two standard deviations about the mean) as the definition of the anomaly threshold is a more stringent definition than the anomaly threshold of one standard deviation about the mean used by Popper and Martin (1982) for uranium data in ground water of the Newark basin.

The geographic distribution of the top 5 percent of the radionuclide data was used to demarcate areas in the Newark basin where concentrations of radionuclides in ground waters were considered anomalously "elevated." Median values for the concentrations of the radionuclides were considered to represent "background" concentrations.

Chemical types of ground water in the Newark basin were delineated by use of a modified version of the classification scheme of Back (1966), based on the distribution of chemical constituents in 125 chemically analyzed samples. Some chemical types of ground water in the classification scheme of Back (1966) were represented by only one or two samples; these samples were grouped with other classes of similar composition, and the result was four broad chemical types of ground water. Median values of radionuclide concentrations were calculated for each of these broad chemical types of ground water and were compared by use of the nonparametric Kruskal-Wallis statistical test, which determines if the median values of ranked concentration values of populations are significantly different.

The relations between concentrations of radionuclides and the concentration of other geochemical constituents were investigated by use of Spearman rank correlations, a nonparametric correlation technique. The magnitude of the correlation coefficient, r, was determined at the 95-percent confidence level.

RADIONUCLIDE ACTIVITIES AND DISTRIBUTION IN GROUND WATER

The range and median value of gross alpha- and gross beta-particle activity, uranium, ^{226}Ra, and ^{222}Rn concentrations are listed in table 45. Because the concentrations of these radioactive constituents are skewed strongly toward lower values, low concentrations of these radionuclides are suggested to be the norm, or background, for most ground water in the Newark basin.

Distribution of Gross Alpha-Particle Activity

The gross alpha-particle activity of a sample measures the total number of alpha emissions derived from radionuclides in solution, including the activity of the isotopes of uranium, ^{226}Ra, and other alpha-emitting

Table 45. Range and median values of radionuclide concentrations in ground water in the Newark basin, New Jersey

[<, less than; values are reported in picocuries per liter, except for uranium, which is reported in micrograms per liter]

Radionuclide	No. of samples	Maximum value	Minimum value	Median value
Gross alpha particle . .	260	124	<1	3.3
Gross beta particle . . .	260	49.7	<1	3.1
Uranium	134	60	<.4	3.1
Radium-226	178	22.5	<.6	<.6
Radon-222	30	15,900	<71	1,390

nuclides in the uranium and thorium radioactive-decay series. However, the activity of ^{222}Rn is not measured, because it is a gas that escapes by outgassing as the sample is evaporated during preparation.

The gross alpha-particle activity generally indicates the total amount of uranium and ^{226}Ra present in the sample; this was true of ground waters in the Newark basin. The combined activities of uranium and ^{226}Ra equaled, or nearly equaled, the gross alpha-particle activity in almost all samples.

Gross alpha-particle activities ranged from less than 1 to 124 pCi/L, with a median value of 3.3 pCi/L. Sixty-five percent of the ground-water samples had gross alpha-particle activities less than 5 pCi/L, and 80 percent had activities of less than 8 pCi/L. Thirteen percent of the samples had gross alpha-particle activities between 8 and 15 pCi/L, 7 percent had activities greater than 15 pCi/L, and 5 percent had activities greater than 19 pCi/L.

Gross beta-particle activities ranged from less than 1 to 49.7 pCi/L, with a median value of 3.1 pCi/L. Five percent of the ground-water samples had gross beta-particle activities greater than 11 pCi/L. Anomalously elevated gross beta-particle activity (greater than 11 pCi/L in this report) is found only in ground-water samples that also had anomalously elevated gross alpha-particle activity. The areal distribution of samples that had elevated gross beta-particle activity parallels the areal distribution of samples that had elevated gross alpha-particle activity. Radiochemical analyses to determine the activity of specific beta-particle-emitting radionuclides were not conducted, and gross beta-particle activity is not discussed further in this report.

Anomalously elevated gross alpha-particle activities (defined in this report as greater than 19 pCi/L) were detected in ground waters from several of the rock formations in the Newark basin. Elevated gross alpha-particle activities are found predominantly in ground water along the south and eastern border of the study area in the lower part of the Passaic Formation and the upper part of the Lockatong Formation, and in the Hopewell and Flemington fault blocks, where these same geologic formations are repeated through faulting. Many of the elevated gross alpha-particle activities are in water from wells that are located very near to the mapped stratigraphic contact of the Passaic and Lockatong Formations. In addition, elevated gross alpha-particle activities are found locally in the lower part of the Lockatong Formation and in the Stockton Formation. Ground water from the upper part of the Passaic Formation, from the basalt units and diabase units, and from the area shown as Jurassic undifferentiated (fig. 98) is characteristically very low in gross alpha-particle activity.

The distribution of anomalously elevated gross alpha-particle activities in ground water from predominantly the same stratigraphic interval basinwide suggests that conditions existed for uranium enrichment of these formations during a specific period of basin deposition. During early diagenesis, this deposited uranium was redistributed and became even more enriched in beds along reducing fronts in proximity with black organic-carbon-rich lake beds.

Distribution of Uranium, Radium-226, and Radon-222

The areas of anomalously elevated concentrations of uranium and ^{226}Ra in ground water correspond to areas of anomalously elevated gross alpha-particle activity in ground water and uranium enrichment in the underlying rock. Therefore, anomalously elevated concentrations of gross alpha particles, uranium, and ^{226}Ra in ground water of the study area are related spatially. However, a major conclusion of this study is that, in the Newark basin, anomalously elevated concentrations of both uranium and ^{226}Ra are rarely found concurrently in the same ground-water sample (fig. 99). The lack of concurrence within a given water sample reflects differences in geochemical behavior between uranium and radium.

Uranium concentrations ranged from less than 0.4 to 60 μg/L, with a median value of 3.1 μg/L. Eleven percent of the ground-water samples had uranium concentrations greater than 10 μg/L, 7 percent had concentrations greater than 15 μg/L, and 5 percent had concentrations greater than 21 μg/L. Eighty-six percent of the samples had uranium concentrations greater than 0.4 μg/L, the MDL.

Radium-226 concentrations ranged from less than 0.6 to 22.5 pCi/L, with a median value of less than the MDL of 0.6 pCi/L; 75 percent of the ground-water samples had ^{226}Ra concentrations less than 0.6 pCi/L. Sixteen percent of the samples had ^{226}Ra concentrations greater than 1 pCi/L, and 5 percent had concentrations greater than 4 pCi/L. Four percent of the samples exceeded 5 pCi/L.

Anomalously elevated concentrations of uranium (defined in this report as greater than 21 μg/L) and (or) ^{226}Ra (defined in this report as greater than 4 pCi/L) are present predominantly in ground water from wells near or along the stratigraphic contact of the Passaic and Lockatong Formations, where gross alpha-particle activities are high.

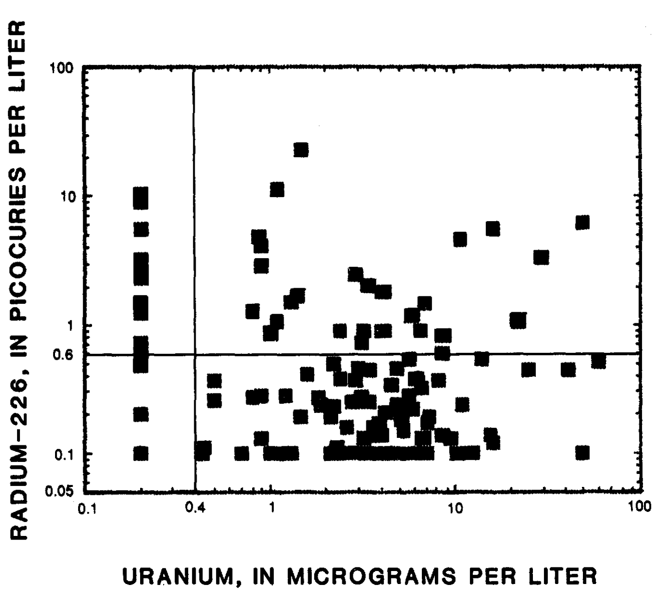


Figure 99. Scatter plot of radium-226 concentration as a function of uranium concentration in ground water in the Newark basin, New Jersey. The solid vertical and horizontal lines represent the minimum detection limits for uranium and radium-226, respectively.

Elevated activities of uranium and ^{226}Ra also are present locally in other parts of the Lockatong Formation and in the Stockton Formation. In these areas, uranium enrichment in the rock has been demonstrated during the course of this study. In contrast, large areas of the upper parts of the Passaic Formation, the basalt and diabase units, and the area shown as undifferentiated rocks (fig. 98) have ground water with low gross alpha-particle uranium and ^{226}Ra concentrations.

In ground water in the lower part of the Passaic Formation, anomalously elevated concentrations of uranium and ^{226}Ra are found with about equal frequency. However, moving up section in the Passaic Formation, away from the interbedded black and red beds characteristic of the lower part of the Passaic Formation and the upper part of the Lockatong Formation, the presence of anomalously elevated uranium concentrations becomes more frequent than the presence of anomalously elevated ^{226}Ra concentrations in the ground water. Anomalously elevated uranium concentrations are present more often in the Stockton Formation, whereas, anomalously elevated ^{226}Ra concentrations are present more often in the Lockatong Formation. A possible explanation for this is that ground-water chemistry is different within each of these formations, and the ability of ^{226}Ra and uranium to go into solution depends upon the chemistry of the ground water.

Radon-222 concentrations ranged from less than 71 to 15,900 pCi/L, with a median value of 1,390 pCi/L. Thirteen percent of the ground-water samples had ^{222}Rn concentrations greater than 10,000 pCi/L. Thirty-five percent of the ground-water samples has ^{222}Rn concentrations greater than 2,000 pCi/L, and 87 percent had ^{222}Rn concentrations greater than 200 pCi/L. Because of the relatively small number of ^{222}Rn samples collected from the Newark basin, and because of uncertainty of the form of the population distribution (Kratochuil, 1985), significance was not attributed to the 95th percentile of ^{222}Rn concentrations in ground water. This lack of strong correlation indicates that ^{222}Rn concentrations in ground water are controlled by different factors than those controlling uranium or ^{226}Ra.

Concentrations of ^{222}Rn greater than 10,000 pCi/L are present in all three of the formations where elevated gross alpha-particle activities are found (the lower part of the Passaic Formation, the Lockatong Formation, and the Stockton Formation). The highest concentrations of ^{222}Rn are present in the lower part of the Passaic Formation, as are the highest concentrations of uranium and ^{226}Ra. However, samples having the highest concentrations of ^{222}Rn do not necessarily coincide with samples having the highest concentrations of gross alpha particles, uranium, or ^{226}Ra. Variation in ^{222}Rn concentrations is considerable and can be an order of magnitude different in proximate wells. Samples that have the highest concentrations of ^{222}Rn correlate better with the highest concentrations of ^{226}Ra than with uranium (fig. 100).

DETERMINATION OF SOURCES OF RADIOACTIVITY BY BOREHOLE GEOPHYSICS AND TEST-CORE ANALYSIS

Borehole geophysics and rock coring were used to determine the subsurface source of radioactivity at specific sites within the Newark basin. Natural gamma-ray logging of wells and boreholes records the background rate of emission of gamma rays from the various formations that were penetrated as well as recording the anomalously high gamma activity of radioactive zones. Gamma-activity measurements vary, depending on the sensitivity of the instrument used for the measurement, changes in borehole diameter, and well-construction materials. Therefore, the data reported for gamma activity in counts per second (cps) are relative numbers. Gamma-spectral logging can be used to ascertain the presence of (and, with the appropriate calculations, to determine the concentration of) uranium, thorium, and potassium in rock if the intensity of gamma emissions is measured at different energy levels.

Background gamma levels for sedimentary rocks of the Newark basin measured in this study typically range from 100 to 160 cps. Natural gamma-ray logs of many wells in the upper part of the Passaic Formation and in the undifferentiated areas (fig. 97) show no significant gamma levels above background. Natural gamma-ray logs of wells in basalt and diabase recorded the lowest gamma counts of all the Newark basin units, with background gamma levels ranging from 20 to 40 cps.

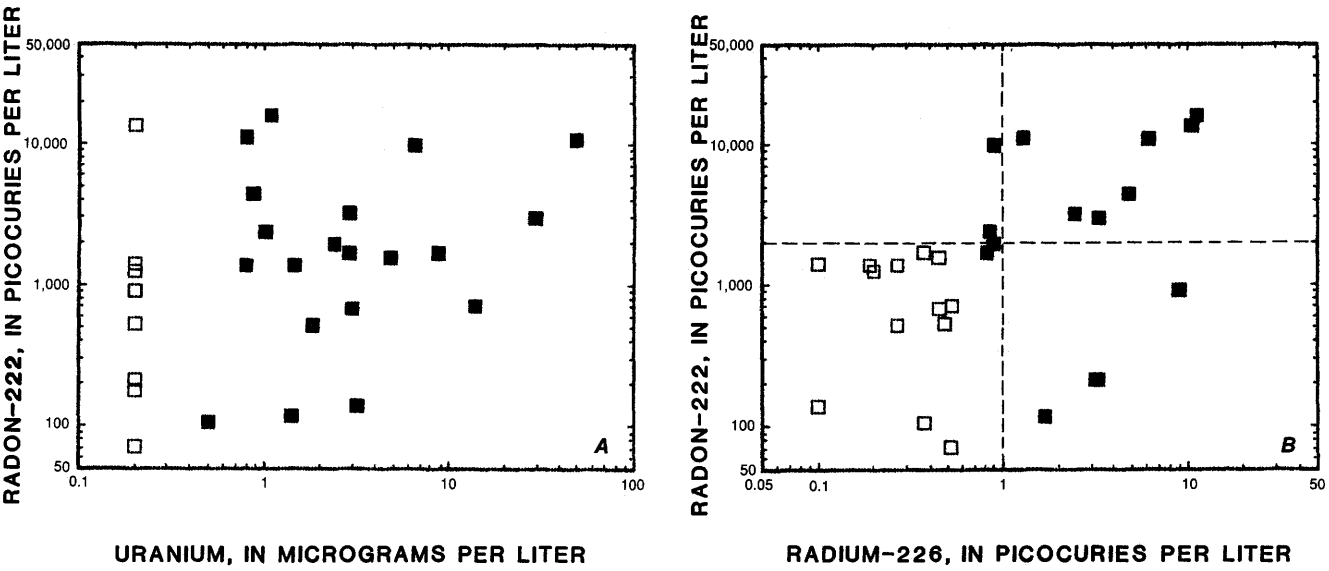


Figure 100. Scatter plot of radon-222 concentration as a function of (*A*) uranium concentration and (*B*) radium-226 concentration in ground water in the Newark basin, New Jersey. A vertical dashed line indicates a radium-226 concentration of 1 picocurie per liter (pCi/L), whereas a horizontal dashed line indicates a radon-222 concentration of 2,000 pCi/L. The open symbols represent uranium concentrations below the detection limit of 0.4 μg/L, plotted as 0.2 μg/L (*A*), and radium-226 concentrations below the detection limit of 0.6 pCi/L, plotted at the reported value measured below the detection limit, with a minimum plotting value set at 0.1 pCi/L (*B*).

Numerous gamma anomalies of several thousand cps were recorded on well logs from the Lockatong Formation and the lower part of the Passaic Formation. These anomalies are associated with rock units that are characteristically thin (less than 1 ft thick). The anomalies are laterally extensive and can be traced in adjacent wells over a lateral distance of several thousand feet. High counts on natural gamma-ray logs of wells in the Stockton Formation also have been observed. These anomalies are also associated with rock units that are thin. However, these anomalies are more localized and cannot be traced in adjacent wells. The distribution of elevated activities of radionuclides in ground water of the Newark basin is consistent with the occurrence and extent of natural radioactive anomalies recorded in the subsurface rock formations.

Three continuous cores were examined for this study (fig. 97). Two corehole sites (A and B) were located near the geologic contact of the Passaic and Lockatong Formations, and one site (corehole C) was located in the Stockton Formation. Sites A and B were cored adjacent to existing wells for which natural gamma-ray logging indicated several gamma anomalies ranging from 1,500 cps to more than 7,000 cps. The depths of the anomalous rock zones were used to determine how deep to drill the coreholes.

Continuous 2-in-diameter core was obtained to a depth of 68 ft in the lower part of the Passaic Formation at corehole site A. A natural gamma-ray log, a generalized lithologic description, and an interpreted environment of deposition are shown in figure 101. The cyclic sequence of red, gray, and black mudstones shows the expansion and contraction of an extensive lake as suggested by Van Houten (1969). The red beds, deposited near the margin of the lake in a mudflat/playa environment, indicate oxidizing conditions prevailed at the time of their deposition. The progressively darker colored gray beds represent expansion and deepening of the lake. The black mudstones formed farthest offshore under anoxic conditions, and coupled with the presence of pyrite, are indicative of a highly reducing environment at the time of their deposition that is ideal for the precipitation of uranium.

The natural gamma-ray log of corehole A (fig. 101) recorded a gamma anomaly at 42 to 43 ft below land surface. Gamma-spectral logging of the zone containing the gamma anomaly in corehole A identified the radioactivity as uranium-series radionuclides (fig. 101).

The spatial distribution of radionuclides within this anomalous rock zone was determined by autoradiography of core material. In autoradiography, radioactive emissions from the rock sample are allowed to strike a zinc-sulfide-coated scintillation plate. Flashes of light that are produced are recorded on photographic film. The zone in the rock containing the most radioactivity appears as the lightest (most developed) on the photographic film.

Autoradiographs of vertical slices of rock core from the depth interval of 42 to 43 ft show abundant radionuclides finely disseminated throughout much of the interval. Radionuclides appear most concentrated in two thin zones on core pieces from depths of 42.3 to 42.4 ft and 42.6 to

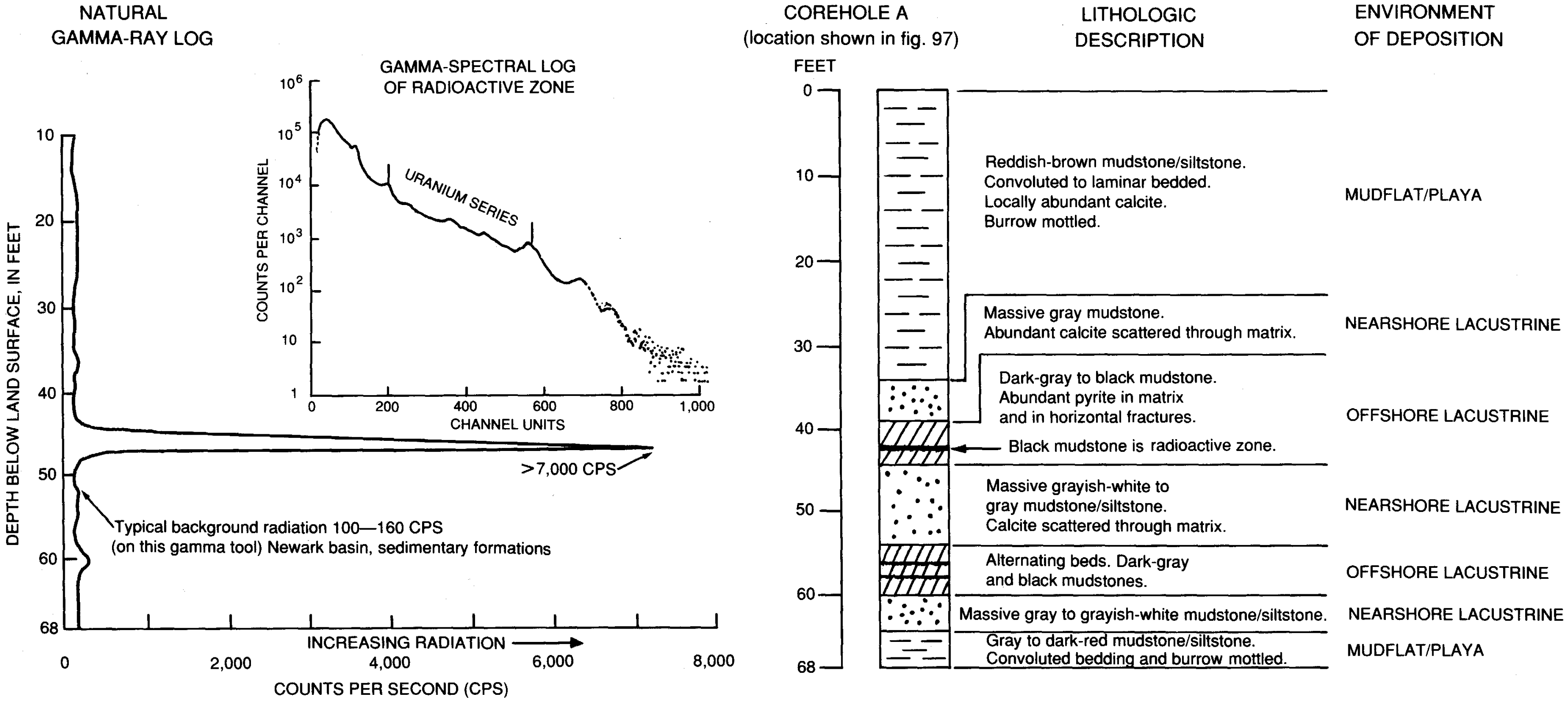


Figure 101. Gamma-ray log, lithologic description, and environment of deposition of corehole A, and gamma-spectral log of radioactive zone in the 42- to 43-ft depth interval.

42.8 ft. The autoradiograph of the core from a depth of 42.3 to 42.4 ft shows elevated concentrations of radionuclides (light color) in association with interbedded carbonate and black mudstone laminae in a zone approximately 0.5 in thick (fig. 102*A*). The autoradiograph of the core from a depth of 42.6 to 42.8 ft shows elevated concentrations of radionuclides partly in association with horizontal bedding planes. However, in the upper left-hand part of the autoradiograph, elevated concentrations of radionuclides crosscut the bedding planes (arcuate shape) and cannot be definitively associated with any lithologic feature on the hand specimen (fig. 102*B*). Within these arcuate radioactive anomalies, the greatest concentration of radionuclides appears along two thin, horizontal, wavy, carbonate-rich beds.

Radiochemical analysis of ground water from the 68-ft corehole (open interval 12 to 68 ft) shows concentrations of 66.4 pCi/L for gross alpha-particle activity, 25.2 pCi/L for gross beta-particle activity, and 22.5 pCi/L for ^{226}Ra. In comparison, an adjacent well that is 409-ft deep (open interval 30 to 409 ft) intersects the same radioactive zone and shows concentrations of 44.9 pCi/L for gross alpha-particle activity, 18.1 pCi/L for gross beta-particle activity, and 9.0 pCi/L for ^{226}Ra. Uranium concentration was below the MDL. These radiochemical analyses indicate vertical changes in ground-water radioactivity at the site. Concentrations of radioactivity in ground water from the 409-ft-deep well are less than those of the shallower corehole because radioactive water in the deeper well is diluted by the mixing of less radioactive water from deeper water-bearing zones within the well.

At corehole B, approximately 20 mi to the southwest of corehole A (fig. 97), a continuous core of the upper part of the Lockatong Formation was collected to a depth of 187 ft. The lithologic sequence encountered was similar to that of corehole A. In corehole B, two radioactive zones, each approximately 0.5 ft thick, were encountered. Both zones are black mudstones that have abundant pyrite mineralization in the matrix and locally along bedding planes. Although these zones are thin, they appear to have great lateral extent. Because these radioactive zones have been recorded on a natural-gamma log from another well more than 1,100 ft from the corehole, these zones can function as stratigraphic marker beds. Gamma-spectral logging of these radioactive zones indicates the radioactive elements present were uranium- and thorium-series radionuclides.

Autoradiographs of core from these two radioactive zones show the radionuclides finely dispersed throughout the rock matrix, similar to the distribution of radionuclides at a depth of 42.3 to 42.4 ft in corehole A (fig. 102*A*). Arcuate bands of radionuclides crosscutting bedding were not observed in either radioactive zone from corehole B.

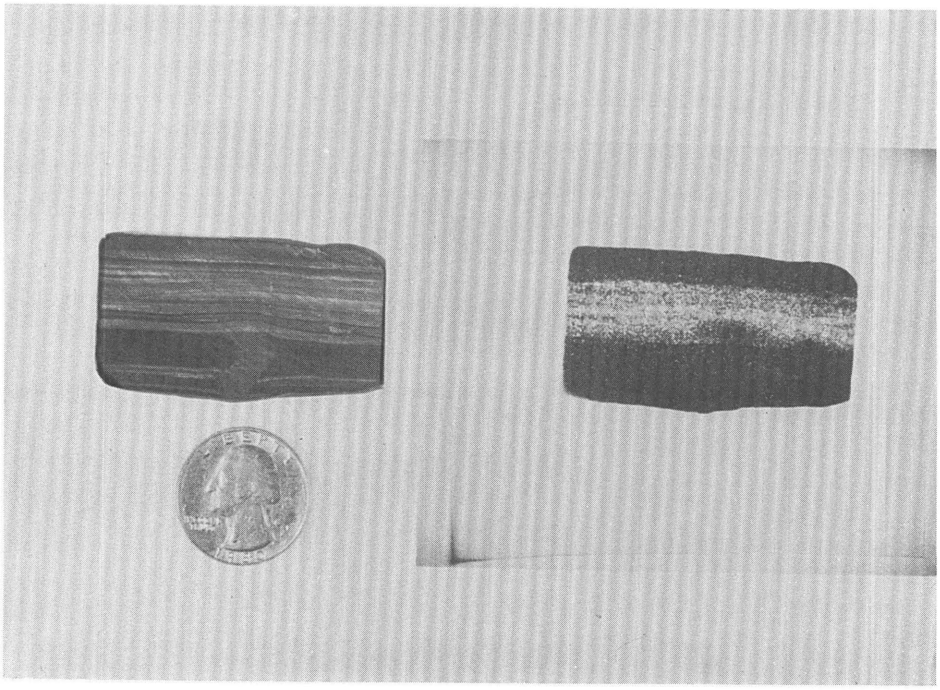
Radionuclide activities of a ground-water sample from a nearby observation well, open to a depth of several hundred feet, were at the median values for the Newark basin (table 45). Because fracturing of the rock material was not observed in the proximity of the radioactive zones, it is possible that significant amounts of ground water do not come in contact with these zones or that ground water from other well-developed fracture zones is present in the observation well in large enough quantities to dilute any radioactivity contributed from the unfractured radioactive-rock zone.

Uniform dispersion of radionuclides within black mudstones of the lower part of the Passaic Formation and the Lockatong Formation probably indicates uranium enrichment during, or soon after, deposition. This uniformity is consistent with syngenetic concentration of uranium in response to reducing conditions near the sediment-water interface of the lake bottom, as suggested by Turner-Peterson and others (1985). The origin of the thin, highly concentrated zones of radioactivity, found in association with horizontal carbonate-mudstone interbeds, and the radioactivity that crosscuts the bedding is probably due to secondary uranium migration and enrichment.

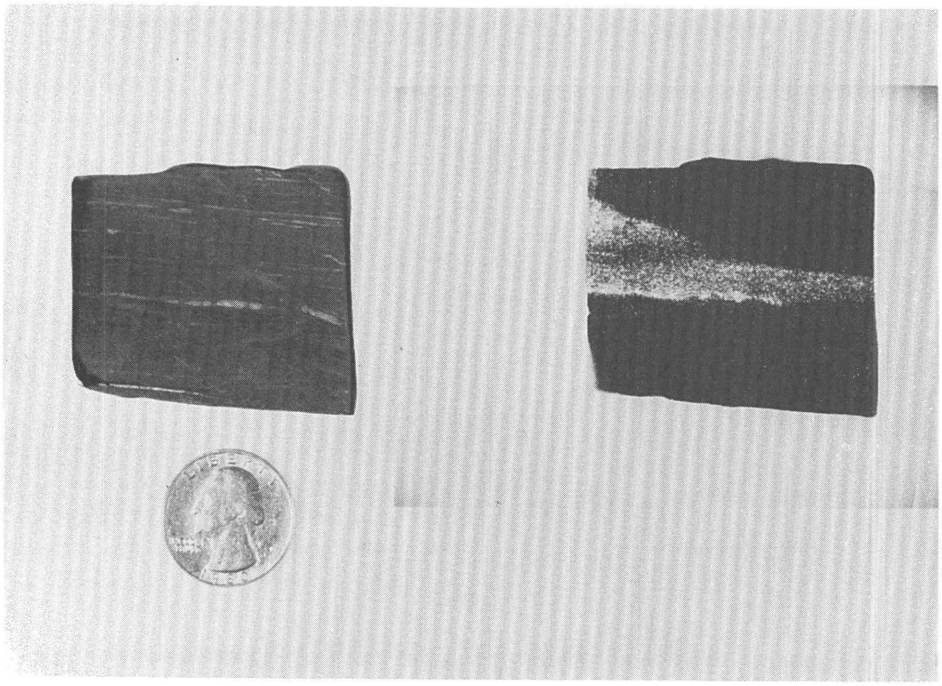
At corehole C (fig. 97), continuous core of the Stockton Formation was collected to a depth of 120 ft (Lewis and Spitz, 1987). Subsequent natural gamma-ray logging indicated a gamma anomaly of 600 cps at a depth of 104 ft. A hand sample from the core of this radioactive zone is composed of fine- to medium-grained sandstone cut by two near-vertical fractures that are coated with a white clay mineral. In reference to uranium occurrences in outcrop of the Stockton Formation, Popper and Martin (1982) indicate that uraniferous sandstones have a higher clay content than adjacent rocks that exhibit only background levels of uranium. They also observe that the highest radioactivity is present in fracture zones or bedding contacts where high amounts of clay are present.

Autoradiographs of the core from 104 ft show abundant radionuclides dispersed along the face of the fracture. Because the fracture face was a highly irregular surface, the autoradiograph was not clear enough to determine whether the radioactivity is associated with the fracture-fill material or the sandstone matrix.

Ground water from a test well adjacent to the corehole and open to the aquifer between a depth of 90 to 110 ft has concentrations of 30.4 pCi/L for gross alpha-particle activity, 15.5 pCi/L for gross beta-particle activity, and 11.4 pCi/L for ^{226}Ra. Uranium concentration was below the MDL. The abundant fracturing of the radioactive zone enabled ground water to come in contact with the minerals hosting the radionuclides in the rock strata or along the fracture. Dilution of radionuclide concentrations in water from the test well was not possible because the test well was only open to rock strata containing the radioactive zone.



A



B

Figure 102. Rock samples (left) and autoradiographs (right) of radiation development (light color) from corehole A at depth intervals of (*A*) 42.3 to 42.4 ft and (*B*) 42.6 to 42.8 ft.

GROUND-WATER GEOCHEMISTRY OF URANIUM AND RADIUM IN THE NEWARK BASIN

The observed distribution of uranium and ^{226}Ra in the ground water is related to their different geochemical behavior, as predicted by thermodynamics and laboratory studies and verified by chemical analysis of ground-water samples from the field. By understanding the geochemistry of uranium and radium, we intend to identify other indicators of the presence of uranium and radium under given geochemical conditions. For this purpose, selected ground-water samples from all rock types in the Newark basin were analyzed chemically. (The range and median of selected constituents are given in table 46.)

Water-Quality Characteristics of Ground Water in the Newark Basin

Ground water in the Newark basin is primarily a calcium magnesium sodium bicarbonate sulfate type, although some calcium magnesium sodium sulfate bicarbonate, sodium bicarbonate, and calcium sodium magnesium chloride bicarbonate type of water also is present. Total dissolved solids range from 103 to 1,700 mg/L (milligrams per liter), with a median value of 260 mg/L (table 46). All the ground water contains abundant bicarbonate (median concentration is 163 mg/L). Calcium, bicarbonate, and sulfate have the greatest range of concentrations among major ions. All major cations and anions increase as dissolved-solids contents increase, except in samples of the sodium bicarbonate type ground water, where calcium, magnesium, and sulfate decrease as sodium and bicarbonate increase. Furthermore, bicarbonate concentrations decrease slightly with increasing dissolved-solids contents in ground-water samples of the calcium magnesium sodium sulfate bicarbonate type. However, bicarbonate, calcium, and sulfate concentrations increase most strongly with increasing dissolved-solids contents in ground-water samples. These results are generally consistent with previous findings in the Newark basin (Herpers and Barksdale, 1951; Longwill and Wood, 1965).

The ground water in most cases is slightly alkaline and strongly oxidizing; however, some waters that are slightly acidic, or are reducing, are present. Values of pH range from 5.5 to 9.8 and have a median of 7.6 (table 46). Fifty percent of the pH values range from 7.2 to 7.8. Dissolved-oxygen values range from less than 0.1 mg/L to 9.8 mg/L and have a median concentration of 2.8 mg/L. Approximately 30 percent of the wells sampled had dissolved-oxygen concentrations of less than 1 mg/L.

Iron, strontium, and manganese have the greatest range of concentrations among trace metals. Iron and manganese concentrations increase most strongly with decreasing dissolved-oxygen content. Strontium concentrations increase strongly (correlation coefficient, r,= +0.63) and barium concentrations decrease strongly (r= −0.48) with increasing sulfate concentrations.

Table 46. Range and median values of selected water-quality characteristics and constituents in ground water from the Newark basin, New Jersey

	No. of samples	Minimum	Maximum	Median
Characteristic				
pH (pH units)	260	5.5	9.8	7.6
Eh (millivolts)	256	24	827	382
Major ions (in milligrams per liter)				
Dissolved oxygen .	248	<.1	9.8	2.8
Bicarbonate	260	26	460	163
Calcium	121	2.5	360	46
Sulfate	121	1.6	1,100	40
Total dissolved solids	121	103	1,700	260
Trace metals (in micrograms per liter)				
Barium	116	<2.0	1,200	110
Iron	121	<3.0	11,000	8.0
Manganese	121	<1.0	1,600	7.0
Strontium	116	50	11,000	425

Relation Between Concentrations of Radionuclides and Chemical Constituents in Ground Water

Chemical and radiochemical analyses of ground water in the Newark basin indicate that several chemical constituents may serve as indicators of the presence of elevated concentrations of uranium or ^{226}Ra. Anomalously elevated gross alpha-particle activity of the ground water coupled with dissolved-oxygen content, or the Eh, accurately predicts the presence of elevated uranium or ^{226}Ra concentration (figs. 103 and 104). Anomalously elevated gross alpha-particle activity in oxidizing waters is associated with anomalously elevated concentrations of uranium; however, anomalously elevated gross alpha-particle activity in reducing waters is associated with anomalously elevated concentrations of ^{226}Ra. With few exceptions, anomalously elevated uranium and ^{226}Ra concentrations were not present concurrently in ground-water samples (fig. 99).

Specific chemical constituents of the ground water also relate to radioactivity (figs. 105–107). Although uranium concentration shows negative correlations with iron and manganese concentrations (r=−0.40 and −0.30, respectively; fig. 105*A*), positive correlations with strontium and sulfate concentrations are noted (r =+0.32 and +0.24, respectively; fig. 107*A*). Radium-226 shows positive correlations with concentrations of iron and manganese (r =+0.46 and +0.48, respectively; fig. 105*B*). These correlations are all significant at the 95-percent confidence

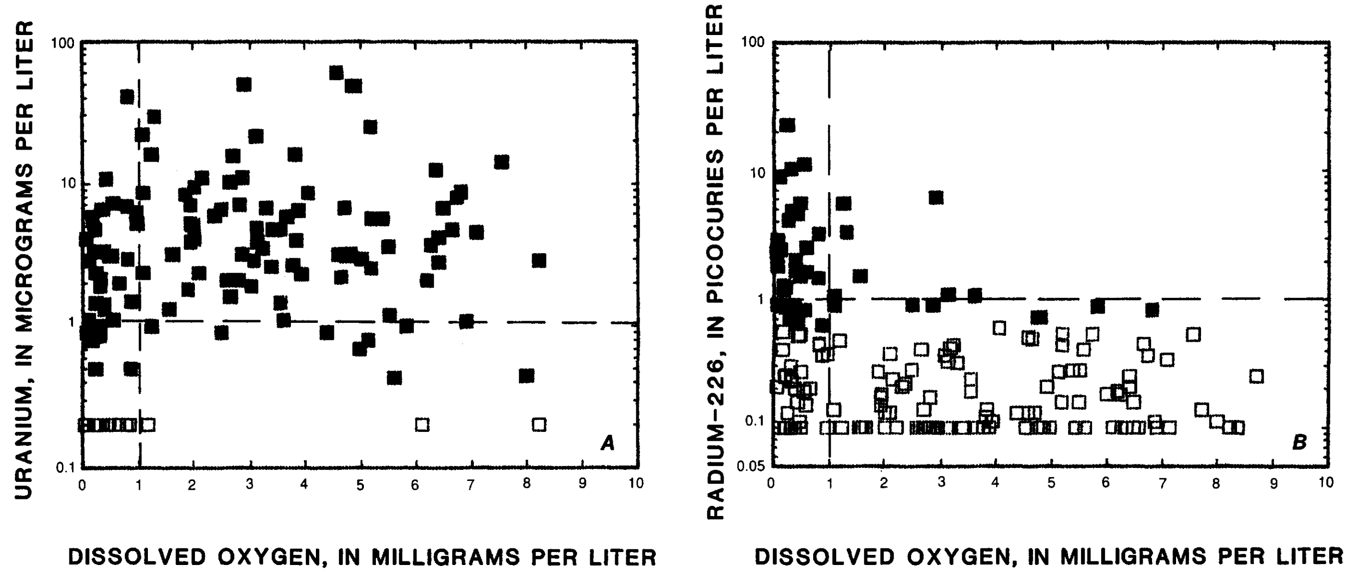


Figure 103. Scatter plots of (*A*) uranium concentration and (*B*) radium-226 concentration as a function of dissolved-oxygen content in ground water in the Newark basin, New Jersey. A vertical dashed line indicates dissolved-oxygen content of 1 mg/L (milligram per liter). A horizontal dashed line indicates uranium concentration of 1 μg/L (microgram per liter) (*A*) and a radium-226 concentration of 1 pCi/L (picocurie per liter) (*B*). The open symbols represent uranium concentrations below the detection limit of 0.4 μg/L, plotted as 0.2 μg/L (*A*), and radium-226 concentrations below the detection limit of 0.6 pCi/L, plotted at the reported value measured below the detection limit, with a minimum plotting value set at 0.1 pCi/L (*B*).

level when nonparametric statistics are used. In addition, ground water that has anomalously elevated uranium concentrations exceeds a pH of 7 (fig. 106*A*) and contains bicarbonate concentrations greater than 100 mg/L. Anomalously elevated ^{226}Ra concentrations are present in ground waters where pH values are below 8 (fig. 106*B*). Ground water that has anomalously elevated ^{226}Ra concentrations usually contains barium concentrations greater than 100 μg/L. Radium-226 concentrations greater than 4 pCi/L were not found in waters where the sulfate anion is present in concentrations above 100 mg/L (fig. 107*B*).

Correlation between radionuclide activities and concentrations of other dissolved chemical constituents, even if statistically strong, does not by itself indicate cause-and-effect relations. Radionuclides and chemical constituents are discussed only in terms of possible geochemical reactions that may control the presence of elevated activities of these radionuclides in ground water of the Newark basin. These postulated geochemical controls on radionuclide activities are based upon published laboratory experiments and calculations using published thermodynamics constants (see the citations in the discussions on radionuclide chemistry that follow) and the correlations noted previously.

The contrasting geochemical behavior of uranium and radium is evident in both the presence or absence of dissolved oxygen. Uranium concentrations above 20 μg/L occur most often in ground water in which the dissolved-oxygen content is above 1 mg/L (fig. 103*A*). Conversely, in ground water that contains less than 1 mg/L of dissolved oxygen, more uranium concentrations are less than 1 μg/L than are concentrations in ground water that contains more than 1 mg/L of dissolved oxygen. Radium-226 concentrations exceed 1 pCi/L predominantly in ground water that contains less than 1 mg/L dissolved oxygen (fig. 103*B*). The contrasting geochemical behavior of uranium and radium also is evident with respect to Eh (the redox potential) (fig. 104). Uranium concentrations increase in the ground water as Eh increases, whereas ^{226}Ra concentrations increase in the ground water as Eh decreases.

Uranium changes valence from the insoluble U^{+4} ion in reducing waters to the highly soluble U^{+6} ion in oxidizing waters (Langmuir, 1978). Where the amorphous uranium oxides contained in the rocks of the Newark basin (Turner-Peterson and others, 1985) come in contact with the typical oxygen-rich ground water, the uranium is easily oxidized and dissolved. Where the ground water is oxygen deficient, the uranium cannot be oxidized to the soluble valence state and does not appear in solution, even if it is abundantly present in the rock (as at corehole A, for example).

Radium in the natural environment is present only as the Ra^{+2} ion. Because the valence state of the radium ion does not change, changing redox conditions (determined primarily, but not exclusively, by oxygen content) of the ground water do not directly influence the solubility of the radium ion. Yet, radium concentrations correlate inversely

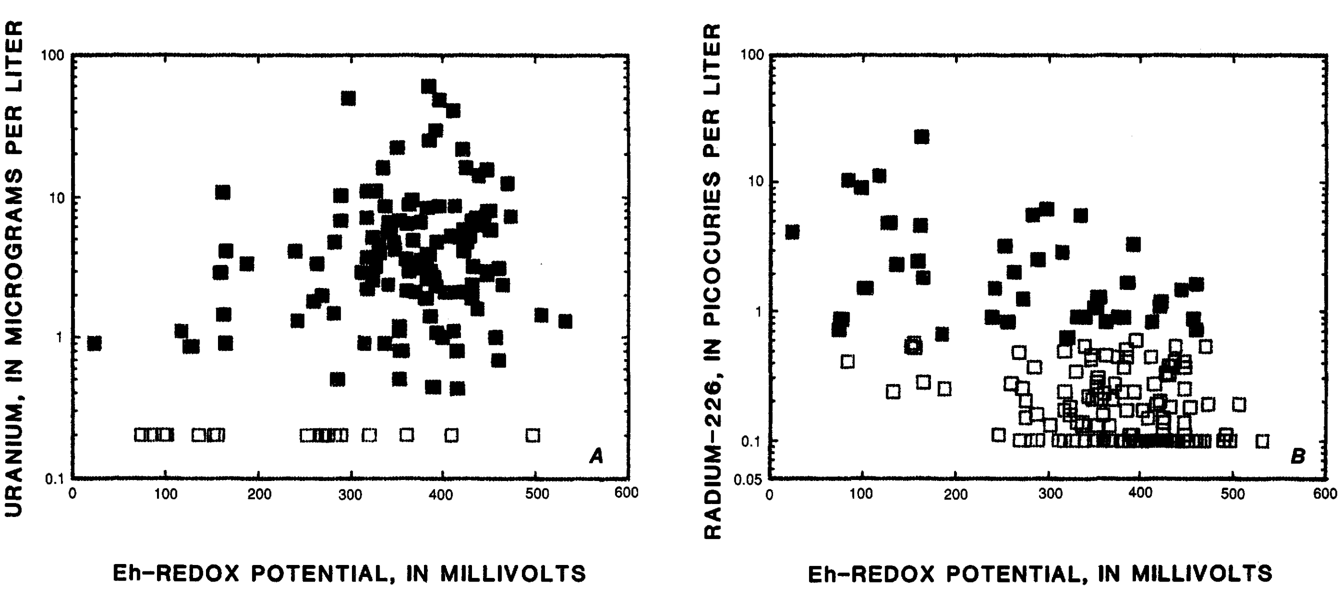


Figure 104. Scatter plots of (*A*) uranium concentration and (*B*) radium-226 concentration as a function of Eh in the ground water in the Newark basin, New Jersey. The open symbols represent uranium concentrations below the detection limit of 0.4 μg/L (microgram per liter), plotted as 0.2 μg/L (*A*), and radium-226 concentrations below the detection limit of 0.6 pCi/L (picocurie per liter), plotted at the reported value measured below the detection limit, with a minimum plotting value set at 0.1 pCi/L (*B*).

with Eh and thus suggest that a redox reaction may indirectly influence the solubility of the radium ion. Consequently, other chemical components of the ground water were investigated to determine their effect on dissolved radium concentrations.

Geochemical Controls on Concentrations of Dissolved Uranium

The negative correlation of uranium with iron (fig. 105*A*) and manganese reflects that in reducing water, uranium is precipitated in the insoluble +4 valence state (Langmuir, 1978), whereas iron and manganese each are present in the soluble +2 valence state (Hem, 1985). Conversely, in oxidizing water, uranium goes into solution in the soluble +6 valence state, whereas iron and manganese, found in essentially insoluble +3 and +4 valence states, respectively, form iron and manganese hydroxide precipitates.

In oxidizing water, iron hydroxides are capable of adsorbing uranium from solution (Ames and others, 1983b). However, uranium is strongly complexed by the carbonate ion in carbonate-rich, oxidizing, slightly alkaline waters (Langmuir, 1978). Uranium that is complexed in solution by carbonate is not adsorbed onto iron hydroxides (Hsi and Langmuir, 1985). Carbonate-rich, oxidizing, slightly alkaline ground waters are predominant in the Newark basin. We propose that uranium in solution in the oxidizing ground waters in the Newark basin is complexed by carbonate and, hence, is not efficiently adsorbed by iron hydroxides. The highest concentrations of uranium are found in moderately alkaline ground waters (pH values exceed 7, see fig. 106*A*) where carbonate complexing of the uranium ion occurs (Langmuir, 1978).

Uranium concentrations show a positive correlation with strontium and sulfate concentrations in the ground waters of the Newark basin. Proportionately more ground-water samples that contain sulfate concentrations above 100 mg/L also contain uranium concentrations above 3 μg/L (the median value) than do ground-water samples that contain sulfate concentrations below 100 mg/L (fig. 107*A*). The median uranium concentration (4.1 μg/L) for ground water that contains more than 100 mg/L of sulfate is nearly double the median uranium concentration (2.7 μg/L) for ground water that contains less than 100 mg/L of sulfate, even though the highest dissolved-uranium concentrations are found in ground waters that contain less than 100 mg/L of dissolved sulfate.

The sulfate-rich waters are mildly oxidizing and contain concentrations of dissolved oxygen and bicarbonate typical (near the medians) of ground water in the Newark basin. Both strontium and sulfate increase with increasing dissolved-solids content in the sulfate-rich waters, with the concentration of strontium correlating strongly with that of sulfate. The median concentration value of strontium in the sulfate-rich ground water (3.2 mg/L) is well above the median concentration value of strontium in ground-water samples from the entire data set (table 46). The correlation between concentrations of sulfate and strontium, a soluble

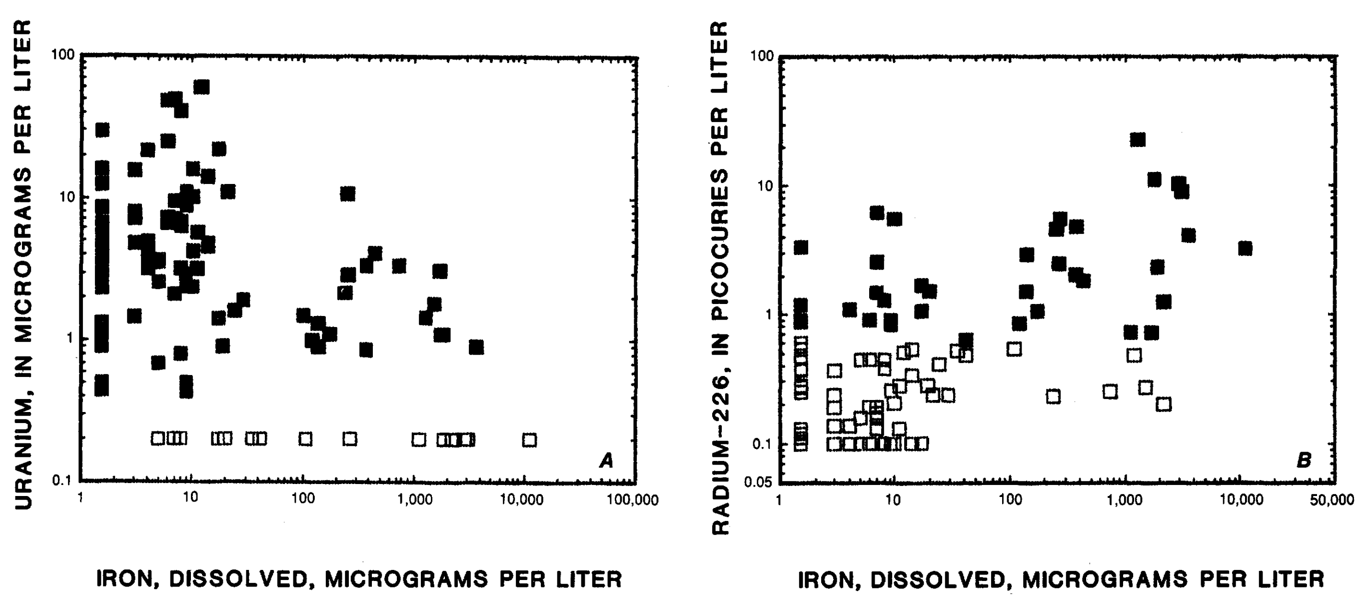


Figure 105. Scatter plot of (*A*) uranium concentration and (*B*) radium-226 concentration as a function of dissolved-iron concentration in the ground water in the Newark basin, New Jersey. The open symbols represent uranium concentrations below the detection limit of 0.4 µg/L (microgram per liter), plotted as 0.2 µg/L (*A*), and radium-226 concentrations below the detection limit of 0.6 pCi/L (picocurie per liter), plotted at the reported value measured below the detection limit, with a minimum plotting value set at 0.1 pCi/L (*B*).

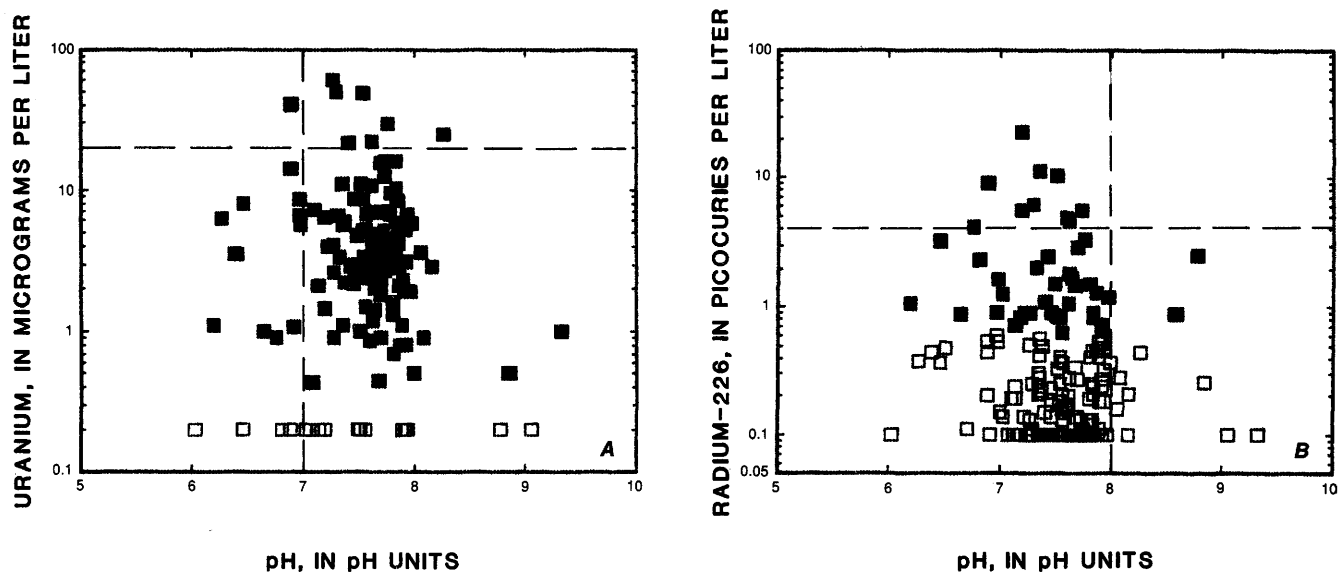


Figure 106. Scatter plot of (*A*) uranium concentration and (*B*) radium-226 concentration as a function of pH in ground water in the Newark basin, New Jersey. A vertical dashed line indicates a pH value of 7 (*A*) and 8 (*B*). A horizontal dashed line indicates uranium concentration of 20 µg/L (micrograms per liter) (*A*) and a radium-226 concentration of 4 pCi/L (picocuries per liter) (*B*). The open symbols represent uranium concentrations below the detection limit of 0.4 µg/L, plotted as 0.2 µg/L (*A*), and radium-226 concentrations below the detection limit of 0.6 pCi/L, plotted at the reported value measured below the detection limit, with a minimum plotting value set at 0.1 pCi/L (*B*).

divalent cation, suggests these sulfate-rich waters have accumulated high amounts of dissolved mineral matter by leaching a large volume of rock or by residing a long time in the rock. Because these waters are oxidizing, uranium

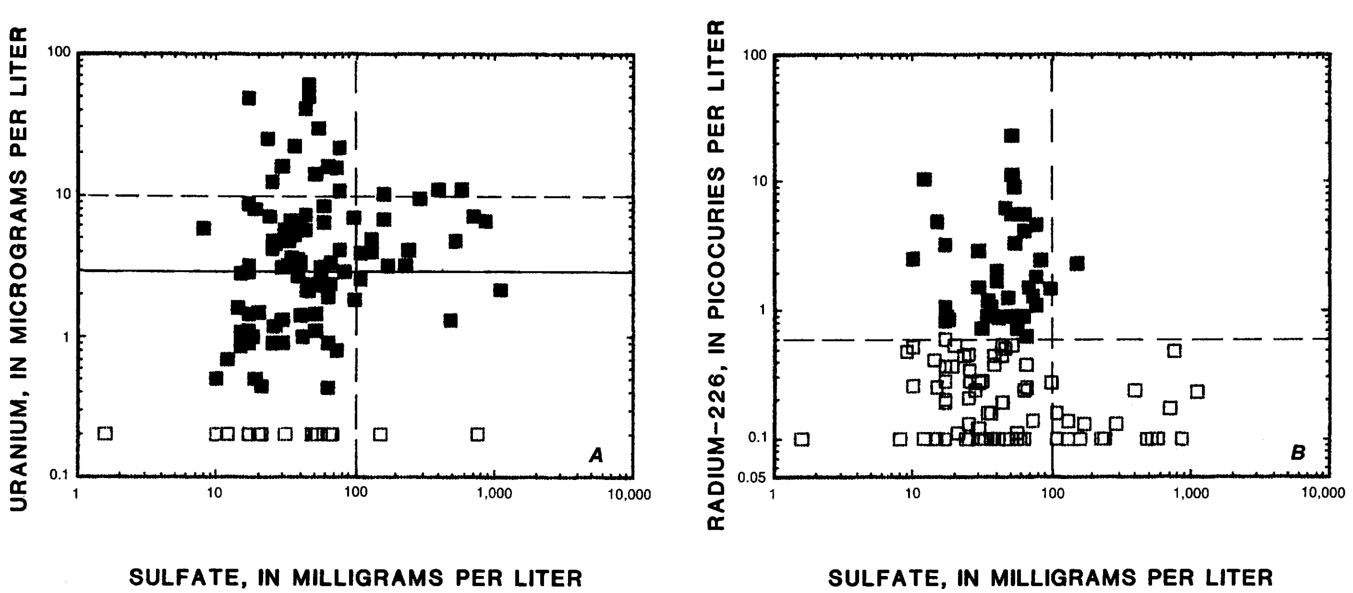


Figure 107. Scatter plot of (*A*) uranium concentration and (*B*) radium-226 concentration as a function of sulfate concentration in ground water in the Newark basin, New Jersey. A vertical dashed line indicates sulfate concentration of 100 mg/L (milligrams per liter). A horizontal solid line indicates uranium concentration of 3 μg/L (micrograms per liter) (*A*); a horizontal dashed line indicates uranium concentration of 10 μg/L (*A*), and a radium-226 concentration of 0.6 pCi/L (picocurie per liter) (*B*). The open symbols represent uranium concentrations below the detection limit of 0.4 μg/L, plotted as 0.2 μg/L (*A*), and radium-226 concentrations below the detection limit of 0.6 pCi/L, plotted at the reported value measured below the detection limit, with a minimum plotting value set at 0.1 pCi/L (*B*).

can accumulate (increase in concentration), as does the other dissolved mineral matter, such as sulfate, calcium, and strontium. In addition, both carbonate complexing of the uranium by the abundant dissolved bicarbonate and the increased competition for adsorption sites with the elevated amounts of dissolved calcium would inhibit uranium adsorption from these sulfate-rich waters and thus allow the accumulated uranium to stay in solution. Therefore, these waters often have moderately, but not anomalously, elevated uranium concentrations relative to the median uranium concentration of ground-water samples from the Newark basin.

In the Newark basin, ground-water samples with relatively low dissolved solids contents that contain uranium concentrations above 20 μg/L are generally found in areas of uranium-enriched rock strata. However, sulfate-rich ground-water samples with relatively high dissolved-solids contents that contain uranium concentrations above 3 μg/L, the median uranium concentration, and occasionally exceed 10 μg/L, are generally found in areas where uranium-enriched rock strata are not known to be present. During this study, geophysical logs collected from areas in the Newark basin containing sulfate-rich ground waters have shown no anomalously radioactive rock zones. The implication is that uranium concentration may be relatively elevated in ground water in the Newark basin, even in the absence of uranium-enriched rock, provided that the ground water has a residence time in the rock long enough to leach significant quantities of uranium (and other mineral matter) and that the uranium leached from the rock is not removed from solution by any geochemical process.

Geochemical Controls on Concentrations of Dissolved Radium-226

Radium-226 is present primarily in samples of the dilute calcium magnesium sodium bicarbonate sulfate type of ground waters that contain less than 1 mg/L dissolved oxygen and elevated concentrations of dissolved iron (as high as 11 mg/L). Elevated concentrations of iron and (or) manganese are present in almost all ground water in the Newark basin that has elevated ^{226}Ra activities (fig. 105*B*).

Ames and others (1983a,b) conducted laboratory experiments and concluded that radium was adsorbed onto the surface of all the secondary minerals they examined; however, iron hydroxide adsorbed far more radium (several orders of magnitude) than any of the other minerals. They also found that this adsorption process was most efficient in dilute waters that had low concentrations of divalent cations (like calcium) that could effectively compete with the radium for adsorption sites on the iron hydroxides. Korner and Rose (1977, p. 127) analyzed iron-hydroxide samples from uranium-ore-bearing sandstones in Pennsylvania and found ^{226}Ra activities significantly higher than the uranium activity of the ore-bearing rock. This finding suggests that iron hydroxides incorporated ^{226}Ra from the surrounding environment, presumably by an adsorption mechanism.

Surface-adsorption mechanisms depend on the nature of the sorbent and sorbate, the concentration of sorbate in solution, the concentration of competing ions in solution, and the pH of the solution (Stumm and Morgan, 1981). At a specific pH for each sorbent, enough hydrogen ions are adsorbed to satisfy the negative charge of the sorbent surface; the affinity of positively charged ions in solution for the sorbent surface is thereby decreased or ended. This pH value is termed the "point of zero charge" (Stumm and Morgan, 1981).

Anderson (1983) reported that radium is adsorbed most efficiently when solutions reach pH values of approximately 8 or above. The point of zero charge for iron hydroxides is reported to be just below this pH value (Stumm and Morgan, 1981).

After reviewing this body of field and laboratory research and examining the data gathered in this study, we propose a conceptual model that relates increasing radium concentration in ground water of the Newark basin to increasing iron (and manganese) concentrations and decreasing Eh and pH values, as observed in this study. We conclude that iron (and manganese) hydroxides, which are abundant in the rocks of the Newark basin (Turner-Peterson, 1980), strongly adsorb radium from solution and thereby exert significant control on ^{226}Ra concentration in the ground water.

Radium-226 is continuously released to ground water contained in the pore or fracture space by several mechanisms, including the alpha-recoil mechanism detailed by Tanner (1964a). However, because radium is so strongly adsorbed by iron (and manganese) hydroxides, ^{226}Ra ejected into a pore space (or fracture) would quickly be adsorbed onto the iron (or manganese) hydroxides coating fracture and grain surfaces.

In reducing waters, iron (and manganese) hydroxide coatings are absent or are less abundant on mineral grains or along fractures. The coatings are unstable in reducing environments and tend to dissolve. When iron hydroxides dissolve, or are not present, fewer adsorption sites are available for radium; hence, radium concentration can increase in solution. Because radium-226 released to the ground water by alpha-recoil and other mechanisms has a lesser likelihood of being adsorbed onto iron (and manganese) hydroxides in reducing environments, it will have a greater likelihood of migrating along with the water in major fractures.

An additional factor limiting radium adsorption is the pH of the ground water. As the pH decreases below a value of 8, proportionately more samples of the ground water in the Newark basin have higher radium concentrations. Concentrations of radium in excess of 4 pCi/L are found only in ground-water samples in which the pH is less than 8 (fig. 106*B*). As the pH decreases, the point of zero charge for iron hydroxides is approached or reached, and radium is less likely to be adsorbed onto the iron hydroxides, even in oxidizing ground water where the iron hydroxides are stable.

In summary, radium is strongly adsorbed from ground water in the Newark basin by iron hydroxides, especially above pH values of 8. As the pH decreases to a value below this, the efficiency with which radium is adsorbed decreases. Furthermore, in reducing waters, iron hydroxides become solubilized (or do not precipitate in the first place), so the number of adsorption sites available for dissolved radium is further reduced.

Because the total amount of iron hydroxides in the rocks is relatively small compared to the abundance of other minerals, these hydroxides often are dismissed as unimportant. However, in fractured rocks, the fractures in the rock along which the water flows become coated with secondary minerals, including iron hydroxides. Therefore, even though the abundance of the iron hydroxides may be minor relative to other components in the bulk rock, the iron hydroxides are distributed in the rock in such a way (along the fractures) that their contact with the ground water is maximized. Ground-water flow in the Newark basin is along fractures in the rock (Herpers and Bardsdale, 1951); this flow pattern enables fracture-coating iron hydroxides to adsorb trace constituents (including ^{226}Ra) from the ground water.

Adsorption onto iron hydroxides is not the only control on ^{226}Ra concentration in ground waters of the Newark basin. The data of Ames and others (1983a,b) illustrated that secondary clay minerals, and even quartz, can adsorb some quantities of radium. During this study, the collected core samples were found to contain many fractures coated with clay minerals; this finding indicates that these sorbents also are abundant in fractures in the rocks of the Newark basin. The point of zero charge for most secondary clay minerals is generally below pH values of 6 (Stumm and Morgan, 1981). Therefore, adsorption onto these secondary clay minerals may also limit ^{226}Ra activities in the slightly alkaline ground water present in the Newark basin.

The sodium bicarbonate type of water is found in some areas in the black mudstone beds in the lower part of the Passaic Formation and in the Lockatong Formation. These waters are low in divalent cations (divalent cations may account for less than 5 percent of the total positive charge). The chemistry of these waters is controlled by cation exchange, where divalent cations are removed from solution by clay minerals and are replaced by monovalent sodium. These waters generally have low radium concentrations (less than 1 pCi/L). The concentration of ^{226}Ra, a divalent cation, also is limited in solution in these waters by the cation-exchange process.

Investigators have reported that ^{226}Ra activities increase with increasing dissolved-solids content in briny or brackish waters (Miller and Sutcliffe, 1985). However, we propose that ^{226}Ra concentrations decrease when dissolved solids content increases in ground waters in the Newark basin because of the increasing sulfate concentrations associated with increasing dissolved-solids content.

Radium-226 concentrations greater than 0.6 pCi/L are absent in ground water where sulfate concentration exceeds 100 mg/L, with the exception of one sample (fig. 107*B*). Because the sulfate-rich ground water is oxidizing, it may be argued that the absence of detectable ^{226}Ra is probably because iron hydroxides adsorb ^{226}Ra from solution. However, the efficiency of this adsorption may be somewhat reduced by competition from the abundant divalent Ca^{+2} ion (Ames and others, 1983b).

Radium is not abundant enough in nature to precipitate radium sulfate salts, even though radium sulfate is essentially insoluble (Langmuir and Riese, 1985). Saturation indices, calculated from the solubility-product constants given by Langmuir and Riese (1985), indicate that radium-sulfate precipitation cannot occur in ground waters in the Newark basin. The presence of any significant ^{226}Ra concentrations in sulfate-rich water may be limited by the precipitation of other sulfate minerals. Langmuir and Riese (1985) determined that radium can coprecipitate with barium sulfate (barite) and calcium sulfate (gypsum). Both barite and gypsum are reported to be present in the rocks of the Newark basin (Herpers and Barksdale, 1951; Dombroski, 1980). Thermodynamic calculations made with the computer program WATEQF (Plummer and others, 1976) indicate that the calcium magnesium sodium sulfate bicarbonate type waters in the Newark basin are supersaturated with respect to barite. Barium concentrations show a strong inverse correlation with the concentration of both sulfate and strontium, and the median concentration value of barium in the sulfate-rich waters (57 μg/L) is well below the median concentration value of barium in ground-water samples from the entire data set (table 46). Coprecipitation of radium with barite may be another mechanism that limits the activities of ^{226}Ra in the sulfate-rich ground water. This constraint in sulfate-rich water explains why water samples containing ^{226}Ra concentrations above 0.6 pCi/L are solely the reducing bicarbonate type of waters (with the exception of one sample).

In summary, uranium and radium concentrations in ground water in the Newark basin are determined not only by uranium abundance in the rock material but also by geochemical processes involving these elements. Uranium concentrations are controlled by (1) the solubility of the U^{+6} ion and (2) carbonate complexing and the lack of adsorption due to carbonate complexing. Radium-226 concentrations in the ground water of the Newark basin are constrained by the simultaneous occurrence of at least three important geochemical processes, which are adsorption onto iron and manganese hydroxides, ion exchange onto clay minerals, and precipitation as alkaline-earth sulfate minerals. All these reactions, in turn, may be limited by the pH and Eh of the ground water.

Relations Among Ground-Water Chemistry, Lithology, and Radionuclide Activities

The geographic distribution of uranium and ^{226}Ra in ground water depends upon the outcrop pattern of the different formations in the Newark basin, because the chemical characteristics (pH and Eh) and constituents differ somewhat from formation to formation. Ground water in the red siltstone of the Passaic Formation and the arkose of the Stockton Formation is generally oxidizing, whereas in the black organic mudstones of the lower part of the Passaic Formation and the Lockatong Formation it is generally reducing. Where the underlying rock is uranium rich, uranium is present in oxidizing waters (mostly the Passaic and Stockton Formations), and ^{226}Ra is present in reducing waters (mostly the lower part of the Passaic Formation and Lockatong Formation).

CONTROLS ON DISSOLVED RADON-222 CONCENTRATION IN THE NEWARK BASIN

Radon-222 concentration in the ground water is highly variable and is two to three orders of magnitude greater than the corresponding uranium and ^{226}Ra concentration (fig. 100). This finding is consistent with the conclusions of Tanner (1964b) that the dissolved ^{222}Rn concentration is too high to be derived from dissolved ^{226}Ra and must be derived from alpha-recoil (Tanner, 1964a,b) of ^{222}Rn from ^{226}Ra decay in the rock matrix. Because the half-life of ^{222}Rn is relatively short (3.82 days), it cannot migrate far from its source rock. Therefore, the dissolved ^{222}Rn concentration of a water sample from a well is likely to be proportional to the radioactivity in the rock around the well borehole, and not necessarily to the concentration of radionuclides in solution.

Radon-222 concentration in the ground water does not correlate with concentrations of dissolved uranium (fig. 100*A*), but ^{222}Rn concentration does correlate with the concentrations of dissolved ^{226}Ra (fig. 100*B*). In general, ground-water samples from the Newark basin that contained concentrations of ^{226}Ra greater than 1 pCi/L also contained ^{222}Rn concentrations greater than 2,000 pCi/L (fig. 100*B*), whereas all samples that contained concentrations of ^{226}Ra less than the detection limit of 0.6 pCi/L also contained ^{222}Rn concentrations less than 2,000 pCi/L. However, several wells containing abundant ^{226}Ra concentrations contain dissolved ^{222}Rn concentrations below the median value.

Uranium concentrations do not correlate with ^{222}Rn concentrations in ground water of the Newark basin, because uranium concentrations in the ground water are controlled by chemical reactions, and because uranium can migrate far from its source bed, as it has a long half-life.

Radium-226 concentrations correlate with ^{222}Rn concentrations in ground water of the Newark basin, while uranium concentrations do not, because in the reducing waters of the uranium-rich black mudstones in the Newark basin, ^{226}Ra can go into solution to some degree as can ^{222}Rn. In addition, ^{226}Ra has a much shorter half-life than uranium and, therefore, cannot migrate as far from its source bed as can uranium. However, the half-life of ^{226}Ra is still much longer than that of ^{222}Rn, so ^{226}Ra may migrate at least some distance from its source bed and not represent radioactivity in the rock around the well borehole. This ability to migrate may cause the abundant scatter on the scatter plot of ^{222}Rn and ^{226}Ra (fig. 100*B*). Additional scatter may be caused by the fact that ^{226}Ra does not go into solution in the oxidizing ground water of the Newark basin.

Dissolved ^{222}Rn concentration does not correlate with the concentration of any chemical constituent in the ground water in the Newark basin. This is not surprising in light of the fact that ^{222}Rn is an inert gas that is not chemically reactive. In the Newark basin, many uranium-enriched rock zones are found in black mudstones containing reducing ground water; this environment prohibits uranium from going into solution while allowing the chemically inert ^{222}Rn to go into solution. Therefore, the concentration of dissolved uranium in reducing environments does not represent the radioactivity in the rock around the well borehole. Conversely, in oxidizing ground water in the Newark basin (particularly in the sulfate-rich water), uranium is soluble, can migrate far from its source bed, and, therefore, may not represent the radioactivity in the rock around the well borehole.

Radon-222 concentrations in ground water in fractured-rock terrain are reported to be controlled by ^{226}Ra content of the local bedrock and by several physical characteristics, not by geochemical reactions involving radon. These characteristics include pumping rate, hydrodynamic dispersion, fracture length (Chen and Wilson, 1987), fracture width, and the ^{226}Ra content and radon emanation coefficient of the material in the radium-containing aquifer (Korner and Rose, 1977). These characteristics have not yet been evaluated in the fractured rocks of the Newark basin.

Applications to Indoor Airborne Radon

One of the most important factors in identifying areas of elevated activities of indoor airborne ^{222}Rn is locating areas of rock (soil) material containing anomalously high concentrations of uranium or ^{226}Ra (Otten, 1987). This study of the Newark basin has shown that areas of the Newark basin that have elevated radioactivity in ground water are areas that generally have underlying rock units enriched in uranium. In the lower part of the Passaic Formation, and in the Lockatong and Stockton Formations, anomalous activities of radionuclides are present in ground water, and anomalous amounts of radioactivity usually are found in the rock material intercepted by wells. Therefore, the outcrop areas of these formations may be areas that have the potential for indoor airborne ^{222}Rn contamination. Measurements of radioactivity in ground water and in the adjoining rock units in the Newark basin may prove valuable in the study of the distribution of indoor airborne ^{222}Rn concentrations (Thomas, 1987).

CONCLUSIONS

Elevated concentrations of naturally occurring radionuclides in ground water are associated with uranium enrichment in the rocks of the Newark basin in New Jersey. Elevated gross alpha-particle activities are found primarily in ground water along the southeastern border of the study area, in the upper part of the Lockatong Formation and lower part of the Passaic Formation of Olsen (1980a), and in the Hopewell and Flemington fault blocks, where these formations are repeated. Many wells located near the mapped stratigraphic contact of these formations had elevated alpha-particle activities.

The source of radioactivity was determined by borehole geophysical testing, analysis of lithologic cores, and autoradiography. Thin, laterally extensive, uranium-enriched black mudstones within the Lockatong Formation and lower part of the Passaic Formation are the primary source of natural radioactivity in ground water in the study area.

Elevated radionuclide activities in ground water also are found locally in wells open to uranium-enriched zones of arkosic sandstone of the Stockton Formation and in the Lockatong and lower part of the Passaic Formation in the northeastern part of the basin. Ground water from the upper part of the Passaic Formation, basalt and diabase units, and the undifferentiated sedimentary units interbedded with and overlying the basalt flows is predominantly low in radionuclides.

The distribution of ^{226}Ra and uranium concentrations in ground water of the Newark basin is controlled not only by uranium abundance in the rock but also by ground-water geochemistry. Elevated concentrations of uranium and ^{226}Ra generally are not found concurrently in the same ground water. Elevated concentrations of uranium are present in ground water that exhibits high gross alpha-particle activity and that is oxidizing, low in iron and manganese, and often high in sulfate and bicarbonate. In contrast, elevated concentrations of ^{226}Ra occur in ground water that has high gross alpha-particle activity and that is reducing, contains elevated concentrations of iron, manganese, and, locally, barium, and low concentrations of sulfate.

Uranium concentrations in ground water in the Newark basin are determined by two geochemical processes, the solubility of the U^{+6} ion and carbonate complexing. Radium-226 concentrations are determined by three important geochemical processes: (1) adsorption onto iron and manganese hydroxides, (2) adsorption and (or) ion exchange onto clay minerals, and (3) coprecipitation with alkaline-earth sulfate minerals. All these reactions are, in turn, governed by the pH and Eh of the ground water. The dissolved concentration of ^{222}Rn, an inert gas, is not constrained by the geochemical composition of the ground water. Radon-222 concentrations in ground water are limited by the ^{226}Ra content and the radon-emanation coefficient of the rock matrix, and by physical characteristics of the aquifers, such as ground-water flow conditions, and the length and width of the fractures in the rock.

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RADIUM-226, RADIUM-228, AND RADON-222 IN GROUND WATER OF THE CHICKIES QUARTZITE, SOUTHEASTERN PENNSYLVANIA

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Abstract

The Chickies Quartzite, a Cambrian quartzite, slate, and conglomerate, typically forms ridges and borders uplands on a 112-square-mile area of the Piedmont physiographic province of southeastern Pennsylvania. The Chickies is a minor water-table aquifer recharged primarily by precipitation.

Radium-226 concentrations for ground water from 107 wells range from less than 0.1 to 31 picocuries per liter, and radium-228 concentrations range from less than 0.5 to 100 picocuries per liter. Fifty-one of the 107 analyses exceeded the Environmental Protection Agency maximum contaminant level for radium of 5 picocuries per liter and had radium-228 concentrations in excess of those for radium-226. Elevated radium concentrations in ground water are distributed throughout the Chickies Quartzite. The range of radium-228/radium-226 activity ratios for 70 analyses is 0.7 to 30, and the median is 2.7. Radon-222 concentrations range from 86 to 24,500 picocuries per liter.

Total dissolved-solids concentration of the ground water is low (as low as 7 milligrams per liter), and pH is acidic (as low as 4.3). The ground waters are slightly reducing with respect to iron and manganese and contain detectable dissolved concentrations of these constituents.

Nonparametric Spearman rank correlation was used to determine the relation between concentrations of radionuclides, other chemical constituents, and well construction characteristics. Total radium (sum of dissolved radium-226 and dissolved radium-228) relations that were found to be significant at the 95-percent confidence level are a negative correlation with pH and a positive correlation with dissolved organic carbon, barium, and manganese. Total radium concentrations also showed a slight negative correlation with well yield and a slight positive correlation with specific conductance, sodium, chloride, and sulfate. Radon-222 relations found to be significant are slightly negative correlations with sulfate, magnesium, total dissolved solids, and specific conductance.

Low pH may be used as a predictor of elevated radium concentrations in ground waters of the Chickies Quartzite. At pH below 4.7, all samples of ground water contained concentrations of total radium that exceeded the Environmental Protection Agency maximum contaminant level of 5 picocuries per liter.

Possible controls on radium in solution in ground water of the Chickies Quartzite are adsorption and complexation. The low pH of the ground water decreases the ability of quartz and kaolinite in the aquifer matrix to serve as adsorption sites for radium. The dissolution of iron and manganese oxyhydroxides, possible adsorption sites, decreases the number of available adsorption sites and may release the radium into solution. Also, complexation with dissolved organic carbon may increase radium mobility.

Lack of correlation between concentrations of radon-222 and its parent, radium-226, suggests that radon-222 is not supported by radium-226 in solution. Concentrations of radon-222 in solution differ locally.

INTRODUCTION

The Chickies Quartzite is a Cambrian quartzite, slate, and conglomerate that crops out in the Piedmont physiographic province of southeastern Pennsylvania, generally forming narrow ridges (fig. 108). The Chickies Quartzite occurs in parts of Adams, Berks, Bucks, Chester, Lancaster, Montgomery, and York Counties. Although the Chickies is a minor aquifer, it is the only source of water for thousands of people served by private wells.

In 1985, routine sampling by the Pennsylvania Department of Environmental Resources (PaDER) at three trailer parks that derive their water supply from the Chickies Quartzite showed gross alpha-particle radioactivity, excluding uranium and radon, above the Environmental Protection Agency (EPA) 15 pCi/L (picocuries per liter) maximum contaminant level (MCL) (EPA, 1986). Subsequent sampling in 1985 showed total radium concentrations (sum of dissolved radium-226 and radium-228) that exceeded the MCL of 5 pCi/L for radium.

A study of the occurrence and distribution of radium and radon in ground water of the Chickies Quartzite in southeastern Pennsylvania was conducted by the U.S. Geological Survey in cooperation with the PaDER Bureau of Topographic and Geologic Survey and the PaDER Bureau of Radiation Protection from October 1986 through December 1987. An understanding of the conditions and processes that favor the dissolution and retention of radium and radon in solution may help predict concentrations in the Chickies Quartzite and similar geologic units.

Purpose and Scope

This paper identifies the extent and magnitude of radium and radon concentrations in ground water in the Chickies Quartzite in southeastern Pennsylvania and briefly characterizes the geochemical environment associated with elevated radium and radon concentrations. Inductive statistics are used to determine correlations between radionuclide concentrations, other chemical constituents, and well construction characteristics.

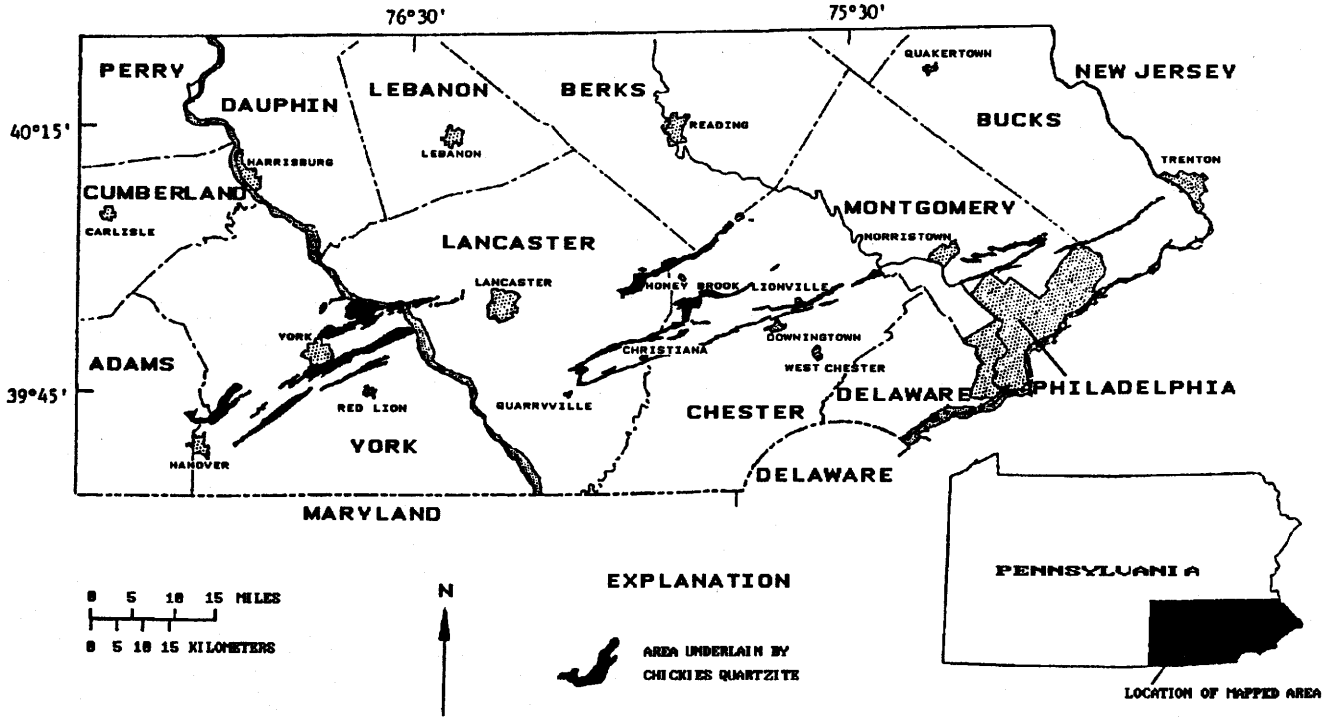

Figure 108. Location of the Chickies Quartzite in southeastern Pennsylvania. Base by the Pennsylvania Geological Survey.

Phase one of the sampling program, a reconnaissance to determine the extent and magnitude of radium and radon concentrations, was conducted between October 1986 and January 1987. The sampling program was based on available data and adjusted as new data became available. Fifty-one water samples were collected to supplement available radionuclide and chemical data and provide evenly distributed areal coverage in the six counties where the Chickies occurs. Data collection was concentrated in Chester, Lancaster, and York Counties; a few sites were sampled in Berks, Bucks, and Montgomery Counties. During phase two of the sampling program (March to October 1987), an additional 56 samples were collected for chemical analyses. Sampling sites were distributed throughout the Chickies, but sampling was concentrated in several smaller areas to characterize the geochemical and hydrologic conditions indicative of elevated radium and radon concentrations.

Acknowledgments

The authors are indebted to the many individuals who provided assistance and information essential for the successful completion of this study. Special thanks are given to Robert C. Smith II of the PaDER Bureau of Topographic and Geologic Survey, Margaret A. Reilly of the PaDER Bureau of Radiation Protection, and Arthur W. Rose of the Pennsylvania State University for their many suggestions for this paper. Sincere thanks are given to Michael Webb of the PaDER Bureau of Laboratories for his assistance in quality-assurance analyses. The authors gratefully acknowledge the cooperation of the many individuals who kindly permitted access to their wells for the collection of data essential to this study.

GEOHYDROLOGIC SETTING

Geology

The Chickies Quartzite is Early Cambrian in age and lies unconformably on Precambrian deformed and metamorphosed crystalline rocks. The Lower Cambrian Antietam Quartzite and underlying Harpers Phyllite—undivided quartzite, quartz schist, and phyllite units—lie conformably above the Chickies Quartzite. The Chickies Quartzite has three mapped members: the upper quartzite, upper slate, and lower Hellam Member, a conglomerate. The upper slate member has been mapped in York County (Stose and Stose, 1944), and both other members have been mapped in all counties underlain by the formation. Overall thickness of the Chickies Quartzite is reported to range from 430 to 1,300 ft (feet) (Lyttle and Epstein, 1987); the Hellam Member ranges from 200 to 500 ft in thickness (Bascom and Stose, 1938; Stose and Jonas, 1939; Willard and others, 1959). Thickness of the upper slate member is estimated to be 350 ft (Stose and Stose, 1944). The Chickies Quartzite has been folded and faulted repeatedly and, where exposed, forms narrow ridges that trend generally east-northeast. The formation is highly resistant to weathering.

The quartzite is light gray to white, vitreous to granular, and massive to thin bedded, with sericitic quartz schist (Bascom and Stose, 1938). In general, the quartzite is very clean, with small amounts of feldspar, has undergone some recrystallization, and may contain nondetrital tourmaline (commonly stretched) and mica. Other minerals present as accessories include zircon, rutile, apatite, monazite, limonite after pyrite, hematite, magnetite, and unidentified opaque minerals (Bascom, 1905; Bascom and Stose, 1938; Dryden and Dryden, 1964; Berkheiser, 1985). Thin, discontinuous beds of "white siliceous clay" or kaolinite in the quartzite have been interpreted as weathered mica-rich feldspars layers (Bascom and Stose, 1938). The iron content of the quartzite is reported to increase to the west from Lancaster County (Berkheiser, 1985). The basal Hellam Member is a coarse feldspar and quartz-pebble conglomerate that grades into conglomeratic quartzite having a quartz and sericitic matrix and accessory zircon, magnetite, and hematite (Knopf and Jonas, 1929; Bascom and Stose, 1938). Quartz pebbles in the conglomerate are commonly blue. Interbeds of slate are reported to occur in both members. The upper slate member is interbedded slate, phyllite, and quartzite; the member contains quartz, albite, chlorite, muscovite, magnetite, zircon, and tourmaline (Stose and Stose, 1944).

Because zircon, monazite, and apatite are known to contain minor amounts of uranium and thorium, these minerals are possible sources of radionuclides within the formation. Generally, zircon may contain up to 13.1 percent thorium and 2.7 percent uranium, monazite may contain up to 26.5 percent thorium and up to 0.1 percent uranium, and apatite may contain up to 3 percent thorium and 0.02 percent uranium (Frondel and others, 1967; Durrance, 1986). Detrital zircons from the Devonian Catskill Formation in east-central Pennsylvania are reported to contain up to about 0.17 percent thorium and 1.2 percent uranium (Tole, 1979).

Goodwin and Anderson (1974) described the environment of deposition of the quartzite as a mosaic of subtidal channels, intertidal flats, and tidal flat ponds. They base this interpretation on identifiable physical and biogenic structures within the formation, such as stratification, bed form, grain size, and fossil burrow forms of *Scolithus* and *Monicraterion*.

Aquifer Characteristics

The Chickies Quartzite is a water-table aquifer. Springs and artesian conditions occur locally on slopes. Recharge to the aquifer is predominately by local precipitation. Soil cover is thin (1 to 5 ft), and depth to consolidated bedrock commonly is 10 ft or less, although some weathered zones may extend deeper. Beds dip steeply in places. Openings to the surface have developed along bedding, jointing, and structurally controlled planes such as cleavage, faults, and fractures caused by folding. This secondary porosity is estimated to be very slight to moderate (McGlade and others, 1972; Wilshusen, 1979). In very well cemented sandstones and recrystallized quartzites, porosities may be less than 1 percent (Freeze and Cherry, 1979).

Well Yield, Well Depth, and Specific Capacity

Reported yields for 167 domestic wells ranged from 0.5 to 73 gal/min (gallons per minute); the median yield was 9.0 gal/min. Reported yields for 37 nondomestic wells ranged from 3 to 125 gal/min; the median yield was 23.5 gal/min. Some of the difference between the median yield for domestic and nondomestic wells may be attributable to the generally larger (greater than 6 in (inches)) diameter of nondomestic wells. Also, nondomestic wells are drilled for the largest yield obtainable and are generally deeper than domestic wells. Well depths for 196 domestic wells ranged from 16 to 500 ft; the median depth was 140 ft. Well depths for 48 nondomestic wells ranged from 42 to 600 ft; the median depth was 155 ft.

Specific capacities of 76 domestic wells ranged from less than 0.01 to 5.0 (gal/min)/ft (gallons per minute per foot of drawdown); the median was 0.2 (gal/min)/ft. Specific capacities are less than 1.0 (gal/min)/ft for 87 percent of the domestic wells. Specific capacities for 21 nondomestic wells ranged from 0.2 to 3.0 (gal/min)/ft, and the median was 0.25 (gal/min)/ft. Specific capacities are less than 1.0 (gal/min)/ft in 81 percent of the nondomestic wells.

DATA COLLECTION AND ANALYSIS

Data for this study consist of chemical analyses of ground-water samples and physical characteristics of the wells. A total of 107 wells completed in the Chickies Quartzite was sampled from November 1986 through October 1987. Several wells were sampled more than once, but only the results of the initial sampling are reported and included in data analysis. Temperature, pH, alkalinity, specific conductance, and dissolved oxygen of ground-water samples were measured in the field at each sampling site. Water samples were analyzed in the laboratory for dissolved radionuclides, inorganic constituents, and dissolved organic carbon. Information about well construction and characteristics was obtained from driller records, if available, from owners, and from the U.S. Geological Survey data base.

Sampling Sites

Of the 107 sampling sites, 101 were domestic wells and 6 were nondomestic wells. Generally, these wells were drilled by the air-rotary or air-hammer method, cased with steel or plastic casing set 20 to 40 ft into bedrock, and finished as open holes. Depth to consolidated rock from land surface is usually less than 30 ft. Most domestic wells have a 6-in diameter and are equipped with submersible pumps. The nondomestic wells range in diameter from 6 to 8 in. Driller records indicate that more than one fracture or water-bearing zone contributes water to some of the wells.

Information from driller logs, geologic maps, and field water chemistry was used to ensure that sampled wells were completed in the Chickies Quartzite. However, it is possible that some wells near contacts penetrate other formations.

Sample Collection

Wells were pumped until water temperature stabilized (about 30 to 60 minutes). Water samples were collected as close to the well head as possible, ahead of filters or treatment equipment. Water for analysis of dissolved radionuclides and inorganic constituents was filtered through a 0.45-μm (micrometer) filter by use of a peristaltic pump. The radiochemical samples were preserved in the field by treatment with concentrated hydrochloric or nitric acid. For radon- 222 (^{222}Rn) analysis by liquid scintillation, two 10-mL (milliliter) aliquots per sample site were collected by the method outlined by the EPA (1978).

Field Analysis

Field measurements of pH, temperature, specific conductance, alkalinity, and dissolved oxygen were obtained by use of methods described by Wood (1981). Total alkalinity is assumed to be the sum of bicarbonate and carbonate, and samples were titrated to pH 4.5. If the pH of a water sample was below 4.5, alkalinity is reported as 0.0 mg/L (milligrams per liter) as $CaCO_3$. The presence of organic acids or other titratable anions not identified in the sample may interfere with the alkalinity titration by lowering the endpoint. Other errors in determination of alkalinity may be caused by inaccurate pH measurements of the sample.

Dissolved oxygen was measured by the azide modification of the Winkler method (American Public Health Association and Water Pollution Control Federation, 1985). Reported values for dissolved oxygen may be elevated and may not reflect actual ground-water concentrations because oxygenation may have occurred during pumping or sampling. Possible interference by ferrous iron (greater than 1,000 μg/L (micrograms per liter)) would cause a low value for dissolved oxygen that had been determined by the azide modification of the Winkler method. Dissolved-oxygen measurements were made at 37 sites sampled during the first phase of the project. Both a membrane polarographic cell probe and the Winkler method were used. Air was used to calibrate the membrane polarographic cell probe. Comparison of the two methods shows that the Winkler method generally measured greater values for dissolved oxygen (the range of difference for the Winkler and membrane methods combined is -1.2 to $+3.6$ mg/L, the median is $+0.6$ mg/L, and the mean is $+0.8$ mg/L). Dissolved-oxygen measurements from independent work in 1985 (Cecil and others, 1987) confirm that the values for dissolved oxygen in ground water of the Chickies Quartzite are elevated.

Eh was measured during phase one sampling as a semiquantitative indicator of redox potential. For 48 sites, Eh ranged from $+210$ mv (millivolts) to $+660$ mv, and the median was $+430$ mv. Given the observed range of pH, the Eh measurements, which agree with previous work from 1985 (Cecil and others, 1987), indicate that the ground water is slightly reducing for iron and manganese. Dissolved ferrous and total iron were field determined at a few sites during phase two by use of a spectrophotometric method (American Public Health Association and Water Pollution Control Federation, 1985). The field determinations indicate that the ferrous form is predominant; this determination is consistent with observed Eh and pH values.

Generally, ground water from the Chickies Quartzite is acidic, exhibits low alkalinity and specific conductance, and apparently has elevated concentrations of dissolved oxygen. Median and ranges of field values are presented in table 47. The highest value of pH (7.3) and second highest specific conductance (626 μS/cm (microsiemens per centimeter at 25 °C)) are for a water sample from a well that probably penetrates the Ledger Dolomite and represents mixed waters.

Radionuclides

Analysis of water samples for radionuclides was contracted to private laboratories. Concentrations of dissolved radium-226 (^{226}Ra), radium-228 (^{228}Ra), total uranium, and gross alpha- and beta-particle radioactivities were measured by Teledyne Isotopes, Westwood, N.J. Concentrations of ^{226}Ra were determined by radon emanation, ^{228}Ra by coprecipitation with barium sulfate, and beta-gamma counting and total uranium by laser fluorometry. Lower limits of detection for analytical methods are reported as less-than (<) values and differ with background count of the laboratory at the time of analysis. All values less than the lower limit of detection were set equal to the same rank for use in nonparametric statistical analysis

Table 47. Field measurements of temperature, pH, specific conductance, alkalinity, and dissolved oxygen for ground water from the Chickies Quartzite

[°C, degrees Celsius; µS/cm, microsiemens per centimeter at 25 °C; mg/L, milligrams per liter]

Physical property or constituent	Number of measurements	Range		Median
		Minimum	Maximum	
Temperature (°C)	107	9	15	12.0
pH (pH units)	107	4.3	7.3	5.3
Specific conductance (µS/cm)	107	115	663	137
Alkalinity (mg/L as $CaCO_3$)	95	.0	296	22.4
Dissolved oxygen (mg/L)	102	.0	12.1	6.7

Table 48. Laboratory determinations for dissolved concentrations of radium-226, radium-228, total uranium, and radon-222, and gross alpha-particle and gross beta-particle radioactivity

[pCi/L, picocuries per liter; µg/L, micrograms per liter]

Constituent	Number of samples	Number of samples below detection limits	Range		Median
			Minimum	Maximum	
Radium-226 (pCi/L)	107	19	<0.1	31±2	1.0±0.4
Radium-228 (pCi/L)	107	31	<.5	100±10	2.7±.5
Uranium, total (µg/L)	107	29	<.05	6.6±1.0	.2±.03
Radon-222 (pCi/L)	103	0	86	24,500	2,000
Gross alpha-particle radioactivity (pCi/L)	107	29	<.7	55±6	3.5±1.1
Gross beta-particle radioactivity (pCi/L as cesium-137)	107	1	<1.0	210±10	6.7±1.0

Table 49. Distribution by county of total radium and radon-222 in samples of ground water in the Chickies Quartzite

[pCi/L, picocuries per liter]

County	Total radium[1] (pCi/L)					Radon-222 (pCi/L)			
	Number of samples	Number (percent) exceeding 5 pCi/L	Range		Median	Number of samples	Range		Median
			Minimum	Maximum			Minimum	Maximum	
Berks	1	0 (0)	3.2±0.7			1	114		
Bucks	5	2 (40)	<.8	54±2.1	<1.4	5	760	3,530	1,630
Chester	57	34 (60)	<.6	86±6.3	6.5±.4	53	228	9,270	2,070
Lancaster	18	10 (56)	<.8	107±10	6.6±.6	18	475	24,500	1,950
Montgomery	4	0 (0)	<1.0	1.5±.4	<1.2	4	86	2,680	500
York	22	5 (23)	<.7	14±1.1	<1.4	22	180	20,320	2,300
Total	107	51 (48)	<.8	107±10	4.1	103	86	24,500	2,000

[1] Sum of dissolved radium-226 and radium-228.

(D.R. Helsel, U.S. Geological Survey, oral commun., 1988). Less-than values were set equal to the method detection limit (for example, <0.1=0.1) for use in plots. Less-than values were excluded from ^{228}Ra/^{226}Ra activity-ratio calculations. Radon-222 (^{222}Rn) measurements by the liquid scintillation method were made at the Physics Department, University of Maine, Orono, Maine. Reported ^{222}Rn values are the mean of two samples collected at each site. A summary of the results of the radiochemical analyses is listed in table 48.

Results of total radium and ^{222}Rn sampling by county are presented in table 49. All counties except Berks (1 sample) and Montgomery (4 samples) have wells in which the water exceeds the EPA MCL of 5 pCi/L total radium. Wells in Chester and Lancaster Counties, where much of the formation crops out, show the greatest numbers and percentage of samples above the EPA MCL. Elevated radium concentrations in ground water appear to be distributed throughout the formation. However, some areas of the Chickies Quartzite within each county seem more likely to have elevated radium concentrations in the ground water. This suggests that local lithology and hydrology may be controlling factors.

Radon-222 concentrations differ throughout the formation and appear to be site specific with no regional trend. For example, two wells that are approximately 300 ft

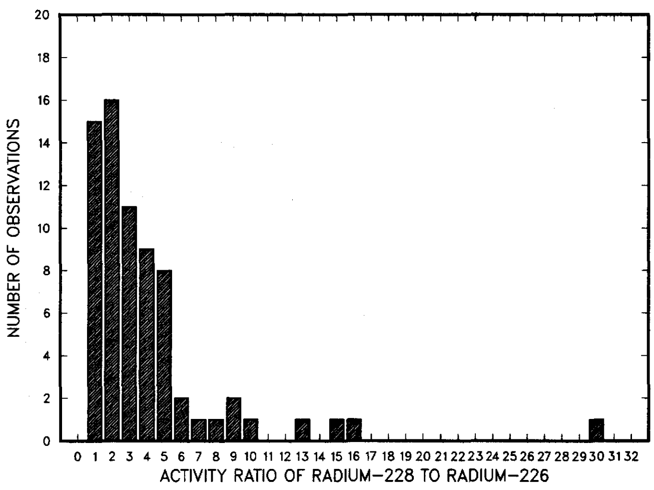

Figure 109. Activity ratios of radium-228 to radium-226 for 70 ground-water samples from the Chickies Quartzite. The range of ratios is 0.75 to 30. The median of ratios is 2.7.

apart and have similar depths and similar hydrologic settings were sampled at the same time, and the mean ^{222}Rn concentrations were 5,000 and 28,400 pCi/L. The larger ^{222}Rn concentration (28,400 pCi/L) was obtained during the second sampling of the well. The concentration of the first sample (24,500 pCi/L) was used in the data analysis.

In 97 samples, ^{228}Ra concentrations exceeded those of ^{226}Ra, and in the 10 samples where the ^{228}Ra concentration was less than or equal to that of ^{226}Ra, total radium concentrations were less than 5 pCi/L, and ^{228}Ra concentrations were at or near the method detection limit. For the 51 wells having total radium concentrations greater than 5 pCi/L, all ^{228}Ra concentrations were greater than those of ^{226}Ra. Because ^{228}Ra is a beta emitter, the screening of gross alpha-particle radioactivity for drinking water that has a potential for high radium values may need to be used with caution.

Activity ratios of ^{228}Ra to ^{226}Ra were calculated for a subset of samples. Analytical determinations reported as less than the method detection limit were excluded from the calculations. Seventy analyses for which both ^{226}Ra and ^{228}Ra concentrations were above the method detection limits were used to calculate radium-isotope ratios. The activity ratios are reported only as a semiquantitative indicator of the apparently unusual ^{228}Ra to ^{226}Ra activity ratios observed in the ground water of the Chickies Quartzite.

The range of ^{228}Ra/^{226}Ra activity ratios is 0.75 to 30 (fig. 109), and the median is 2.7. The abundance of the parent radionuclides, thorium-232 (^{232}Th) and uranium-238 (^{238}U), in the rocks of the Chickies Quartzite is not known. Mean crustal abundances are about 8.5 ppm (parts per million) thorium and 2.7 ppm uranium (Durrance, 1986). For an average sandstone, the thorium content is estimated to be 1.7 ppm, and the uranium content is estimated to be 0.45 ppm (Durrance, 1986). The activity ratio of ^{232}Th to ^{238}U may be estimated to be about 1.1 in the solid phase for an average sandstone or rocks that have mean crustal abundances of thorium and uranium. The expected ^{228}Ra/^{226}Ra activity ratio for water in contact with the rock should be close to unity if dissolution reactions and rates for the two radium isotopes are similar. Although a number of analyses fall near unity, 80 percent exceed 1.5. It is suggested that the abundance of ^{232}Th relative to ^{238}U may be greater in the Chickies Quartzite than for an average sandstone. For example, an average thorium content of orthoquartzites is about 12 ppm (Durrance, 1986). The Hardyston Quartzite, also Early Cambrian in age and of similar lithology to the Chickies Quartzite, but cropping out to the north, contains a basal layer enriched in thorium and uranium. Three analyses of rock samples from the Hardyston yielded high thorium/uranium weight ratios (R.C. Smith II, Pennsylvania Department of Environmental Resources, Bureau of Topographic and Geologic Survey, oral commun., 1988), which are approximately equivalent to ^{232}Th/^{238}U activity ratios of 2.5 or greater. Therefore, differences in the observed ratios of ^{228}Ra to ^{226}Ra in the ground water of the Chickies Quartzite may reflect differences in composition of the aquifer material. Initial aquifer composition may have changed through preferential leaching of ^{238}U or ^{234}U at some time, and this leaching would account for relative depletion of ^{226}Ra with respect to ^{228}Ra in the ground water.

Inorganic Constituents and Dissolved Organic Carbon

Chemical analyses of water samples for inorganic constituents other than radionuclides and for dissolved organic carbon (DOC) were performed at the U.S. Geological Survey National Center. Water samples for determination of major and minor ions and dissolved organic carbon were collected at all sites, and a summary of available results is listed in table 50. The chemical data are not normally distributed and are positively skewed.

Results of chemical analyses indicate that ground water in the Chickies Quartzite is soft (Durfor and Becker, 1964), relatively dilute, and low in dissolved solids. More than 80 percent of samples have a hardness of less than 60 mg/L as $CaCO_3$. The sum of major cation milliequivalents ranges from 0.1 to 7.0 meq/L (milliequivalents per liter), and the median is 0.9 meq/L. Sodium is the dominant cation in some water samples, and sodium concentrations vary directly with chloride concentrations. In other samples, calcium and magnesium dominate. Dominant anions are bicarbonate and chloride, although in a few samples sulfate

Table 50. Determinations of inorganic constituents and dissolved organic carbon in ground water in the Chickies Quartzite

[mg/L, milligrams per liter; µg/L, micrograms per liter; —, no samples below detection limits]

Constituent	Number of samples	Number of samples below detection limits	Range		Median
			Minimum	Maximum	
	Major ions (mg/L)				
Calcium	92	—	0.1	59	4.4
Magnesium	92	—	.2	38	3.3
Sodium	92	—	.7	59	4.2
Potassium	92	—	.3	11	1.9
Chloride	93	—	.3	83	8.2
Sulfate	93	1	<.2	62	6.2
Fluoride	93	43	<.1	.5	.1
Silica	93	—	3.4	28	8.8
	Minor ions (µg/L)				
Barium	92	4	2.0	380	66.5
Manganese	92	8	1.0	870	33.5
Iron, dissolved	92	6	3.0	27,000	35.5
	Other constituents				
Iron, total (µg/L)	91	7	<10.0	51,000	230
Total dissolved solids (mg/L)	93	—	7.0	343	68
Dissolved organic carbon (mg/L)	89	—	.5	10.0	.8

is dominant. Associations between ions differ; sulfate and bicarbonate show a tendency to pair with magnesium or calcium.

The laboratory and field data suggest that recharge to the ground water is local precipitation that has enriched sodium and chloride concentrations that are due to evapotranspiration and, possibly, contamination with road salt. Chemical analysis of precipitation collected since 1979 at a rain gage in central Chester County shows sodium, calcium, and magnesium concentrations of up to 1 mg/L, chloride concentrations of up to 1 mg/L, and sulfate concentrations of up to 10 mg/L, with pH as low as 3.6. The relatively low concentrations of calcium and magnesium, and low pH observed in the ground water, may be accounted for by the sparsity of easily dissolved or weathered feldspar and other silicates in the formation. The presence of dissolved iron and sulfate in the ground water may reflect oxidation of pyrite, a mineral that is known to occur in the Chickies Quartzite. Such a reaction would be favored in the acidic and oxygen-rich geochemical environment observed in the ground water. Relatively elevated concentrations of dissolved and total iron and dissolved manganese were measured in a few samples.

Thirty percent of the samples had dissolved organic carbon concentrations greater than or equal to 1 mg/L. Median dissolved organic carbon concentrations for crystalline rock aquifers are reported as 0.5 mg/L, and for sandstone, limestone, and gravel aquifers as 0.7 mg/L (Thurman, 1985). Three samples had elevated (greater than 5.0 mg/L) concentrations of dissolved organic carbon for ground water; the well that had the highest concentrations of dissolved organic carbon in water was resampled later, and the ground water was found to be contaminated with manmade organic compounds. Naturally elevated dissolved organic carbon values may indicate short residence time or rapid transport of organic-rich recharge waters.

RADIUM AND RADON IN GROUND WATER

To evaluate the physical and chemical factors that may affect the source and mobility of radionuclides in the Chickies Quartzite, the nonparametric Spearman rank correlation test was used to determine the correlation between concentrations of radionuclides and well depth, well yield, and concentrations of other chemical constituents. The correlation coefficient, Spearman's rho (r_s), measures monotonic trends, which may not be linear. Total radium and ^{222}Rn concentrations were tested for correlation with well depth, well yield, pH, specific conductance, temperature, concentrations of alkalinity, dissolved organic carbon, calcium, magnesium, sodium, potassium, chloride, sulfate, silica, barium, total iron, dissolved iron, manganese, uranium, total dissolved solids, and dissolved oxygen. Because of the EPA MCL of 5 pCi/L for combined ^{226}Ra and ^{228}Ra, correlations were calculated for total radium. Radon-222 was tested for correlation with its parent, ^{226}Ra.

A positive or negative correlation between the physical property or chemical constituent and radionuclide concentration may be used to help predict anomalous radionuclide concentrations in ground water of the Chickies Quartzite. Because a significant correlation coefficient (positive or negative) does not necessarily imply a cause-

Table 51. Spearman rank correlation for chemical constituents and well yield with total radium (sum of dissolved radium-226 and radium-228) and radon-222 significant at the 95-percent confidence level

Chemical constituent or physical property	Correlation coefficient (Spearman's rho)	p value
Total radium		
Dissolved organic carbon	0.68	$<1\times10^{-6}$
pH	−.66	$<1\times10^{-6}$
Silica	−.57	$<1\times10^{-6}$
Alkalinity	−.50	$<1\times10^{-6}$
Barium	.46	4×10^{-6}
Manganese	.35	.000632
Chloride	.26	.013651
Specific conductance	.24	.012464
Sodium	.24	.0229
Sulfate	.23	.027777
Well yield	−.23	.047777
Radon-222		
Sulfate	−.25	.016846
Magnesium	−.23	.027426
Total dissolved solids	−.23	.030419
Specific conductance	−.20	.04667

and-effect relation, the correlations were evaluated in terms of possible geochemical reactions or physical processes that may control the concentration and distribution of radionuclides. The Spearman rank correlations significant at the 95-percent level of confidence are listed in table 51. If the p value (the smallest level of significance that allows the null hypothesis to be rejected) for the Spearman rank correlation test is less than or equal to 0.05 (the significance level), then the null hypothesis that the two variables are independent is rejected.

Radium

A negative correlation between total radium concentration and pH in ground water of the Chickies Quartzite was first noted by Cecil and others (1987). Figure 110 is a scatter plot of total radium concentration and pH for 106 wells in the Chickies Quartzite. Figure 110 shows that all samples having a pH lower than 4.7 equal or exceed the MCL of 5 pCi/L for total radium. No water samples having a pH greater than 6.5 exceed the MCL for total radium.

Low pH may reduce the adsorption of radium by mineral surfaces and thereby increase radium concentrations in the water. Experiments conducted by Riese (1982) showed that adsorption of radium by quartz and kaolinite is inhibited by low pH. The data presented in figure 110 suggest that radium is not being adsorbed or otherwise immobilized in the low-pH water in the Chickies Quartzite.

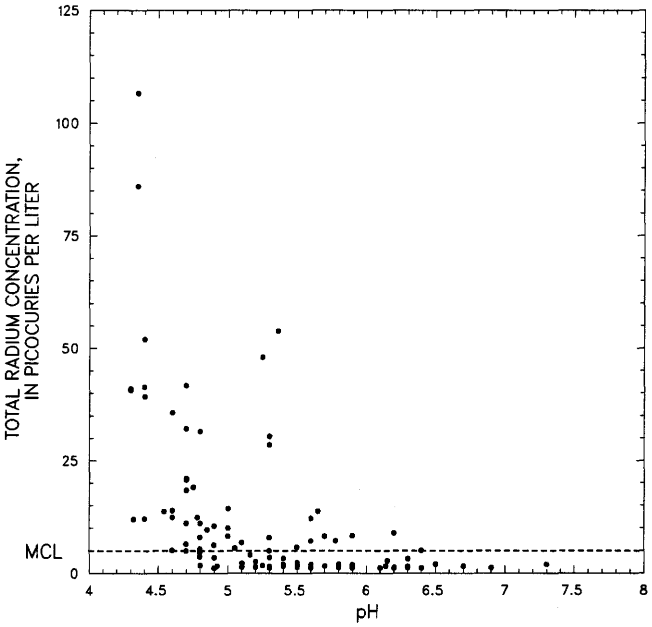

Figure 110. Relation between total radium concentration and pH for 106 wells in the Chickies Quartzite. MCL, the Environmental Protection Agency maximum contaminant level.

A scatter plot of total radium concentration as a function of manganese concentration (fig. 111) shows a positive correlation ($r_s = 0.35$). Because radium is strongly adsorbed by manganese oxyhydroxides, the dissolution of manganese oxyhydroxides would result in the release of adsorbed Ra^{+2}. Eh and pH are important controls on manganese dissolution; however, the relation (correlation coefficient) between dissolved manganese concentration and pH was not statistically significant.

A scatter plot of total radium concentration as a function of barium concentration (fig. 112) depicts a positive correlation (correlation coefficient, $r_s = 0.46$). This positive correlation may reflect the similar chemical behavior of barium and radium and also may reflect the dissolution of manganese oxyhydroxides. Manganese oxyhydroxides act as effective scavengers of barium and radium (Hem, 1985). Therefore, chemical environments favoring the dissolution of manganese oxyhydroxides may result in elevated concentrations of barium and radium in solution. This factor is supported by the correlation coefficient, $r_s = 0.34$, between manganese and barium that is significant at the 95-percent level of confidence.

The positive correlation between total radium concentration and dissolved organic carbon shown in figure 113 and supported by a correlation coefficient of 0.68 suggests the possible complexation of radium by dissolved organic carbon. Thurman (1985) states "complexation of metal ions by organic matter is an important organic-inorganic process." Dissolved organic carbon has a cation-exchange capacity of about 1 microequivalent per milligram (Thurman, 1985). Given the low concentrations of cations in ground water of the Chickies, some exchange sites on dissolved organic carbon may be available for radium. The

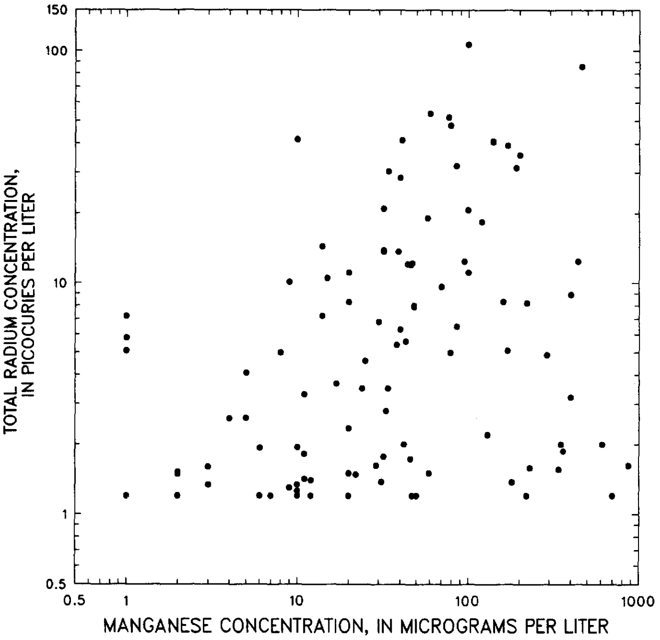

Figure 111. Relation between total radium and manganese concentrations for 92 wells in the Chickies Quartzite.

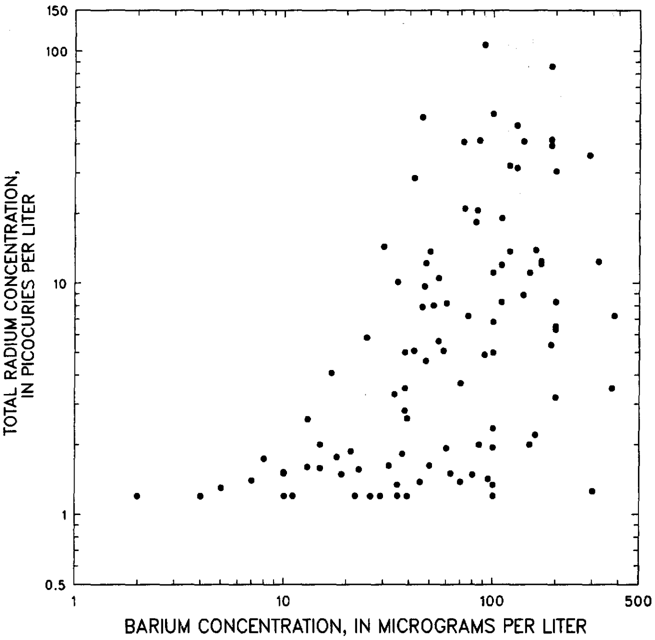

Figure 112. Relation between total radium and barium concentrations for 92 wells in the Chickies Quartzite.

relation between DOC and radium also may be coincidental, reflecting greater mobility of DOC and radium in low pH waters.

Total radium shows a negative correlation with alkalinity and silica (table 51). Alkalinity and silica show positive correlations with pH (r_s = 0.89 and r_s = 0.53, respectively). These correlations are significant at the 95-percent confidence level. However, ^{226}Ra and ^{228}Ra concentrations have a negative correlation with pH, and this factor may explain the negative correlations between radium and alkalinity and radium and silica. Other physical properties or chemical constituents that show a slight positive correlation with total radium are specific conductance, sodium, chloride, and sulfate (table 51). This suggests that radium solubility increases with greater ionic concentrations. Total radium shows a slight negative correlation with well yield.

Radon

The data-point scatter in figure 114 indicates the lack of a linear relation between concentrations of ^{222}Rn and its parent, ^{226}Ra, in solution. Low total uranium concentrations[1] (less than 0.05 to 4.49 pCi/L) and low ^{226}Ra concentrations (fig. 114) indicate that ^{222}Rn is unsupported in ground water of the Chickies Quartzite. The apparent lack of correlation between ^{222}Rn and ^{226}Ra concentrations in ground water has been reported by other investigators (Tanner, 1964; Snihs, 1972; Lee and others, 1979). The variability of ^{222}Rn concentrations in ground

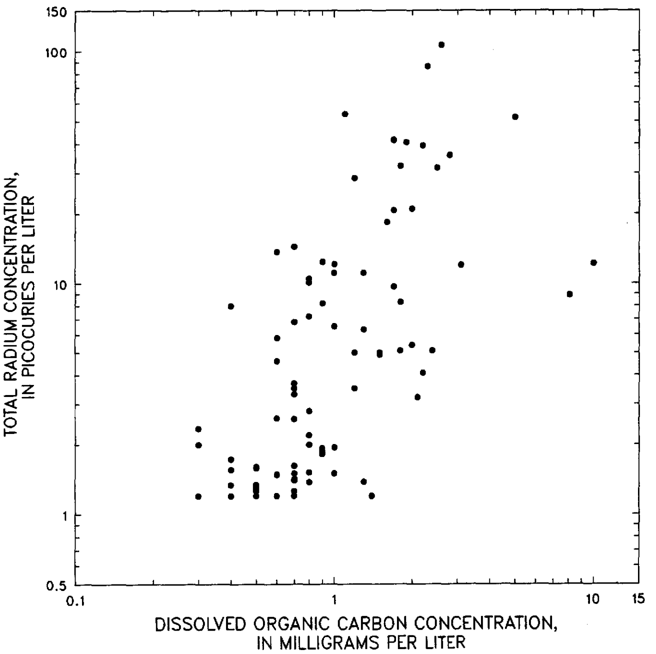

Figure 113. Relation between total radium and dissolved organic carbon concentrations for 89 wells in the Chickies Quartzite.

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[1] Uranium-isotope ratios are assumed to equal natural crustal abundances. The measured concentrations in micrograms per liter are multiplied by 0.68 pCi/μg to convert uranium concentrations to picocuries per liter.

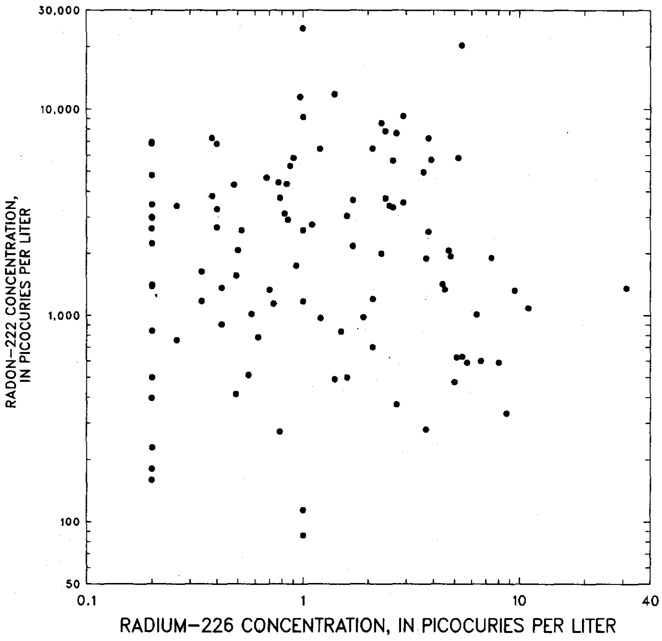

Figure 114. Relation between radon-222 and radium-226 concentrations for 103 wells in the Chickies Quartzite.

water from the Chickies Quartzite suggests that controls on ^{222}Rn concentrations are site specific. In a fractured-rock aquifer, the proximity of ^{226}Ra to water bearing zones, ground-water flow distance, and flow time will affect concentrations of ^{222}Rn in solution. Wathen (1987) and Rumbaugh (1983) suggest that low uranium concentrations and elevated ^{222}Rn concentrations in ground water indicate a ^{226}Ra source in the aquifer matrix close to the well rather than in the ground water.

Radon-222 has a slightly negative significant correlation with specific conductance, sulfate, magnesium, and total dissolved solids (table 51). Mechanisms responsible for these relations are not presently understood.

CONCLUSIONS

Elevated concentrations of ^{226}Ra and ^{228}Ra in ground water in the Chickies Quartzite indicate that the geochemical environment favors the mobility of radium. Elevated radium concentrations in ground water are distributed throughout the formation. The ground water is dilute, soft, and acidic. The ground water contains detectable concentrations of dissolved iron and manganese and is slightly reducing with respect to these constituents.

Adsorption is an important control on radium concentrations in solution. Low pH decreases the ability of quartz and kaolinite in the aquifer matrix to adsorb radium. Iron and manganese oxyhydroxides also provide possible adsorption sites, and the dissolution of these minerals decreases the number of available adsorption sites and may release adsorbed radium into solution.

Radium in solution may complex with dissolved organic carbon. Concentrations of dissolved radium increase with increasing concentrations of dissolved organic carbon. This fact suggests that complexation of radium with dissolved organic carbon may be an important control on dissolved radium concentrations and mobility in the ground water or that DOC and radium exhibit similar chemical behavior.

Barium and radium exhibit similar chemical behavior. Data from this study show a positive correlation between barium and manganese concentrations and radium and manganese concentrations. Similar behavior of radium and barium with respect to manganese oxyhydroxide adsorption is implied.

The slight positive correlation between dissolved radium concentrations and specific conductance indicates that radium solubility may increase with ionic strength in the ground water of the Chickies Quartzite.

At pH less than 4.7, all samples of ground water contained concentrations of total radium that exceeded the EPA MCL of 5 pCi/L. Low pH may be used as a predictor of elevated radium concentrations in ground water of the Chickies Quartzite.

Sources of ^{226}Ra, ^{228}Ra, and ^{222}Rn in the ground water appear to be from within the aquifer matrix of the Chickies Quartzite. Because the formation typically forms ridges and is a water-table aquifer recharged primarily by local precipitation, it is unlikely that radionuclides in solution have sources outside the formation. Observed activity ratios of ^{228}Ra and ^{226}Ra may reflect the ^{232}Th and ^{238}U content of the aquifer matrix and distribution of the thorium- and uranium-bearing minerals within the aquifer.

Lack of correlation between ^{222}Rn and its parent, ^{226}Ra, suggests that ^{222}Rn is not supported by ^{226}Ra in solution and that the source is in the aquifer matrix. Regional and local variability of ^{222}Rn concentrations in ground water of the Chickies Quartzite indicates that controls on concentrations are site specific.

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RADON IN GROUND WATER OF CARSON VALLEY, WEST-CENTRAL NEVADA

Michael S. Lico and Timothy G. Rowe

Abstract

Radon-222 activities in ground-water samples from the basin-fill sedimentary aquifers of Carson Valley were determined to delineate the areal distribution of, and possible geochemical controls on, the radon-222. Activities of radon-222 in 30 ground-water samples ranged from less than 100 to 10,000 picocuries per liter and have a median activity of 480 picocuries per liter. The highest radon-222 activities are in ground water from the confined alluvial-fan deposits along the western periphery of Carson Valley.

Several factors may contribute to the high radon-222 activity in ground water of Carson Valley. First, Cretaceous granitic rocks of the Carson Range (western border of Carson Valley) contain uranium in concentrations as high as 10 milligrams per kilogram. These rocks, both as alluvial detritus and bedrock, may contribute significant amounts of radon-222 to ground water in the alluvial-fan deposits. Second, ground water enriched in radon-222 may flow from fractured bedrock into the alluvial-fan aquifer. Third, water rich in radon-222 also may be moving upward along a major fault bordering the eastern side of the Carson Range. Fourth, organic-rich sediments along the western side of Carson Valley contain high uranium concentrations (greater than 53 milligrams per kilogram) and may be releasing radon-222 to the ground water. These factors, along with the composition of radon-222-enriched water and the slope of the piezometric surface, indicate that most of the radon-222 is derived from the Carson Range.

INTRODUCTION

The major source of domestic drinking water in Carson Valley is ground water. Recent increases in population have resulted in the construction of many new private and municipal wells. Although little is known about the distribution and occurrence of radon-222 (^{222}Rn) in ground water of western Nevada, previous studies suggest that high activities may exist, and these activities would pose some risk to the population. Otton and others (1985, p. 21) found anomalous activities of ^{222}Rn in water from springs in the Lake Tahoe basin (fig. 115). The same study (Otton and others, 1985, table 1) revealed that slightly elevated concentrations of uranium, 5–53 mg/kg (milligrams per kilogram), are present in the valley-fill sediments derived from the Carson Range along the western margin of Carson Valley. A study of public drinking-water supplies (Horton, 1985, p. 123) found high activities of ^{222}Rn in the Gardnerville, Reno, and Yerington areas.

Radon-222 is a naturally occurring, colorless, odorless, inert gas that is formed by the radioactive decay of uranium-238 (Wanty and Schoen, this volume). Uranium-238 is present in trace concentrations in all rocks, with certain types such as granitic rocks, late-stage volcanic rocks, shales, and limestones generally having higher uranium concentrations (Lanctot and others, 1985, p. 68). Two intermediate radioactive decay products of the uranium-238 decay series, thorium-230 and radium-226, are important in determining the amount of ^{222}Rn generated. Because both thorium and radium tend to be immobile under most geohydrologic conditions, the uranium content of rocks generally is a good indicator of ^{222}Rn activity in associated ground water.

Several natural geochemical processes can concentrate uranium-238 in the environment. Late-stage igneous rocks, present in western Nevada, are rich in uranium because of the occlusion of uranium from the crystal lattice of early-forming minerals. The same process occurs in high-grade metamorphic rocks during recrystallization and enriches the late-crystallizing minerals with uranium. Uranium also can be concentrated by transport, by way of ground water, from an oxidizing to a reducing environment. Roll-front uranium deposits (Turner-Peterson and Fishman, 1986) exemplify this type of process. Otton and others (1985, p. 7) have reported several areas in the Carson Range where organic-rich bog sediments have accumulated uranium. Hydrothermal fluids also can produce deposits of extremely high uranium content.

Radon-222 is radioactive, and at high concentrations it and its daughter products can cause lung cancer in humans (Cothern, 1987, p. 7). Radon-222 released from drinking-water sources has been estimated to cause as much as 7 percent of the lung cancer fatalities (between 5,000 and 20,000 per year) attributed to ^{222}Rn (Cothern and others, 1986, p. 40). Radon gas is released from water to the atmosphere by any one of several household activities, including showering and laundering (Lowry and others, 1987, p. 363).

Human exposure to ^{222}Rn, and its radioactive daughter products (isotopes of polonium, lead, and bismuth), is possible through (1) inhalation of ^{222}Rn gas or decay products that adhere to small dust particles and directly affect the lungs and (2) ingestion of ^{222}Rn-containing water, by which the dose of radiation to the stomach and intestines is increased (Brutsaert and others, 1981, p. 407). Potential health effects are greatest to private domestic well-water users by these exposure pathways because of the generally short time between pumping and consumption of well water. The half-life of ^{222}Rn is relatively short (3.82 days); thus, the residence time of water in public-supply systems and loss to the atmosphere in storage tanks would determine the potential effects of high ^{222}Rn activity in these systems.

Purpose and Scope

The primary purpose of this report is to describe the areal distribution of ^{222}Rn in ground water in Carson Valley, west-central Nevada. A secondary purpose is to relate high ^{222}Rn activities with certain types of rocks or sediments. This investigation was made during 1987 by the U.S. Geological Survey.

Description of Study Area

Figure 115 shows the location and general features of the study area. Most of Carson Valley lies in west-central Nevada; the extreme southwestern part of the valley lies in California. Carson Valley is bordered on the west by the Carson Range of the Sierra Nevada (maximum altitude, about 10,000 ft (feet)) and on the east by the Pine Nut Mountains (maximum altitude, nearly 9,000 ft). Carson Valley is approximately 15 mi (miles) wide and 24 mi long, with a valley floor that slopes from altitudes of about 5,000 ft in the south to about 4,600 ft in the north (Maurer, 1986, p. 7). The major population center in Carson Valley is the Minden-Gardnerville area. The economy of Carson Valley is dominated by agriculture and related activities.

The climate on the valley floor is semiarid, with mean annual precipitation of less than 9 in (inches) (Glancy and Katzer, 1976, table 4) and mean annual evapotranspiration exceeding 40 in (Pennington, 1980, table 9). The Carson Range and the Pine Nut Mountains receive 45 and 26 in of precipitation, respectively, in an average year, most of which is winter snowfall (Maurer, 1986, p. 7).

Methods of Sample Collection and Analysis

Water samples considered representative of the sampled aquifers were obtained at each well by pumping or bailing until water temperature, specific conductance, and pH were constant, and a minimum of three well-bore volumes was extracted. With the public supply and domestic wells (table 52), existing in-well pumps were used, and the sample was obtained as near to the well as possible. The observation wells (table 52) were sampled by use of a Teflon bailer. Loss of ^{222}Rn gas due to aeration by the pump may be significant in water samples from public-supply wells. Radon-222 values produced for samples from these wells represent minimum ^{222}Rn activities.

A sample for radon analysis was obtained by filling a 1-L (liter) polyethylene bottle gently to avoid aeration. A 10-mL (milliliter) water sample from the bottle was withdrawn with a syringe below the water surface. The 10-mL sample was added to a 30-mL glass scintillation vial that contained 5 mL of a mineral-oil-based liquid scintillator by careful injection of the water sample underneath the mineral oil. The vial then was capped tightly and shaken. The vials were shipped by express mail to the contract laboratory for analysis by liquid scintillation counting. Samples were shipped as soon as possible after collection so radon decay would be minimized prior to analysis.

Duplicate water samples were collected from each well for ^{222}Rn analysis. The duplicate analyses generally agreed to within 5 percent and were averaged and reported as one value. For differences greater than 5 percent, the larger value is reported.

The radon concentrations in five well waters (sites 14, 15, 19, 28, and 29 in table 54) were measured by use of a field-screening method. A 1-L polyethylene bottle was filled with 750 mL of water and capped with a septum cap. The bottle was shaken vigorously for 30 seconds and then allowed to stand for 3 minutes. A 50-mL sample of the head-space gas was removed through the septum cap with a syringe and needle. The gas sample was injected, through a septum connector, into an evacuated Lucas-type, phosphor-coated cell. The cell was inserted into an EDA RDA-200 alpha scintillometer, and the gas activity counted for 30 minutes. The counts were converted to radon activities in picocuries per liter.

GEOHYDROLOGIC SETTING

Geology

The area discussed in this report is entirely within the borders of the Cretaceous Sierra Nevada batholith (Moore, 1969, p. 1). Pre-Cretaceous rocks, which are present as roof pendants in the batholith, include for the most part Triassic and Jurassic metamorphosed volcanic rocks and volcanically derived sedimentary rocks (fig. 116 and table 53). Some limestone and gypsum are interbedded with the clastic and volcanic rocks. The Cretaceous batholith is, in this area, composed of quartz monzonite, granodiorite, and some mafic rocks.

Overlying the granitic and metamorphic rocks is a thick sequence of Cenozoic volcanic and interbedded sedimentary rocks. Generally, this sequence begins with Miocene rhyolitic tuff that is several thousand feet thick. Overlying the rhyolite is a thick accumulation of Miocene and Pliocene andesitic and dacitic breccias interbedded with lacustrine and fluviatile sediments. The youngest volcanic rocks are Pliocene and Pleistocene basaltic flows that cap many of the mountain ranges.

Alluvium is present as valley-fill sediments throughout the area. The older alluvial deposits include pediment gravel of Pliocene to Pleistocene age capping poorly con-

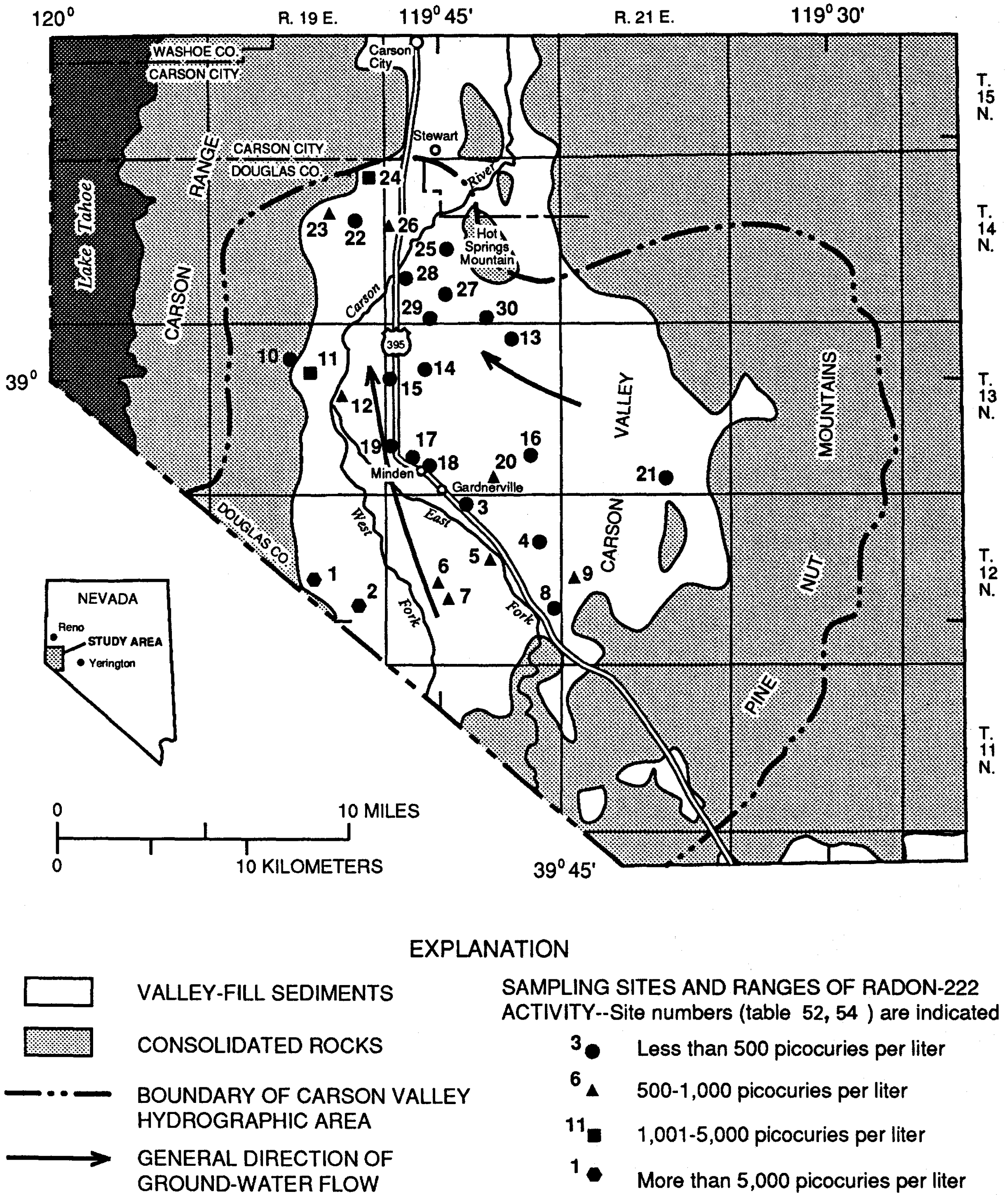


Figure 115. Location of study area and sampling sites. Symbols indicate levels of radon-222 activities.

solidated Tertiary sediments. Younger alluvium, mainly alluvial-fan gravel and stream-laid gravel, sand, and silt, occupies the valley floor and attains a thickness of at least 5,000 ft (Maurer, 1986, plate 2).

The geologic structure of the study area is dominated by normal faulting. Most of the deformation was during two general periods, one preceding and accompanying the emplacement of the Cretaceous granitic rocks and the other consisting of normal faulting, warping, and tilting, in late Tertiary and Quaternary time. The Carson Range, a major part of the study area, is an uplifted block that has prominent normal faults bordering it on the eastern and

Table 52. Data on sampled wells, Carson Valley, Nev.
[—, data not available]

Site no. (fig. 115)	Local identification[1]	Depth (feet below land surface) Producing interval	Depth (feet below land surface) Total	Aquifer unit[2]	Use[3]	Reported lithology of producing interval
1	105 N12 E19 15ABAD1	125–300	300	A	P	Sand, gravel, clay lenses.
2	105 N12 E19 23DDD1	121–141	141	A	D	Decomposed granite, clay lenses.
3	105 N12 E20 04ADA1	100–300	300	C	P	Sand, gravel, some clay lenses.
4	105 N12 E20 11ADD1	105–125	125	B	D	Gravel and sand.
5	105 N12 E20 15ADD1	183–372	375	C	P	Gravel, cobbles, cemented gravel, clay.
6	105 N12 E20 17CCD1	67–87	91	B	D	Gravel.
7	105 N12 E20 20ABAA1	200–450	450	—	P	Gravel, sand, clay lenses.
8	105 N12 E20 24ADC1	122–142	145	—	D	Gravel.
9	105 N12 E21 18CAB1	—	—	—	D	No report.
10	105 N13 E19 09ADCA1	156–176	180	A	D	Gravel, broken rock, clay lenses.
11	105 N13 E19 10DBB1	80–115	115	A	D	Sand, gravel, clay lenses.
12	105 N13 E19 13BCC1	150–500	518	A	I	Sand, gravel, clay lenses.
13	105 N13 E20 02CBB1	156–176	176	B	D	Sand.
14	105 N13 E20 08CAA1	110–125	135	B	D	Sand.
15	105 N13 E20 18BAAA1	—	20	B	O	No report.
16	105 N13 E20 26ABB1	72–85	150	B	D	Decomposed granite, sand, clay lenses.
17	105 N13 E20 29BDDD1	93–114	118	C	D	Sand, gravel.
18	105 N13 E20 29CDC1	—	400	—	P	No report.
19	105 N13 E20 30BCAD1	200–360	360	C	P	Gravel, clay lenses.
20	105 N13 E20 34ACC1	—	—	—	D	No report.
21	105 N13 E21 33BCAB1	140–160	163	—	D	Gravel, clay lenses.
22	105 N14 E19 12ADA2	120–150	155	B	D	Gravel, sand, and clay lenses.
23	105 N14 E19 14BBD1	76–96	100	A	D	Decomposed granite, clay lenses.
24	105 N14 E20 06CBAB2	73–93	96	B	D	Sand.
25	105 N14 E20 17ADCA1	—	27	B	O	No report.
26	105 N14 E20 18ABAB1	151–301	425	C	P	Gravel and clay.
27	105 N14 E20 28CDC1	68–88	88	B	D	Gravel.
28	105 N14 E20 30DCCB1	—	20	B	O	No report.
29	105 N14 E20 32DCCC1	—	21	B	O	No report.
30	105 N14 E20 34BDC1	—	100	—	D	No report.

[1] The local site-identification system used in this report is based on an index of hydrographic areas in Nevada (Rush, 1968) and the rectangular subdivision of the public lands referenced to the Mount Diablo base line and meridian. Each site designation consists of four units separated by spaces. The first unit is the hydrographic area number. The second unit is the township, preceded by an N or S to indicate location north or south of the base line. The third unit is the range, preceded by an E to indicate location east of the meridian. The fourth unit consists of the section number and letters designating the quarter section, quarter-quarter section, and so on (A, B, C, and D indicate the northeast, northwest, southwest, and southeast quarters, respectively), followed by a number indicating the sequence in which the site was recorded. For example, site 105 N12 E20 4ADA1 is in Carson Valley (hydrographic area 105). It is the first site recorded in the NE 1/4 of the SE 1/4 of the NE 1/4 of section 4, Township 12 North, Range 20 East, Mount Diablo base line and meridian.

[2] Aquifer unit refers to unit of unconsolidated Quaternary sedimentary aquifer: A, confined alluvial-fan unit; B, unconfined fluviatile and lacustrine unit; and C, confined fluviatile and lacustrine unit.

[3] Use of well: D, domestic-supply well; I, irrigation well; O, observation well; P, public-supply well.

western flanks. These faults have displacements of at least 4,000 ft (Moore, 1969, p. 17). The Pine Nut Mountains constitute a west-tilting structural block that borders the study area on the east. The west flank of these mountains is dominated by many north-trending normal faults that have relatively small displacements (Moore, 1969, p. 18).

The uranium and thorium concentration of sediment derived from the various rock types in the area has been determined by the National Uranium Resource Evaluation program (Bennett, 1980). Samples were taken from source areas of Jurassic, Tertiary, and Quaternary rocks. Generally, uranium concentrations range from about 2 to 5 mg/kg (milligrams per kilogram), and thorium from 4 to 16 mg/kg in stream sediment, regardless of the age of the source rocks. Forty-seven shallow soil samples collected in Carson Valley by the U.S. Geological Survey (Ron Tidball, written commun., 1988) had a thorium concentration that averaged 11 mg/kg and ranged from 5 to 42 mg/kg. Data for uranium and thorium content of the silicic volcanic rocks in the study area are lacking. Elsewhere in the northern Basin and Range province, however, Walker (1981) found the uranium and thorium content of silicic volcanic complexes, mostly rhyolites, to range from 2 to 17 and 6 to 66 mg/kg, respectively.

Table 53. Generalized stratigraphy of the Carson Valley study area (modified from Moore, 1969)

Age	Rock unit (map symbol)	Description
Quaternary	Younger alluvium (Qal)	Mainly alluvial-fan gravel, stream-laid gravel, sand, and silt, with some talus and dune sand. Also includes some fine sand, silt, and clay of river flood plains, and playa clay and sand.
Quaternary and Tertiary	Older alluvium (QToa)	Predominantly fanglomerate and pediment gravel; includes terrace gravel and late Pleistocene lake beds.
	Basaltic rocks (QTb)	Predominantly thin lava flows, with interbeds of scoriaceous basalt breccia and diatomaceous sediments.
Tertiary	Andesitic rocks (Ta)	Flow breccias, lava flows, and agglomerates, with interbedded sediments. Locally includes basaltic and rhyolitic rocks.
	Sedimentary rocks (Ts)	Lacustrine and fluviatile sediments. Sandstone, shale, mudstone, marl, diatomite, limestone, and calcareous tufa. Includes interbedded tuffaceous rocks, lava flows, and breccias.
	Rhyolitic rocks (Tr)	Rhyolitic tuffs, flows, and intrusions. Biotite rhyolite pumice tuff-breccia and welded tuff are widespread.
Cretaceous	Granitic rocks of the Sierra Nevada batholith (Kg)	Granite porphyry, porphyritic quartz monzonite, nonporphyritic quartz monzonite, granodiorite, and hybrid mafic rocks.
Jurassic and Triassic	Metasedimentary rocks (JTRs)	Shale, slate, tuffaceous siltstone, sandstone, and graywacke, largely derived from volcanic rocks. Includes interbeds of conglomerate, limy shale, limestone, dolomite, and gypsum.
	Metavolcanic rocks (JTRv)	Andesite breccias, tuffs, and flows; basalt; and rhyolite; with interbedded volcanic-derived sedimentary rocks and limestone. Metamorphosed to greenschist or higher facies.

Hydrology

Three generalized aquifers are present in the Carson Valley study area (fig. 117): (1) an unconsolidated Quaternary sedimentary aquifer, (2) a poorly consolidated Tertiary sedimentary aquifer, and (3) a granitic and metamorphic bedrock aquifer surrounding and underlying the basin-fill sediments (Maurer, 1986, p. 16).

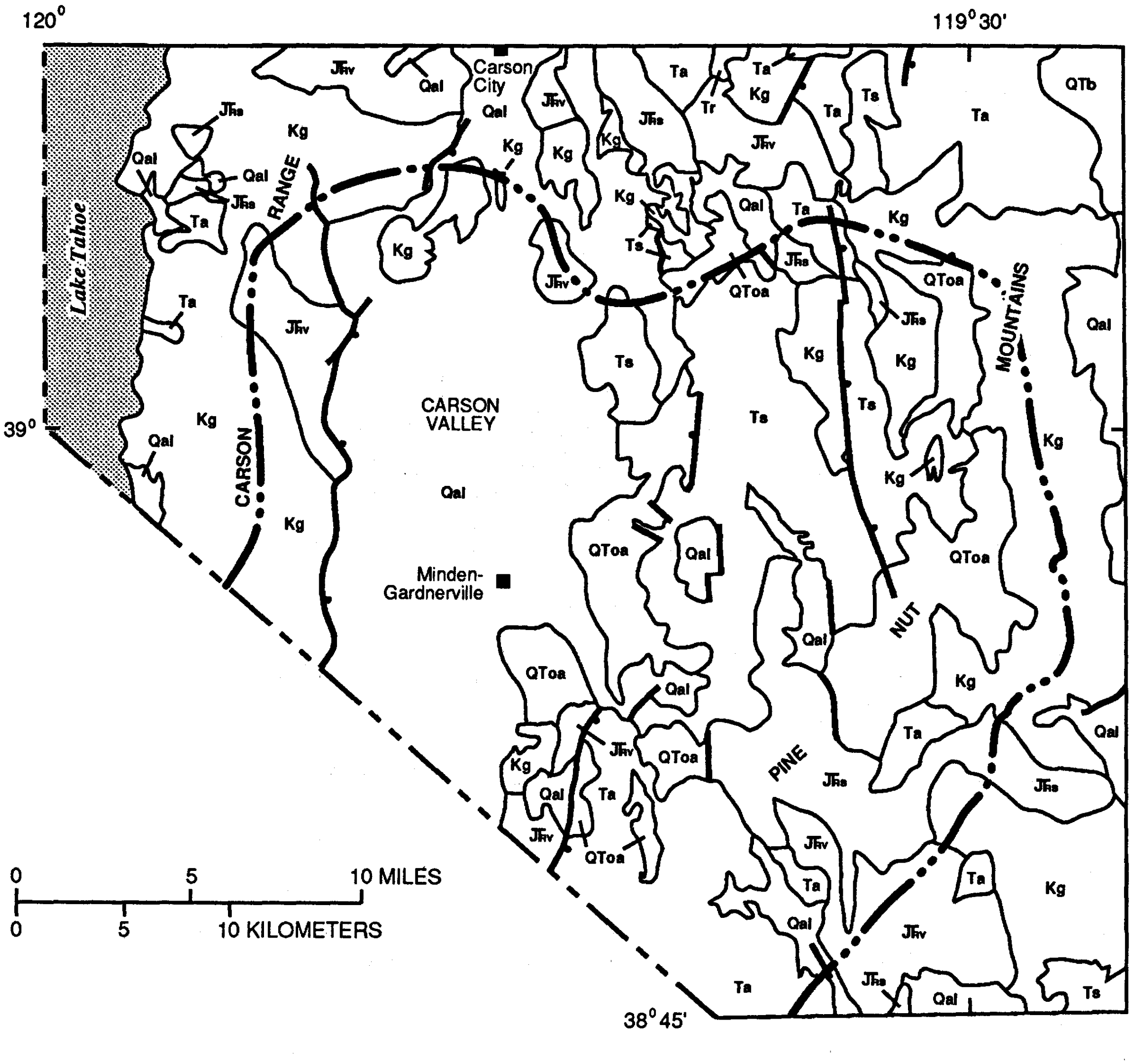
The unconsolidated Quaternary sedimentary aquifer is the most common source of water for domestic, municipal, and irrigation supply in Carson Valley. This aquifer has been further subdivided by Maurer (1986, p. 16) into (1) a confined unit consisting of alluvial-fan deposits along the western side of the valley, (2) an unconfined unit of fluvial and lacustrine sediments present throughout the valley, and (3) a confined unit consisting of fluvial and lacustrine sediments present throughout the valley.

Ground-water flow in the unconfined (water-table) unit is generally toward the center of the valley from the east and west, and then north along an axis approximately 2 mi west of U.S. Highway 395 (see fig. 115). Gradients range from about 5 ft/mi in the north to as great as 100 ft/mi in the southwestern and western parts of the valley (Maurer, 1986, p. 17). Water in the underlying confined units generally flows in the same direction as in the water-table unit but with reduced gradients. The depth to water ranges from about 5 ft on the valley floor to more than 100 ft on the alluvial fans along the margins of the valley.

DISTRIBUTION OF RADON IN THE GROUND WATER

Ground water is the major source of drinking water in the Carson Valley. Most households outside of the urban Minden-Gardnerville area use private wells as water sources. Residences are distributed throughout the valley in such a way as to allow good areal coverage for the sampling of ground water. Samples were collected from the deeper confined units and the shallow water-table unit (table 52).

Radon-222 activities in ground water sampled for this study ranged from less than 100 to 10,000 pCi/L (picocuries per liter; table 54). The distribution of ^{222}Rn activities in Carson Valley is shown in figure 118. Activities of ^{222}Rn are log-normally distributed about a median value of 480 pCi/L, except for the two highest values (5,600 and 10,000 pCi/L). Without exception, the highest ^{222}Rn activities (>1,000 pCi/L) determined in this study were present in



EXPLANATION

Qal	YOUNGER ALLUVIUM	Kg	GRANITIC ROCKS OF THE SIERRA NEVADA BATHOLITH
QToa	OLDER ALLUVIUM	J̄R̄s	METASEDIMENTARY ROCKS
QTb	BASALTIC ROCKS	J̄R̄v	METAVOLCANIC ROCKS
Ta	ANDESITIC ROCKS	━━●━━	FAULT -- Ball on downthrown side
Ts	SEDIMENTARY ROCKS	━━ ● ● ━━	BOUNDARY OF CARSON VALLEY HYDROGRAPHIC AREA
Tr	RHYOLITIC ROCKS		

Figure 116. Generalized geology of the study area and adjacent areas. Modified from Moore (1969); see table 53 for description of rock units.

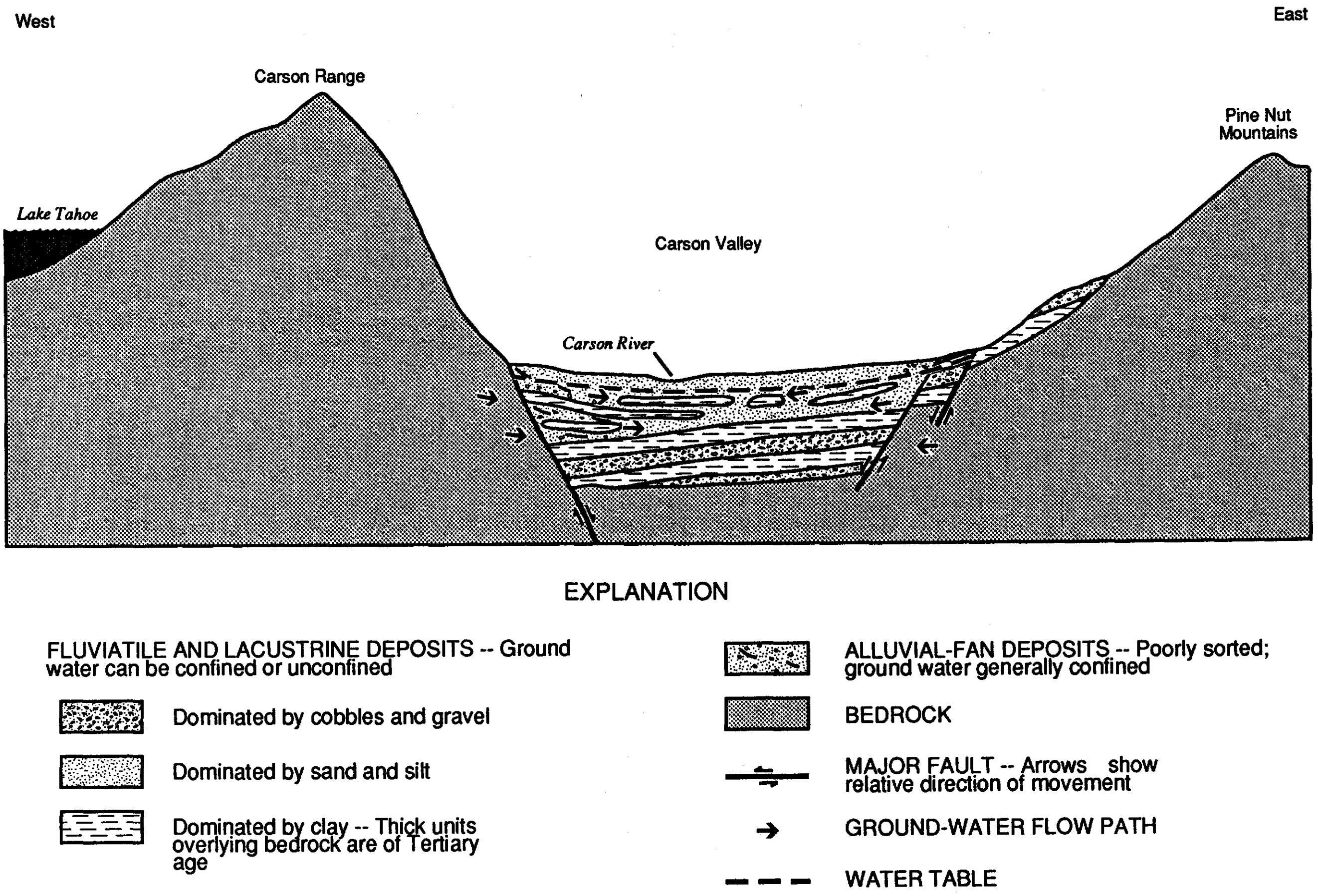


Figure 117. Cross section of hydrogeologic relations in Carson Valley. Upper part of basin-fill section is Quaternary age and lower part is Tertiary age. View looking north, modified from Maurer (1986, p. 44).

ground water from wells along the extreme western side of Carson Valley. These wells (table 52) are completed in or close to the alluvial-fan deposits at the base of the Carson Range.

GEOLOGIC AND GEOCHEMICAL CONTROLS ON RADON IN THE GROUND WATER

The high activities of ^{222}Rn in ground water from the western side of Carson Valley may be caused by several factors. The alluvial-fan deposits along the western side of the valley consist primarily of coarse-grained, relatively fresh detritus from the Cretaceous intrusive rocks of the Carson Range. These rock fragments have been mechanically weathered but show little evidence of chemical weathering (Otton and others, 1985, p. 25). Uranium concentration ranges from 2.9 to 10.1 mg/kg and averages 5.4 mg/kg in granitic rocks in the Carson Range (Otton and others, 1985, p. 23). Because of their high uranium concentrations, the granitic rocks may generate sufficient ^{222}Rn to contribute to the associated ground water and cause the high ^{222}Rn activities measured.

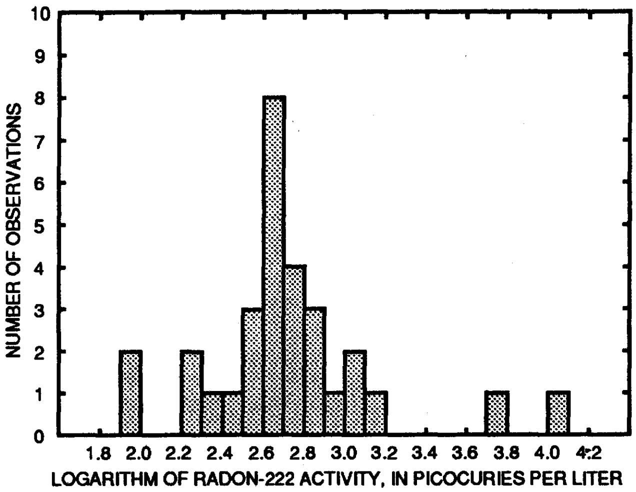


Figure 118. Distribution of radon-222 activities in sampled ground water.

Another possible factor that may be contributing to elevated ^{222}Rn activities in ground water associated with the alluvial-fan deposits is direct flow into the aquifer

Table 54. Analyses of water from sampled wells in the Carson Valley

[Chemical analyses are measures of dissolved concentration in milligrams per liter, except as indicated. Abbreviations: °C, degrees Celsius; µS/cm, microsiemens per centimeter; µg/L, micrograms per liter; pCi/L, picocuries per liter; —, data not available]

Site no. (fig. 115)	Sample date	pH (standard units)	Water temperature (°C)	Specific conductance (µS/cm)	Calcium	Magnesium	Sodium	Potassium	Bicarbonate	Carbonate	Sulfate	Chloride	Fluoride	Iron (µg/L)	Dissolved organic carbon	Radon-222 (pCi/L)[1]
1	87–06–23	6.9	12.0	82	8.4	1.0	7.4	0.6	41	<1	7.3	0.6	0.2	12	4.5	10,000
2	87–04–16	7.8	15.0	102	5.2	.5	18	.7	36	<1	21	1.1	.8	59	.5	5,600
3	87–04–13	7.2	12.0	385	44	13	21	2.8	195	<1	32	11	.1	<3	.7	480
4	87–04–15	8.1	11.0	273	34	6.4	16	2.2	127	<1	21	9.1	.1	<3	.5	440
5	87–04–10	7.3	12.0	219	22	6.8	14	1.8	97	<1	24	5.5	.1	7	.8	530
6	87–04–16	6.9	13.5	200	21	8.7	11	2.3	102	<1	11	5.3	<.1	<3	.8	800
7	87–06–23	8.7	17.5	351	8.2	2.0	70	.9	146	5	39	5.9	1.4	8	4.4	760
8	87–04–28	7.9	14.0	578	78	14	26	2.5	248	<1	64	22	<.1	<3	.9	<100
9	87–04–14	7.5	21.0	414	34	5.0	53	2.4	168	<1	34	17	.1	7	.8	590
10	87–04–29	7.2	12.0	282	42	5.7	15	3.1	184	<1	12	1.2	.2	14	.6	380
11	87–04–29	7.6	13.0	199	28	5.7	10	1.4	125	<1	2.1	2.0	<.1	14	.5	1,100
12	87–04–28	8.0	15.5	196	15	4.3	17	4.4	100	<1	18	4.4	.3	—	.6	500
13	87–04–15	8.0	20.0	339	9.2	3.0	59	3.8	138	<1	37	10	1.5	10	.6	320
14	87–04–27	8.0	15.0	383	38	2.9	41	3.6	101	<1	82	18	.4	26	.9	[2]500
15	87–06–29	7.2	12.0	889	100	23	65	5.4	310	<1	130	71	.2	—	8.8	[2]200
16	87–04–13	7.7	15.5	318	31	9.3	24	2.9	172	<1	30	7.7	.2	36	.6	440
17	87–04–15	7.9	12.0	476	63	16	20	3.4	275	<1	19	5.6	<.1	<3	1.1	480
18	87–04–27	8.4	14.0	183	21	3.9	13	2.1	93	1	17	4.4	.1	<3	.5	350
19	87–06–25	8.9	12.0	164	15	3.0	19	2.5	78	7	16	4.6	.4	79	.4	[2]20
20	87–04–16	7.8	14.0	300	33	7.4	23	1.6	164	<1	20	5.2	.2	3	.7	780
21	87–06–24	8.0	14.5	331	35	11	25	3.9	136	<1	66	6.9	.2	32	.4	310
22	87–04–30	8.2	13.0	243	18	2.0	33	1.7	107	<1	20	12	1.4	16	.8	460
23	87–04–29	8.6	15.0	117	13	.07	14	1.0	65	2	0.7	1.0	<.1	120	.5	660
24	87–05–30	7.4	13.5	518	61	7.1	41	2.6	227	<1	23	17	.6	5	1.2	1,200
25	87–04–29	6.7	14.0	3,670	230	6.9	570	6.8	143	<1	1,700	200	5.6	9,600	2.5	<100
26	87–04–16	8.6	19.0	260	14	1.7	42	.7	111	5	17	12	.4	.5	3	560
27	87–04–17	8.0	15.0	473	38	4.3	63	3.5	169	<1	76	13	1.7	5	.7	200
28	87–06–30	7.5	12.0	401	43	7.6	31	5.1	195	<1	50	11	.4	—	—	[2]300
29	87–06–30	7.9	13.5	397	20	8.1	58	1.9	183	<1	54	8.5	.9	—	2.8	[2]400
30	87–04–17	7.7	16.0	353	15	4.9	56	3.3	128	<1	47	16	1.5	<3	—	460

[1] Samples for ^{222}Rn analyses were collected August 24–27, 1987.

[2] Values are results from gas-scintillation counting, where extraction procedures produced values lower than the liquid scintillation method.

from subsurface fractures in the granitic bedrock. Most recharge is from spring snowmelt in the Carson Range and along the alluvial fans at the base of the range. Some of this water flows directly from the bedrock into the alluvial-fan aquifer. This water would likely contain an elevated ^{222}Rn activity because of its intimate contact with the Cretaceous granitic rocks.

Wells along the western side of the valley typically are artesian. Upward head gradients exist that may be carrying ^{222}Rn-rich water up from depth. At the edge of the valley, lateral distance to bedrock is less, and bedrock presumably is closer to land surface, a situation that allows for the short-time flow paths needed to minimize ^{222}Rn decay.

The major normal fault along the eastern base of the Carson Range (fig. 116) separates bedrock from the basin-fill sediments. This fault has a displacement exceeding 4,000 ft and dips 36° to 60° to the east beneath the valley-fill sediments (Moore, 1969, p. 17). The fault zone may act as a conduit for deeper ^{222}Rn-rich water from the bedrock aquifer to migrate up into the confined alluvial-fan unit.

Relatively high uranium concentrations (greater than 53 mg/kg) are present in sediments from shallow auger holes along the bottom of Carson Valley adjacent to the eastern side of the Carson Range (Otton and others, 1985, table 1). These sediments are rich in organic materials that tend to sorb dissolved uranium from the ground water. These sediments are young, and, thus, their ^{222}Rn-generation potential is relatively low. Similar depositional processes have been dominant since late Tertiary time (Moore, 1969, p. 14); therefore, uranium-rich sediments probably are present at depth in the valley-fill sediments along the western side of Carson Valley. These older deposits may contain sufficient uranium to cause elevated activities of ^{222}Rn in ground water.

Most of the ground water in the unconsolidated Quaternary sedimentary aquifer in Carson Valley is dilute and of the sodium-calcium-bicarbonate type. Although most of these waters have specific conductances of less than 500 µS/cm (microsiemens per centimeter), ground water from one well in this study (well 25 in tables 52 and 54), which has been influenced by seepage from sewage-effluent ponds, and possibly by geothermal water, is slightly saline. In general, ground water is more mineralized toward Hot Springs Mountain in the northeast part of the valley (Garcia, 1988).

Water-quality relations suggest that the ^{222}Rn is derived from the granitic rocks of the Carson Range. Sulfate and specific conductance are inversely related to ^{222}Rn activities (fig. 119); that is, waters having high specific conductances or sulfate concentrations tend to have low ^{222}Rn activities. No apparent relation exists between ^{222}Rn activity and other water-quality constituents in ground water from Carson Valley. Water from springs and runoff in the Carson Range typically is dilute (less than 200 µS/cm), low in sulfate (less than 4 mg/L), of the sodium-calcium-bicarbonate type (Feth and others, 1964, table 1), and similar to ground water on the western side of Carson Valley. Ground water on the eastern and northern sides of the valley commonly has higher specific conductance and sulfate concentrations (see fig. 115 and table 53) than water on the western side of the valley (Garcia, 1988). Apparently, water east of the major ground-water trough in the Carson Valley, as indicated by the slope of the piezometric surface, is derived largely from recharge in the Pine Nut Mountains (fig. 115), where Tertiary sedimentary rocks predominate. Although the uranium concentration of these rocks is unknown, the ^{222}Rn generation potential of these rocks and their derived sediments probably is low, as reflected in the low ^{222}Rn activities in ground water from the east side of the valley.

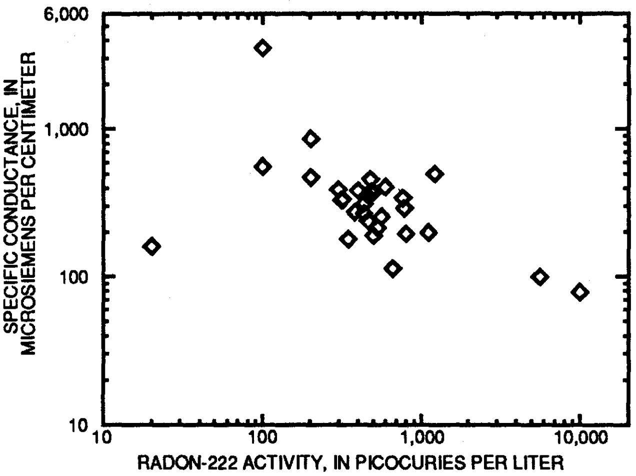


Figure 119. Relation between radon-222 and specific conductance in sampled ground water.

SUMMARY

Ground water in the unconsolidated Quaternary sedimentary aquifer of Carson Valley contains ^{222}Rn activities ranging from less than 100 to 10,000 pCi/L. The median activity for 30 ground-water samples is 480 pCi/L. Radon-222 activities are highest along the western periphery of Carson Valley in the alluvial-fan deposits.

Several possible factors may contribute to elevated ^{222}Rn activities. Cretaceous granitic rocks that have high uranium content, both as bedrock and alluvial detritus, may release ^{222}Rn to the ground water. Significant amounts of ^{222}Rn-enriched ground water from the fractured bedrock may flow into the confined alluvial-fan unit. The major normal fault bordering the eastern side of the Carson Range may function as a conduit for ^{222}Rn-rich water from depth. Organic-rich sediments that are present along the western side of Carson Valley contain high uranium concentrations and may be releasing ^{222}Rn to the ground water.

Relations between water quality and ^{222}Rn activities indicate that the major source of ^{222}Rn is the Carson Range. Radon-222 activity is related inversely to sulfate and specific conductance. The chemical character of ^{222}Rn-rich ground water in the unconsolidated Quaternary aquifer is similar to that of water from the Carson Range.

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GEOCHEMISTRY OF GROUND WATER AND RADIONUCLIDE MOBILITY IN TWO AREAS OF THE READING PRONG, EASTERN PENNSYLVANIA

Richard B. Wanty, Paul H. Briggs, and Linda C.S. Gundersen

Abstract

High levels of indoor radon-222 have been found in homes built on the Reading Prong, a continuous exposure of Proterozoic metamorphic rocks trending northeastward from Reading, Pa. The radon-222 is derived from radioactive decay of uranium-238, in which these rocks are enriched. Radon-222 is present also in the ground water in these fractured crystalline aquifers. Ground-water samples were taken from 31 domestic supply wells in two areas and were analyzed for common major and minor elements, as well as for total uranium, radium-226, and radon-222. The waters are dilute, usually less than 200 milligrams per liter in total dissolved solids, but many of the water samples had high concentrations of the radionuclides. In a study area near Boyertown, Pa., measured pH values ranged from 5.8 to 9.0. The water quality in that area is determined by the aquifer rock types, which include hornblende gneiss and quartz-feldspar gneiss; some of the quartz-feldspar gneiss is mylonitized. In Easton, Pa., the water samples had low total dissolved solids and slightly acidic pH values (5.5 to 6.7). The dominant rock type in Easton is also quartz-feldspar gneiss, but one well appeared to be penetrating a local carbonate aquifer.

In Boyertown, the highest radon-222 and uranium concentrations were in water drawn from the mylonitized quartz-feldspar gneiss. Dissolved radon-222 concentrations as high as 100,000 picocuries per liter and total uranium concentrations as high as 200 picocuries per liter were measured. Radium-226 concentrations ranged from <0.1 to 4.2 picocuries per liter but seldom exceeded 1 picocurie per liter. In Easton, dissolved radon-222 values were generally higher, ranging from 6,000 to 80,000 picocuries per liter. Radium-226 concentrations also were higher, ranging from 0.2 to 10.1 picocuries per liter. Secular equilibrium between radon-222 and uranium-238 is not attained in any of the samples from either area. In every case, radon-222 activity greatly exceeds that of uranium-238 in the water. Thus, the source of the dissolved radon-222 lies ultimately in uranium-238 in the solid phase. Likewise, the aqueous radium-226 activity is lower than that of radon-222 in every sample. Dissolved uranium concentrations are controlled by the oxidizing or reducing nature of the ground water. However, the data suggest that for oxidizing ground water, uranium concentrations are independent of major chemical parameters such as pH and carbonate concentration and depend on aquifer rock type. Waters from the mylonitized quartz-feldspar gneiss have a higher average uranium concentration. Reduced ground water, restricted to two narrow zones of hornblende gneiss within the Boyertown study area, contains uniformly lower uranium concentrations (<1 picocurie per liter). Therefore, aquifer rock type appears to be a dominant factor in determining the radon-222 potential of ground water in the two sites studied in the Reading Prong.

INTRODUCTION

Although the issue of indoor radon and its sources has received considerable attention, there is still little information concerning the source of radon-222 (^{222}Rn) dissolved in ground water. Most studies of ^{222}Rn in ground water have been aimed at characterizing ^{222}Rn values for statistical or geochemical exploration purposes (Dyck and Tan, 1978; Prichard and Gesell, 1983; Loomis, 1987) or at understanding the hydrologic processes governing ^{222}Rn availability (Brutsaert and others, 1981; Rumbaugh, 1983; Wathen, 1986). Few studies have been performed in which specific chemical processes have been examined for the role they may play in controlling ^{222}Rn availability. This paper presents the results of a field study designed to understand the source of the radionuclides uranium-238 (^{238}U), radium-226 (^{226}Ra), and ^{222}Rn in domestic ground-water supply wells from two areas in the Reading Prong physiographic province of eastern Pennsylvania.

A thorough understanding of radionuclide mobility can be derived from a detailed examination of the overall ground-water quality. Therefore, major geochemical parameters of the ground-water samples were measured, including pH, dissolved oxygen, and the concentrations of major and trace cations and anions. A key feature of this study is the examination of a broad range of geochemical processes that affect ground-water quality as well as the mobility of ^{238}U, ^{226}Ra, and ^{222}Rn. Insights into the availability of ^{222}Rn in ground water may be gained from studying these geochemical processes. A detailed summary of the geochemistry of the radionuclides is presented in an earlier paper in this volume (Wanty and Schoen, this volume).

GEOLOGY AND GROUND-WATER CHEMISTRY OF THE STUDY AREAS

The study was performed in two areas in the Reading Prong (figs. 120 and 121), near Boyertown and Easton, Pa. The geology of the two areas is dominated by a complex sequence of metamorphic rocks of varying grade and composition. Structural features such as folding, faulting, and foliation further complicate the geologic interpretation. Specific geologic characteristics of the two areas will be summarized briefly here; for a more detailed description of the geology, the reader is referred to Drake (1967), Gundersen and others (1988), and Gundersen (this volume).

Rocks in the Boyertown study area (fig. 120) constitute a metasedimentary sequence including interlayered biotite and quartz-feldspar gneisses (QFG), and hornblende-pyroxene gneisses. The more mafic gneisses contain relatively low amounts of uranium and may contain sulfide minerals such as pyrite. Ground water in the mafic gneisses typically has low or below-detection concentrations of dissolved oxygen, high iron concentrations (up to several parts per million), variable pH, low levels of uranium, and variable ^{222}Rn concentrations. The QFG contains abundant uranium, sometimes in discrete grains of uraninite or in uraniferous minerals such as allanite, monazite, or sphene. The QFG has been severely mylonitized, and ground-water flow through this rock unit is restricted to fractures. Ground water in the QFG has a generally low pH (5.2–6.2), a high dissolved oxygen content, and high values of dissolved uranium and ^{222}Rn. Weathered surfaces of the QFG typically are iron stained.

The Easton study area (fig. 121) contains gneisses of variable composition, which represent a metasedimentary sequence. The rock units present in the area include interlayered quartz-feldspar and biotite gneisses, sillimanite gneiss, and marble. The QFG are enriched in uranium, which is contained in uraninite or other exotic uraniferous minerals. Limestones of the Lower and Middle Cambrian Leithsville Formation underlie the QFG and crop out to the north of them. Most of the homes in Easton are connected to a community water supply; only six private domestic wells were sampled in this area. All of the sample sites appear within the outcrop of the QFG on the geologic map (fig. 121). Ground water in the QFG generally has a low pH, a high dissolved oxygen content, and high uranium, ^{226}Ra, and ^{222}Rn contents. One of the well waters sampled had a higher pH (6.7), alkalinity (100 ppm (parts per million) HCO_3), and total dissolved solids (TDS; nearly twice the average of the other samples), all of which indicate a mixed origin for this water between the QFG and the underlying limestone.

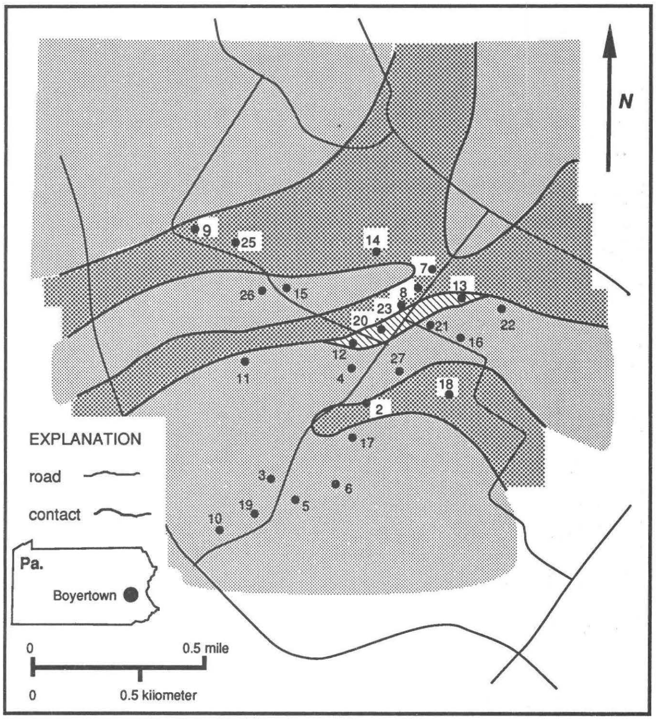

Figure 120. Geology of the Boyertown study area. Dots show the sample locations. Numbers correspond to table 56. The light and intermediate stippled patterns represent various mafic gneisses. The hachured pattern near the center is the mylonitized quartz-feldspar gneiss unit. For a detailed description of the geology of the Boyertown area, see Gundersen and others (1988).

METHODS OF SAMPLING, ANALYSIS, AND DATA TREATMENT

Samples were collected in the field from privately owned domestic-supply wells. Each well was pumped while temperature and specific conductivity were monitored. When these two quantities had stabilized (usually 10–20 minutes), the sampling procedure was begun. Field analyses were performed for concentrations of dissolved oxygen and ^{222}Rn, as well as pH, temperature, specific conductivity, and alkalinity. A summary of the methods used and detailed references for each method are listed in table 55. In addition to the field analyses, samples were collected for later laboratory analysis. These samples first were passed through a 0.45-μm (micrometer) filter and then collected in polyethylene bottles. The samples were acidified to pH <1 with concentrated nitric acid and returned to the laboratory for analysis of major and trace cations and major anions. The cations were analyzed by inductively coupled plasma atomic emission spectroscopy (Lichte and others, 1987), and the anions were analyzed by ion chromatography (Fishman and Pyen, 1979). The chemical data were entered into the program PHREEQE (Parkhurst and others, 1980), which was used to calculate the equilibrium distribution of aqueous species and to determine the saturation state of various phases in the ground water. The saturation state of the water with respect to a given solid phase is quantitatively expressed as the saturation index (SI), defined as the logarithm of the ratio of the ion activity product of the solid to its solubility product constant. Positive values of SI indicate supersaturation of a water with respect to the solid, negative values indicate undersaturation, and an SI of zero indicates equilibrium. For further details of the method of calculation and theoretical fundamentals, see Truesdell and Jones (1974) and Parkhurst and others (1980).

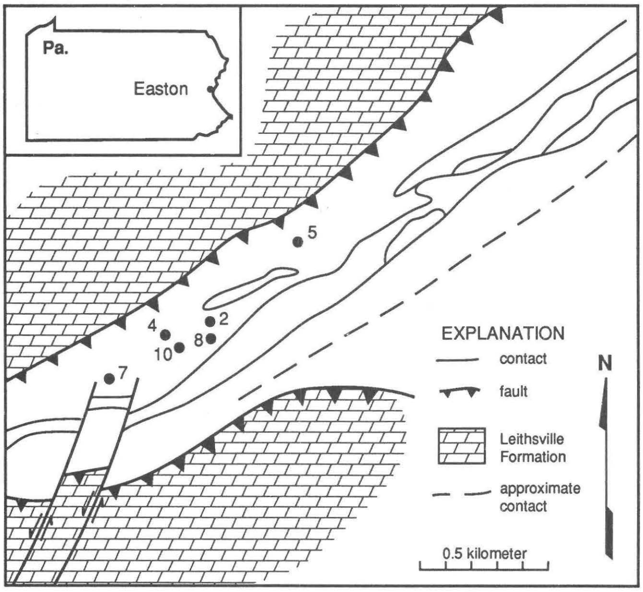

Figure 121. Geology of the Easton study area. Dots show the sample locations. Numbers correspond to table 57. All samples studied lie within a quartz-feldspar gneiss unit. For details of the geology, see Drake (1967) and Gundersen and others (1988).

Table 55. Field methods used during ground-water sampling in the Reading Prong, Pa.

Property	Method	Reference
pH	Electrode	Wood, 1981.
Conductivity	Conductivity bridge	Wood, 1981.
Alkalinity	Titrimetric	Skougstad and others, 1979.
Dissolved O_2	Electrode	Wood, 1981.
Dissolved H_2S	Precipitation with zinc acetate, gravimetric determination.	Skougstad and others, 1979; Tuttle and others, 1986.
Dissolved ^{222}Rn	Degassing followed by scintillometry.	Reimer, this volume.

RESULTS AND DISCUSSION

Boyertown Study Area

The Boyertown study area provided the greatest number of wells, with an approximate sample density of 8 per square kilometer. Twenty-five wells, representing several aquifers, were sampled in the Boyertown area; analytical results for the water samples are presented in table 56. Although the water samples were of generally good quality (all meet Environmental Protection Agency drinking water standards for TDS), many of the wells contain high levels of the various radionuclides. The highest ^{222}Rn values occur in waters sampled from mylonitized zones within the QFG (figs. 122 and 123).

The QFG contains anomalously high uranium, particularly in mylonitized zones. The residence of this uranium is either in discrete uranium minerals or incorporated in other minerals such as sphene. Uranium also is adsorbed onto the iron oxides on weathered surfaces. Ground-water samples from the QFG contained the highest concentrations of dissolved oxygen (fig. 124). Because uranium is most soluble in oxidized waters (Langmuir, 1978), high dissolved uranium concentrations are expected in water from the QFG (fig. 125). Furthermore, the saturation indexes of uraninite, a reduced uranium solid (UO_2), are lowest in water samples drawn from this rock unit (fig. 126). Water samples from the hornblende gneiss had low uraninite SI values because those rocks contain much lower amounts of leachable uranium. In addition, the waters in the QFG had the lowest pH values observed in the Boyertown study area. This effect is due in part to the oxidation of pyrite in the rocks. The iron released to the solution during this oxidation reaction reprecipitates as ferric oxyhydroxide. The iron oxides are effective adsorbents for uranium when the ground water has pH values above 5 to 6 and low total carbonate concentrations (Hsi, 1981). Qualitatively, other substrates, such as clay minerals, should behave similarly. In the waters that have the lowest pH (<6), particularly in the mylonitized zones, high concentrations ($>$ 1 ppb (part per billion)) of dissolved uranium are observed because adsorption reactions are less effective at limiting uranium mobility. Nevertheless, the observed uranium concentrations are still many orders of magnitude lower than the ^{222}Rn concentrations.

Radium-226 values in the Boyertown ground-water samples were generally low; all but one sample tested below 1 pCi/L. The highest value (4.1 pCi/L) was found in the mylonitized QFG. Iron oxides are abundant on weathered surfaces of the QFG (whether mylonitized or not) and effectively adsorb the aqueous radium (Ames and others, 1983 a,b,c). Therefore, ^{226}Ra levels in the ground waters probably are limited by adsorption processes. Dissolution of biotite or pyrite in the rocks is a probable source for dissolved iron. Iron oxide minerals then precipitate as the aqueous Fe^{2+} is oxidized to Fe^{3+}. Calculations made by use of the computer programs BALANCE (Parkhurst and others, 1982) and PHREEQE (Parkhurst and others, 1980) suggest that dissolution of both of these phases is necessary to produce the observed iron concentrations.

The highest concentrations of ^{238}U, ^{226}Ra, and ^{222}Rn all occur in waters drawn from the mylonitized QFG. However, they are not in secular equilibrium. Dissolved ^{222}Rn concentrations are several orders of magnitude higher than would be expected if secular equilibrium were established in the ground water alone (table 56). Therefore, virtually all of the dissolved ^{222}Rn is derived from ^{226}Ra and ^{238}U in the solid phase. It is logical to assume that the dominant control on ^{226}Ra (and, to some extent, ^{238}U) solubility is adsorption onto mineral surfaces at the

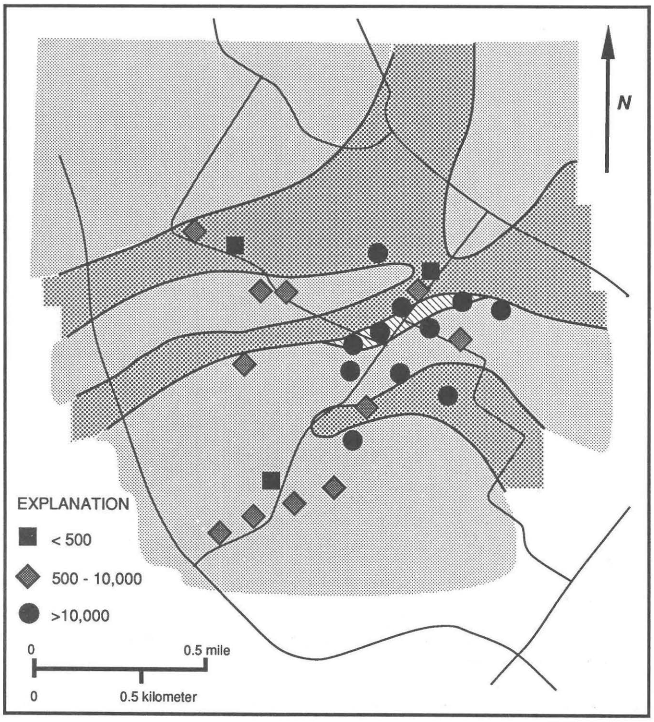

Figure 122. Radon-222 concentrations in ground water of the Boyertown study area. All values are in picocuries per liter; actual values are listed in table 56. Patterns depict lithologies as in figure 120.

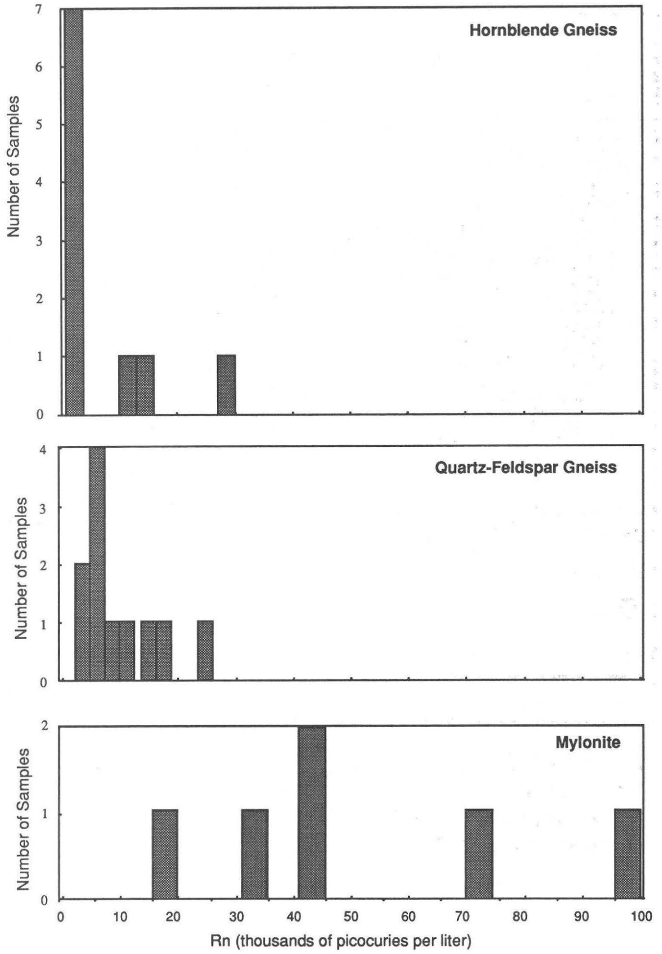

Figure 123. Dissolved-radon-222 concentrations in ground water from three aquifers in the Boyertown study area.

aqueous-solid interface. Thus, the ^{222}Rn concentrations in the ground water are unsupported by the parents in the water, although the parents are concentrated in the solid phase. The implications of this observation are extremely important: rocks having the highest uranium concentrations generally have the highest concentrations of ^{222}Rn in the ground water. In order for this statement to be true, a portion of the solid-phase uranium must be located near the rock-water interface. This condition is met if the uranium and radium are adsorbed on mineral surfaces rather than bound in crystal structures.

Although the waters themselves are not in secular equilibrium, the local water-rock system may be. Chemical evidence from rocks and soils in the Boyertown area suggests that this may be the case. Analyses of rock samples collected in Boyertown indicate that ^{238}U and ^{226}Ra contents are within the correct order of magnitude to support the observed ^{222}Rn in the ground water (Gundersen and others, 1988). In support of this argument, Rumbaugh (1983, 1987) successfully modeled ^{222}Rn concentrations in ground water when assuming secular equilibrium in the local water-rock system.

Easton Study Area

Sampling from the Easton study area was limited by the small number of homes having wells; most Easton residents are connected to a municipal water supply. Six wells were sampled in the area, all of which were situated in the same geologic unit (fig. 121). Indeed, the overall water chemistry is similar for these wells, with the exception of site 5. It is apparent from the ionic strength, pH, and ^{222}Rn and bicarbonate concentrations (table 57) that well 5 is drawing some portion of its water from the Leithsville Formation limestones, which underlie the main QFG body.

Except for site 5, the Easton samples closely resemble the samples drawn from the QFG in Boyertown, and many of the observations and conclusions derived from the Boyertown QFG samples also apply to Easton. The Easton ground water generally is of good quality, except for the high radionuclide concentrations. The Easton ground water is characterized by high concentrations of dissolved oxygen, low pH, low TDS, and high dissolved ^{222}Rn. As in Boyertown, dissolved ^{222}Rn is almost completely unsup-

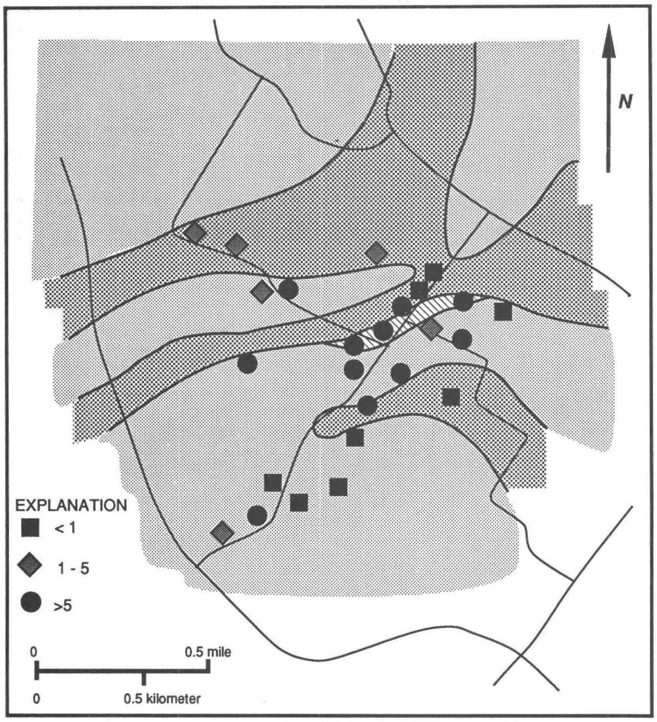

Figure 124. Dissolved-oxygen concentration in the Boyertown ground water. All values are in parts per million, actual values are listed in table 56. Patterns depict lithologies as in figure 120.

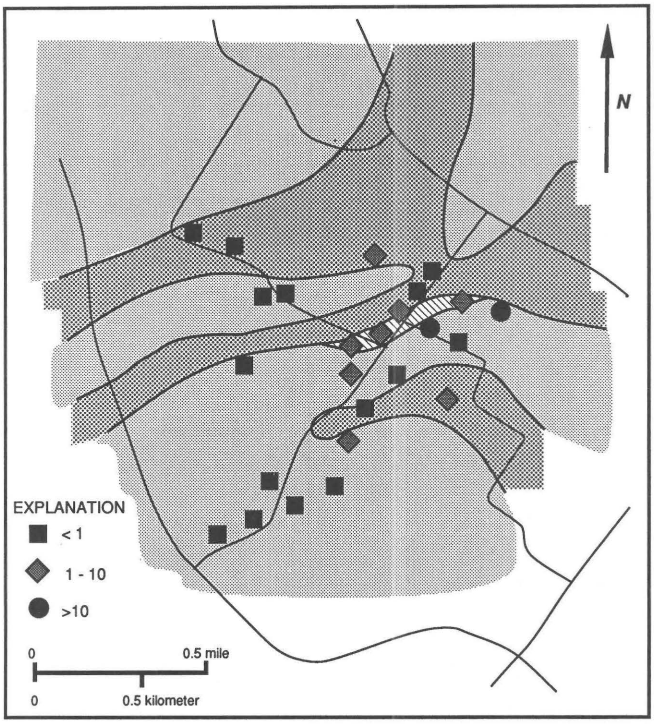

Figure 125. Dissolved-uranium concentration in the Boyertown ground water. All values are in parts per million, actual values are listed in table 56. Patterns depict lithologies as in figure 120.

ported by dissolved ^{238}U and ^{226}Ra. Nevertheless, the rock-water system, taken as a whole, may be in secular equilibrium. As in the Boyertown study area, ^{226}Ra and ^{238}U contents of the aquifer rocks are within the right order of magnitude to support the dissolved ^{222}Rn (Gundersen and others, 1988).

Total uranium concentrations in the Easton ground waters ranged from 0.5 to 14 ppb (table 57). Radium-226 concentrations ranged from 0.2 to 10.1 pCi/L. Uranium-238 and ^{226}Ra approach secular equilibrium in some cases, although this is probably fortuitous. Uraninite is extremely undersaturated in every sample from the Easton area (maximum uraninite SI value is -7.1; most are less than -23). Therefore, reduced uranium solids do not control the mobility of uranium in this ground water. Similarly, no radium solids approach saturation in these waters. Two processes may limit ^{238}U and ^{226}Ra concentrations in these waters: slow kinetics of dissolution of uranium and radium minerals, or, more likely, the adsorption of both elements onto various aquifer materials. Iron oxides and quartz probably provide the majority of adsorption sites. Iron oxides are less abundant but more effective adsorbents (Hsi, 1981; Ames and others, 1983a,b,c). Quartz, although a weak adsorbent, is much more abundant in the rock. On the other hand, ^{222}Rn is weakly adsorbed onto mineral surfaces, if at all. Thus, radon is partitioned into the aqueous phase, and its concentration is high relative to that which would be expected if the water itself were in secular equilibrium.

CONCLUSIONS

Radionuclide concentrations in ground waters from crystalline aquifers in two sites in eastern Pennsylvania are controlled primarily by aquifer lithology. Within the geologic framework, specific chemical processes, including adsorption/desorption and mineral dissolution and precipitation, act to control the concentrations of the individual elements. Weathering reactions that take place during recharge of oxygenated ground waters include pyrite oxidation and dissolution of uranium-bearing minerals. Iron released during pyrite oxidation later reprecipitates as a ferric oxyhydroxide. This ferric oxyhydroxide is an effective adsorbent for ^{238}U and ^{226}Ra. Uranium concentrations are highest in oxidizing ground water but only in rocks that contain leachable uranium. For the Boyertown and Easton study areas, the highest dissolved uranium concentrations occur in uraniferous QFG. Mylonitized areas within the QFG are enriched in uranium over the surrounding rocks, and waters in this aquifer have particularly high dissolved ^{222}Rn concentrations. Radium-226 concentrations are highest in low-pH ground waters, where adsorption onto min-

Table 56. Chemical analyses of the Boyertown study area well waters

[Units are as follows: Temp., temperature in degrees Celsius; SpC, specific conductance in micromhos per centimeter; DO (dissolved oxygen), TDS (total dissolved solids), SO_4, HCO_3, Cl, Ca, Mg, Na, K, Si, and Fe, all in parts per million; Rn, Ra, both in picocuries per liter; U, B, Mn, Cu, Zn, Li, Ba, Sr, and Cr, all in parts per billion. Rock type: QFG, quartz-feldspar gneiss; HG, hornblende gneiss; and My, mylonitized quartz-feldspar gneiss. na, not analyzed; nd, not detected]

Site no.	Rock type	Temp.	SpC	DO	pH	Rn	Ra	U	TDS	SO_4	HCO_3	Cl	Ca	Mg	Na	K	Si	B	Fe	Mn	Cu	Zn	Li	Ba	Sr	Cr
1	QFG	13.6	na	na	5.95	4,300	na	na	na	na	na	na	na	na	na	na	na	na	na	na	na	na	na	na	na	na
2	QFG	13	190	9.2	6.05	3,900	.2	.18	112	32	38	nd	11.9	5	7.3	nd	17.8	nd	.02	nd	nd	13	4	129	67.9	2
3	HG	12	360	.3	6.75	100	.2	.36	214	59	89	nd	28.2	10.1	8.2	1	16.4	40	1.73	115	nd	8	7	93	71.1	1
4	QFG	13	240	5.5	6.3	16,600	.2	6	159	43	57	10	21.4	7.03	6.3	nd	14.2	30	.02	7	nd	7	nd	2	62.6	1
5	HG	12.4	310	.3	6.65	1,200	.2	0	200	50	84	nd	20.1	9.46	7.4	nd	20.1	80	9	103	nd	26	9	14	55	1
6	HG	12	222	.22	6.45	670	.2	.18	164	46	63	nd	19.6	7.09	6.1	1	18.2	nd	3.24	77	nd	15	6	32	47.6	1
7	HG	13.3	336	.3	7.9	120	.1	.14	216	50	97	nd	30.8	8.13	9.1	1	19.8	90	.07	98	nd	nd	5	10	116	1
8	HG	13.7	280	.1	8.95	1,400	.1	.18	167	16	95	nd	4.16	.48	40.6	nd	11.1	130	nd	4	nd	nd	5	nd	20.7	1
9	HG	12.3	200	2.9	6.05	1,200	.1	0	117	31	43	nd	13.5	6.83	5.1	nd	17.1	nd	.63	23	10	42	nd	14	51.5	1
10	QFG	13	258	4.8	6.65	3,700	.1	.33	194	52	79	nd	35.7	6.82	5.7	1	13.5	nd	nd	nd	nd	5	nd	14	110	1
11	QFG	12	180	8.1	6.15	5,500	.2	.33	107	25	40	nd	12.2	4.59	5.7	nd	19.4	nd	.04	1	nd	6	nd	nd	40.9	2
12	My	13.5	201	8.4	6.05	41,000	.3	6.55	112	36	35	nd	13.2	6.22	7.9	nd	14	nd	.02	nd	nd	5	nd	nd	51.4	1
13	My	13.6	300	9.3	5.95	34,000	.5	4.59	127	31	19	30	17.9	8.13	9.6	1	10.1	10	nd	nd	10	147	nd	nd	82.2	nd
14	HG	12.4	370	3.6	6.65	11,100	.2	2.06	214	49	98	nd	31.4	9.73	8.6	nd	17	10	nd	46	nd	10	5	7	63.9	1
15	QFG	13	282	7.6	5.95	8,200	.1	.14	137	53	34	nd	15.8	8.82	9.4	nd	16.4	nd	nd	nd	nd	10	5	16	67.7	1
16	QFG	13.7	280	8.7	5.85	6,300	.2	.48	178	44	35	35	23.2	10.4	13.1	1	15.8	30	.02	2	nd	10	5	4	110	1
17	HG	13.1	240	.69	7.65	28,800	.3	3.48	197	44	94	nd	28.1	8.42	7.6	nd	14.8	40	nd	5	nd	5	4	62	105	1
18	HG	13.1	185	.79	8.55	13,400	.1	1.82	164	38	70	nd	9.28	1.24	29	nd	16.3	100	nd	nd	nd	nd	6	nd	59.2	1
19	QFG	13.9	170	6.8	6.65	26,300	.3	.67	132	36	52	nd	15.5	4.63	5.6	nd	17.8	220	.07	13	nd	12	nd	4	74.6	1
20	My	13.5	130	11.1	6.1	12,000	.2	1.18	101	33	29	nd	12.2	4.46	7.3	nd	14.9	10	.05	nd	20	6	nd	nd	64.4	2
21	My	13.4	235	3.6	6.65	99,000	.6	58	187	42	68	20	27.5	8.89	9.1	nd	12	80	nd	12	nd	1,800	5	nd	83.7	1
22	My	13.1	262	.08	7.65	71,900	4.1	590	203	45	85	15	31.7	7.28	8.4	nd	10.6	80	.18	85	nd	624	5	2	48.1	1
23	My	13.1	182	7.8	5.95	39,600	.4	3.6	113	30	29	15	16	6.77	7.2	1	8.03	10	nd	3	10	20	nd	nd	82.8	2
24	QFG	na	na	na	na	14,200	na	na	na	na	na	na	na	na	na	na	na	na	na	na		na	na	na	na	na
25	HG	12.1	285	3.5	6.35	100	.1	.21	205	57	62	15	33.6	10.6	8.8	2	16.3	nd	nd	188	nd	14	nd	21	138	1
26	QFG	13.1	280	2.3	6.65	1,400	.2	.33	185	54	62	10	23.3	10.8	7.9	1	15.7	10	.04	20	10	53	5	22	72.3	1
27	QFG	12.3	145	9.8	6.05	11,300	.1	.05	106	35	32	nd	12.5	4.44	6.1	nd	16.4	10	.05	2	nd	12	nd	nd	45.5	1

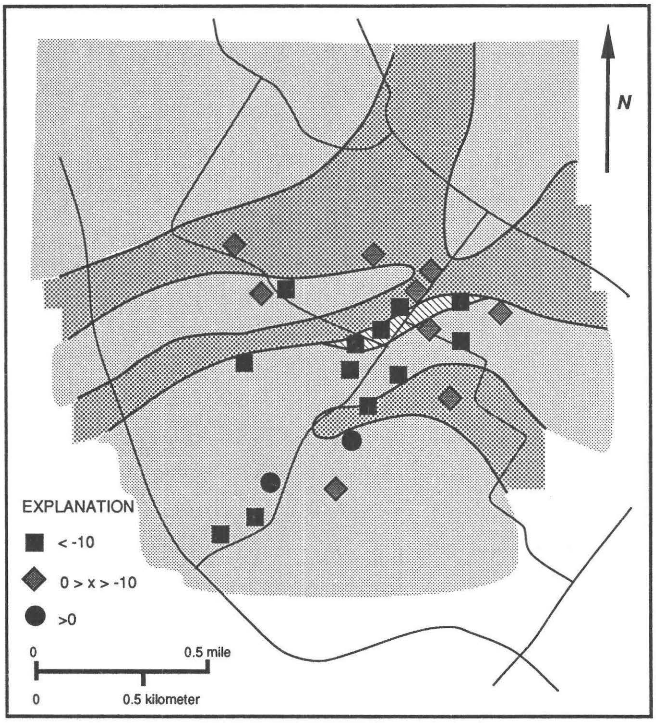

Figure 126. Uraninite saturation indexes for the Boyertown ground-water samples. Positive values indicate supersaturation with respect to uraninite, and negative values represent undersaturation. Actual values are listed in table 56. Patterns depict lithologies as in figure 120.

erals such as iron oxides and quartz is weakest. Groundwater concentrations of ^{222}Rn are much higher than expected, given the relatively low dissolved-^{238}U and dissolved-^{226}Ra concentrations. Uranium-238 and ^{226}Ra concentrations of the aquifer materials are high enough to support the dissolved ^{222}Rn concentrations. Therefore, although the waters themselves are not found to be in secular equilibrium, the overall water-rock system may be.

Specific chemical processes are active in every aquifer at the rock-water interface. Chemically, each phase impacts the others in some way. These processes are predictable from the basis of thermodynamics and kinetics. A detailed examination of these processes should lead to a better understanding of the overall water-rock system.

ACKNOWLEDGMENT

We would like to thank Susan L. Johnson for assistance with the field sampling.

Table 57. Chemical analyses of the Easton study area well waters

[Units are as follows: Temp., temperature in degrees Celsius; SpC, specific conductance in micromhos per centimeter; DO (dissolved oxygen), SO_4, HCO_3, Cl, Ca, Mg, Na, K, Si, Fe, and TDS (total dissolved solids), all in parts per million; Rn, Ra, both in picocuries per liter; U, B, Mn, Cu, Zn, Li, Ba, Sr, and Cr, all in parts per billion; nd, not detected]

Site no.	Temp.	SpC	DO	pH	Rn	Ra	U	SO_4	HCO_3	Cl	Ca	Mg	Na	K	Si	B	Fe	Mn	Cu	Zn	Li	Ba	Sr	Cr	TDS
2	12.9	315	8.6	5.55	80,300	1.6	4.4	55	32	15	26	5.45	9.6	2	12.8	nd	.04	nd	50	8	nd	46	109	2	158
4	12.4	295	8.9	5.60	34,700	1.2	.5	49	16	30	28.5	6.04	9.4	2	10.5	10	.02	1	50	12	nd	65	117	1	151
5	11.1	470	3.6	6.70	10,600	.2	6.5	59	100	30	49.4	8.4	9.6	3	10.2	20	nd	nd	nd	3	7	112	87.8	1	270
7	12.2	240	9.0	5.80	23,400	10.1	12	55	27	10	18.2	4.84	5	1	7.09	nd	nd	nd	nd	4	nd	75	68.8	1	128
9	12.7	240	9.5	5.90	43,900	5.1	14	58	36	10	20.3	5.29	14.3	2	9.92	10	nd	2	30	8	nd	39	77	1	156
10	13.4	180	11.0	5.75	26,300	5.2	6.3	51	18	nd	16.5	4	7.2	2	9.5	10	.05	1	750	8	nd	52	76.7	1	108

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RADIONUCLIDES IN THE PUERCO AND LOWER LITTLE COLORADO RIVER BASINS, NEW MEXICO AND ARIZONA, BEFORE 1987

John R. Gray and Robert H. Webb

Abstract

Radionuclides in the Little Colorado River and tributaries have several sources in the Little Colorado River basin of Arizona and New Mexico. Naturally occurring radionuclides of the uranium-238 and thorium-232 decay series enter the hydrologic cycle through natural erosion. Uranium-mining operations in two areas of the basin—the Grants Mineral Belt of New Mexico and the Cameron Uranium Mining Belt of Arizona—also resulted in releases of radionuclides. A cumulative 22 years of mine dewatering between 1960 and 1986 from the Grants Mineral Belt upstream from Gallup, N. Mex., and a tailings-pond spill in 1979 resulted in releases of radionuclides to the Puerco River. Mine pits left open from wildcat operations in the 1950's and early 1960's in the Cameron Uranium Mining Belt now contain water that has elevated activities of radionuclides. The impact of mining on contamination of water resources of the region, however, is not well understood.

In Arizona, surface water in the Puerco River at Chambers and the Little Colorado River at Cameron typically contains total gross alpha plus gross beta activities of several thousand picocuries per liter, or about 2 orders of magnitude larger than Arizona's maximum allowable limit of 30 picocuries per liter for surface water. Gross alpha plus gross beta activities in streamflow at both stations are associated primarily with the suspended phase. Measured radionuclides include large activities of uranium, thorium, radium, and lead-210.

In December 1986, water from 5 of 14 wells that penetrate the alluvium adjacent to the Puerco River contained gross alpha minus uranium and radon activities equal to or greater than the Environmental Protection Agency's maximum contaminant level of 15 picocuries per liter for drinking water. Water samples from several wells, springs, and open pits near Cameron, Ariz., contain activities of gross alpha plus gross beta in excess of 30 picocuries per liter. If a "plume" of radionuclide-rich water exists in the alluvial aquifer underlying the Puerco or lower Little Colorado River, its areal extent cannot be determined from the data.

The presence and extent of radionuclide activities in the Little Colorado River basin are generally unknown. A lack of understanding of processes of radionuclide movement in and between surface water and ground water presently limits assessments of trends in radionuclide contamination.

INTRODUCTION

Mining activities in the Little Colorado River basin of Arizona and New Mexico have resulted in the dispersion of radionuclides and other potentially hazardous trace elements to the environment. Inhabitants of the sparsely populated drainage basin, which is largely on the Navajo Indian Reservation, rely on ground water for domestic supplies and on surface water for livestock watering and irrigation of forage crops.

This paper describes the occurrence of selected radionuclides in the Puerco and lower Little Colorado Rivers and in near-channel alluvial ground water in the Puerco River basin. The history and impacts of mining on selected water resources in this semiarid region are briefly described, and chemical and physical processes that probably control contaminant movement in surface water and ground water are reviewed.

BACKGROUND

The Little Colorado River basin covers 27,000 mi^2 (square miles) of the southern Colorado Plateau physiographic province. Eighty percent of the drainage area is in northeastern Arizona, and 20 percent is in northwestern New Mexico (fig. 127). The Little Colorado River heads in the White Mountains of east-central Arizona and descends 6,300 ft over 356 mi before joining the Colorado River at Grand Canyon National Park.

In 1975, the population of the Little Colorado River basin averaged six people per square mile. Principal communities are Flagstaff, Winslow, and Holbrook in Arizona and Gallup in New Mexico. The Navajo and Hopi Indian Reservations occupy most of the northern half of the basin (fig. 127).

The climate is arid in the lower elevations and semiarid to subhumid in the uplands. Data from the U.S. Soil Conservation Service (1981, Appendix 1, p. 1–2) indicate that mean annual precipitation in the basin ranges from less than 8 in near Cameron, Ariz., to as much as 32 in at the basin divide south of Winslow, Ariz. Daily average temperatures in the basin range from 26 to 80 °F. Surface altitudes range from 12,630 ft to about 2,730 ft above mean sea level. The Little Colorado River is an ephemeral stream except for about 20 mi in its headwaters, where flow is regulated, and within about 10 mi of its mouth (Loughlin, 1983). Peak runoff typically occurs in March and April from snowmelt and rainfall or in July and August from thunderstorms (U.S. Soil Conservation Service, 1981, Appendix 1, p. 1–2; Appendix 2, p. 5–13).

The Puerco River basin includes about 3,000 mi^2 of the northeastern part of the Little Colorado River basin (figs. 128 and 129). The Puerco River originates in the

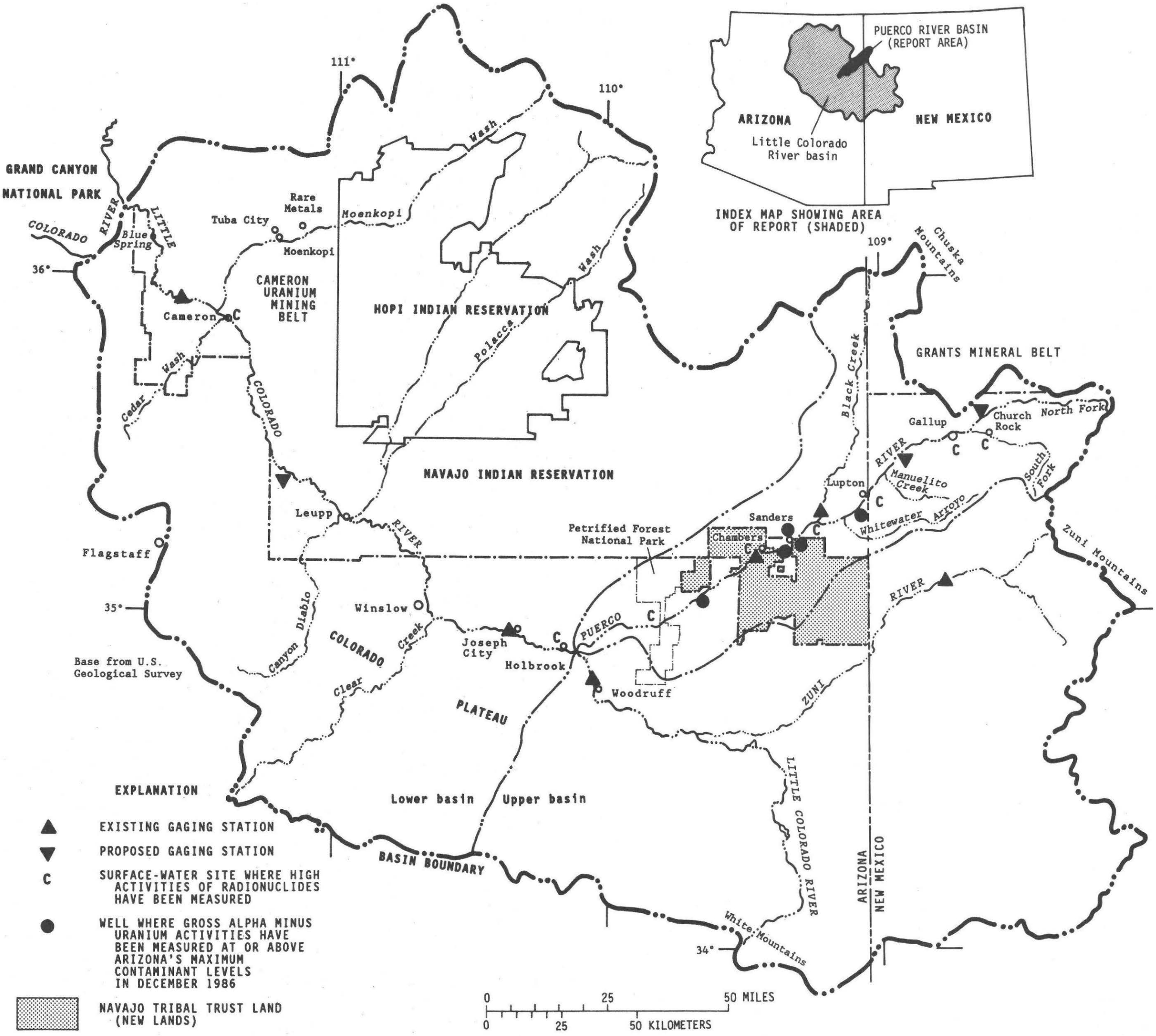

Figure 127. Little Colorado River basin.

Chuska and Zuni Mountains of New Mexico and flows southwest about 140 mi to join the Little Colorado River near Holbrook, Ariz. Peak runoff in the Puerco River usually occurs in response to summer thunderstorms that produce flash floods. Before 1950, the Puerco River was an ephemeral alluvial stream (Kaufmann and others, 1976). For most of the period between the 1950's and the mid-1980's, streamflow in the upper part of the basin (fig. 128) changed from ephemeral to perennial from near Church Rock, N. Mex., to downstream from the State line of Arizona and New Mexico. The principal sources of perennial streamflow were effluent from dewatering of uranium mines northeast of Gallup, N. Mex., and effluent discharged from the sewage-treatment plant in Gallup. In the absence of runoff, all effluent typically infiltrated the alluvium within a few miles downstream from Chambers, Ariz. (Shuey, 1986).

Geology

The Little Colorado River basin is geologically complex, and the regional stratigraphic sequence of sedimentary rocks is structurally deformed. Bedrock exposed in the basin ranges in age from Precambrian to Quaternary although exposures of Precambrian and Cambrian rocks are limited. Rocks of Cretaceous age, which include outcrops of Dakota Sandstone and Mancos Shale, are the most extensive and are the thickest sequence of any rock formations in the New Mexico part of the basin. Extensive volcanic activity began in the Tertiary Period and continued

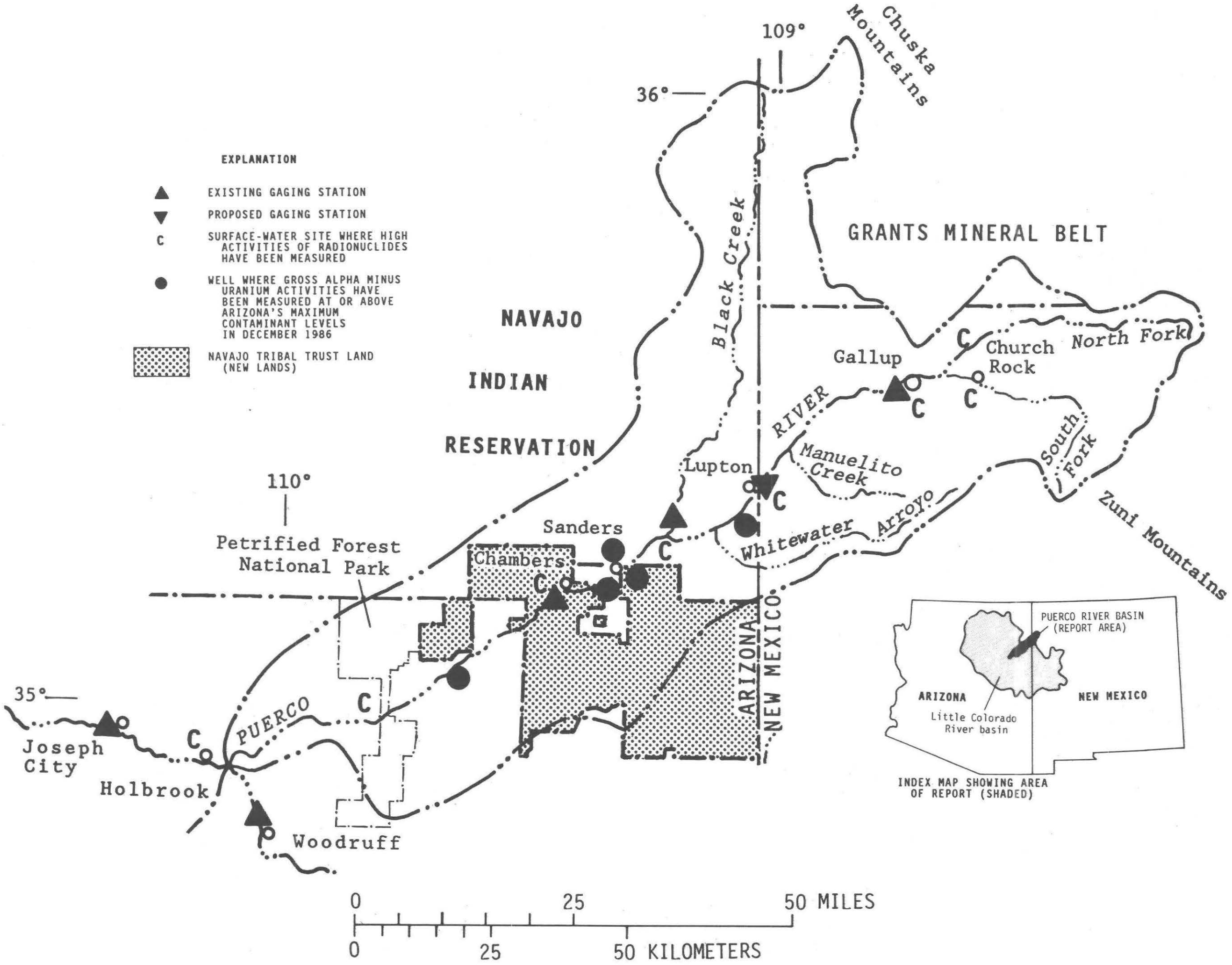

Figure 128. Puerco River basin.

well into the Quaternary Period. Volcanic rocks include rhyolite, dacite, andesite, and latite. Quaternary sediments consisting of sand, silt, and gravel are present throughout most of the basin (U.S. Soil Conservation Service, 1981, Appendix 1, p. 1–11 to 1–15).

A large variety of ore deposits occurs in the Little Colorado River basin. Uranium, coal, bentonite, vanadium, sand, and gravel are commercially important deposits in the basin. In Arizona, uranium is found principally in sandstones and conglomerates of continental origin near Cameron, Chinle, and Round Rock (U.S. Soil Conservation Service, 1981, Appendix 1, p. 1–15 to 1–18).

The bedrock in the Puerco River basin consists of Paleozoic and Mesozoic sedimentary rocks that dip slightly to the northeast. Steeply dipping fault zones that trend north and south displace these rocks. The Defiance and Zuni Uplifts occur to the north and east, respectively (Cooley and others, 1969). Rocks of Tertiary and Quaternary age, including the Tertiary Bidahochi Formation, generally are undeformed. Major uranium deposits and minor coal deposits are present in the Jurassic Morrison Formation to the east and northwest of Gallup (Hackman and Olson, 1977). Uranium deposits also occur in the Petrified Forest Member of the Triassic Chinle Formation in the western half of the Puerco River basin. Chemical and physical weathering of exposed bedrock that contains uranium and other radionuclides is the source for background radiation in the regional water resources and sediments (Weimer and others, 1981).

The alluvial aquifer of the Puerco River consists of interbedded gravel, sand, silt, and clay (Mann and Nemecek, 1983). The thickness of the aquifer is unknown, but several wells in the alluvial aquifer are 200 ft deep and do not encounter bedrock. The stratigraphy and variability of the sediments that compose the alluvial aquifer in the Little Colorado River basin are unknown.

Standards for Radionuclides in Water

Webb and others (1987b) list maximum contaminant levels (MCL's) and maximum allowable limits (MAL's)

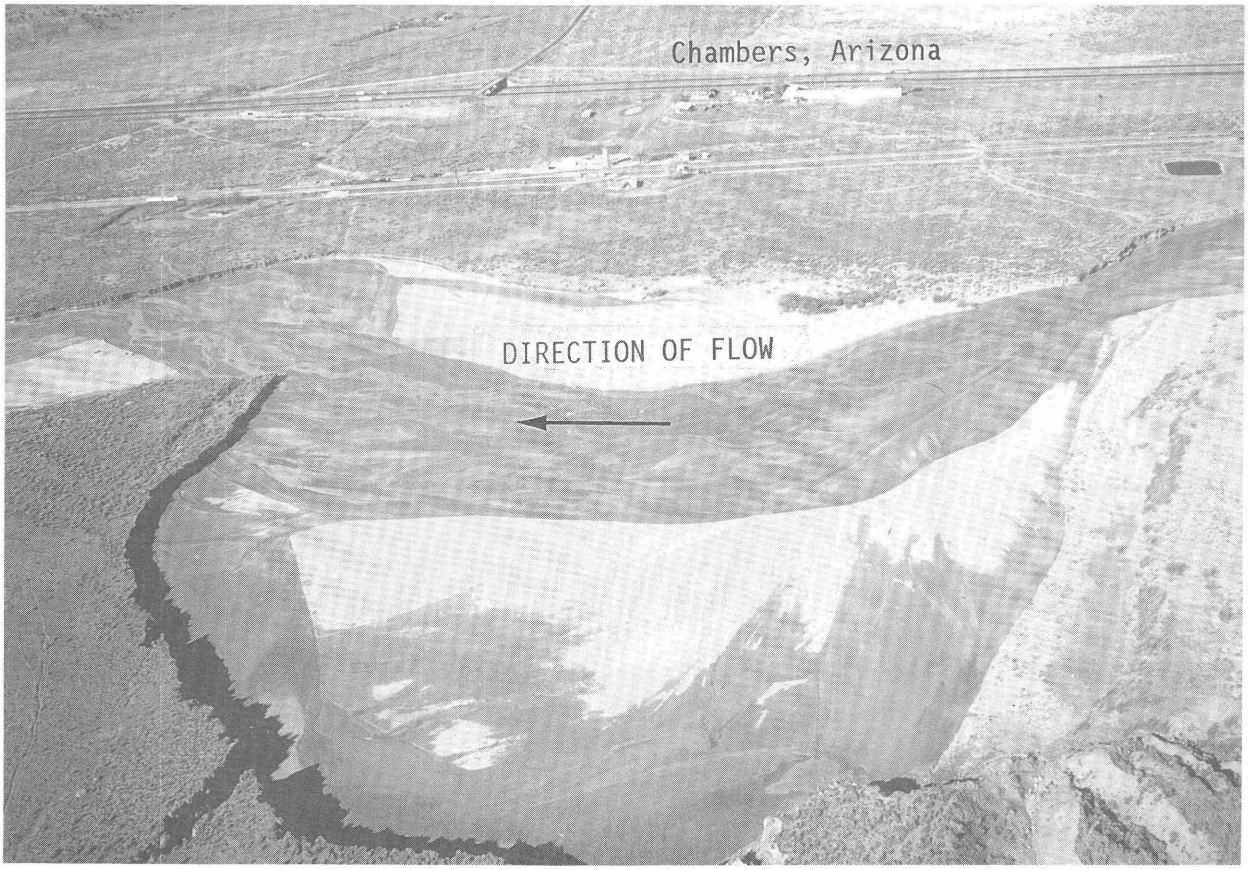

Figure 129. Aerial view of the Puerco River basin. The Puerco River is an ephemeral stream in northeastern Arizona and northwestern New Mexico that has been contaminated by radionuclides and other trace elements. The contaminants are sorbed mostly in channel sediments.

applicable for radionuclides in Arizona ground water and surface water, respectively. The amount of radium-226 (^{226}Ra) in water supplies forms a basis for monitoring natural radionuclides in drinking-water supplies in the United States (Lappenbusch and Cothern, 1985). The MCL is 5.0 pCi/L (picocuries per liter) for total ^{226}Ra plus ^{228}Ra. Total ^{226}Ra plus ^{228}Ra are the only natural radionuclides that have MCL's (McClennan, 1984; Environmental Protection Agency, 1986). The MCL for gross alpha activity minus uranium and radon is 15 pCi/L, and the MCL for total gross beta activity is 50 pCi/L. Although an MCL has not been established for uranium, 35 μg/L (micrograms per liter) of uranium is a recommended limit (Lappenbusch and Cothern, 1985). State of Arizona regulations (State of Arizona Atomic Energy Commission, 1977) also mandate an MCL of 30 pCi/L if the identity or activity of any radionuclide in a mixture of radionuclides in water is unknown.

The MAL for both dissolved ^{226}Ra and dissolved ^{228}Ra is 30 pCi/L. However, the MAL for total ^{226}Ra plus ^{228}Ra is 5 pCi/L. Other MAL's include 45 pCi/L for dissolved plus suspended uranium and 30 pCi/L for gross alpha plus gross beta activity (McClennan, 1986).

History of Radionuclide Releases

Grants Mineral Belt, Puerco River Basin

Effluent from dewatering of uranium mines was discharged to Pipeline Arroyo, a tributary of the Puerco River northeast of Gallup, from 1960 to 1961 and 1967 to 1986 (Perkins and Goad, 1980). Effluent discharges contributed radionuclides to the Puerco River; however, the amount of radionuclides in effluent water from mining activities before 1967 is unknown.

Permits under the National Pollutant Discharge Elimination System (NPDES) were required when the mines reopened in 1969 in an attempt to control the level of radioactive material in effluent water (Environmental Protection Agency, 1975). Under the conditions of the permits, effluent water could contain as much as 3 pCi/L of dissolved ^{226}Ra, as much as 10 pCi/L of total ^{226}Ra, and as

Figure 130. Church Rock tailing pond. The Church Rock Mill was the site of the largest accidental release of uranium tailings liquid in United States history. In July 1979, approximately 94 million gallons of liquid and more than 1,100 tons of tailings were released into the Puerco River. The total activity of gross alpha in the Puerco River on the day of the spill was about 130,000 picocuries per liter. The recommended limit of gross alpha in Arizona surface water is 30 picocuries per liter.

much as 2.0 mg/L of total uranium (Chris Shuey, Southwest Research and Information Center, Albuquerque, N. Mex., written commun., 1987). Shuey (1986) reported 63 violations of the NPDES permits between 1980 and 1983.

Additional discharges of radionuclides into the Puerco River resulted from a tailings-pond dam failure at the United Nuclear Corporation's Church Rock Mill on July 16, 1979 (fig. 130) (Weimer and others, 1981; Shuey, 1982; Millard and others, 1984). An estimated 94 Mgal (million gallons) of liquid were released into the Puerco River (Weimer and others, 1981). The liquid contained an estimated 18,000 tons of suspended sediment in addition to 1,100 tons of tailings eroded from the bottom of the tailings pond (Donald Hendricks, Environmental Protection Agency, written commun., 1982). The total amount that entered the Puerco River is probably less than these amounts because of retention of sediments behind an emergency catchment dam and postspill cleanup efforts (Weimer and others, 1981). A liquid sample taken from the tailings pond before the spill had total activities of 210 pCi/L of ^{226}Ra and 10,225 pCi/L of thorium-230 (^{230}Th). The pH of the liquid was 1.9, and the concentration of uranium was about 4 mg/L (Weimer and others, 1981).

The mines continued to discharge effluents as allowed under the NPDES permits after the spill of 1979. Activities of gross alpha and ^{226}Ra were as large as 400 and 2.5 pCi/L, respectively, in two samples from New Mexico measured in October 1981 (B.M. Gallaher, New Mexico Environmental Improvement Division, written commun., 1982). Activities of lead-210 (^{210}Pb) and polonium-210 (^{210}Po) ranged from 4.5 to 10 pCi/L, and 3.4 to 10 pCi/L, respectively, and activity of ^{230}Th ranged from 0.1 to 3.9 pCi/L (B.M. Gallaher, written commun., 1982). Treated mine water that was released into the Puerco River had gross beta activities that ranged from 320 to 660 pCi/L in 1982 (Shuey, Southwest Research and Information Center, written commun., 1987). Effluent from one mine contained total uranium concentrations that ranged from 1.5 to 2.7 mg/L and a mean total concentration of 1.6 mg/L during 9 months of 1982 (June Buzzell, Environmental Protection Agency, written commun., 1983).

Cameron Uranium Mining Belt, Lower Little Colorado River Basin

Most uranium ore in the lower Little Colorado River basin came from the Cameron Uranium Mining Belt (fig. 127). Uranium mining occurred within a curved belt approximately 2 mi wide extending 6 mi north of Cameron along U.S. Highways 89 and 164, and 5 mi wide extending 18 mi southeast along the Little Colorado River (Don Payne, Navajo Nation, written commun., 1987). Mining in this region began in the early 1950's, reached a production peak in 1957, and ceased in 1962. A total of 289,300 tons of spoil and more than 600 tons of uranium ore were extracted from 98 mining sites. Most ore was processed at Rare Metals near Moenkopi Wash (fig. 127; Don Payne, Navajo Nation, written commun., 1987).

No regulations for stabilization of abandoned mines existed in 1962. As a result, many pits in the Cameron Uranium Mining Belt are open and have filled with water. Don Payne (Navajo Nation, written commun., 1987) collected water samples from 21 uranium mining pits, 14 developed springs, 6 wells, 6 surface-water impoundments, and 2 other locations on the Little Colorado River in the mid-1980's and found elevated activities of gross alpha, gross beta, ^{226}Ra, and uranium.

Water-Quality Investigations Related to Radionuclide Releases

Puerco River Basin

Surface water in the Puerco River contained elevated concentrations of radionuclides and trace elements in the mid-1970's. In two samples taken at the State line of Arizona and New Mexico, one in 1975 and the other in 1976, gross alpha activities were 330 and 100 pCi/L, and gross beta activities were 1,640 and 81 pCi/L, respectively (W.J. Shelley, Kerr-McGee Nuclear Corporation, written commun., 1979). Between 1975 and May 1979, ^{226}Ra activities ranged from 0.1 to 22 pCi/L, and total uranium concentrations ranged from 0.2 to 0.9 mg/L.

The effect of discharges allowed under the NPDES permits and discharges in violation of permits on the quality of ground water in the Puerco River basin is unknown. Scott and Barker (1962) report median values of 0.008 mg/L for uranium and 0.1 pCi/L for radium in ground water in a region that includes the Puerco River basin. Kaufmann and others (1976) studied ground-water contamination near uranium mines and milling operations in the Puerco River basin upstream from Gallup (fig. 128). In 71 ground-water samples that Kaufmann and others (1976) collected near uranium mines throughout northwestern New Mexico, only 6 wells had activities of ^{226}Ra above 3.0 pCi/L. Gross alpha activities exceeded 15 pCi/L in water from 33 of 71 wells (Kaufmann and others, 1976). Although radionuclide activities in some of the samples are above current MCL's, Kaufmann and others (1976) concluded that none of the 13 wells sampled in the Puerco River basin were contaminated by radionuclides.

Water and sediments were sampled extensively throughout the Puerco River basin after the tailings-pond spill of 1979. Activities of ^{230}Th in sediments commonly exceeded 30 pCi/g as far as 40 mi downstream from the mine (Weimer and others, 1981). After the spill of 1979, ^{230}Th activity in sediments decreased with time to an average of 9 pCi/g in the same reach (Millard and others, 1984). In 1979, sediments collected from the Puerco River in Arizona at Chambers and Petrified Forest National Park (fig. 128) had activities of ^{230}Th between 1 and 8 pCi/g (E.K. Swanson, Arizona Department of Health Services, written commun., 1986). Webb and others (1987a) measured radionuclide activities equivalent to background activities in bed sediments collected from five sites along the Puerco River in December 1986.

On July 16, 1979, following the tailings-pond spill, radionuclide activities in unfiltered samples from the Puerco River were 6,910 pCi/L of uranium, 12,000 pCi/L of ^{230}Th, 1.0 pCi/L of ^{226}Ra, 260 pCi/L of ^{210}Pb, and 38 pCi/L of ^{210}Po at sites in New Mexico (Millard and others, 1984). The maximum gross alpha activity was 130,000 pCi/L on the day of the spill near the Church Rock Mill and 26,000 to 40,000 pCi/L at Gallup (Shuey, 1982).

After the spill of 1979, shallow wells in the alluvial aquifer of the Puerco River were monitored in New Mexico and Arizona. Wells in New Mexico near the Puerco River revealed some contamination by radionuclides (Gallaher and Cary, 1986). Gross alpha activity increased from a prespill average of 12 pCi/L to a postspill maximum of 304 pCi/L with concomitant increases in the concentrations of total uranium (Shuey, 1982). Increases in gross alpha, uranium, and sulfate also occurred in alluvial well water in Arizona (Shuey, 1982).

Since 1979, radionuclides and trace elements have been sampled sporadically in surface water of the Puerco River at Chambers (Webb and others, 1987a). Total gross alpha activities have fluctuated between 12 and 11,200 pCi/L between 1979 and 1985 in the Puerco River at Chambers. The Arizona Department of Health Services (1986a) measured 34 violations of MCL's in 11 samples of surface water at 5 sites on the Puerco River in Arizona in February, April, and June 1985. The violations included elevated activities of gross alpha, gross beta, and ^{226}Ra. During a flood on July 16, 1986, Chris Shuey (Southwest Research and Information Center, written commun., 1987) measured total gross alpha activities of 1,700 to 2,200 pCi/L and gross beta activities of 1,800 to 2,100 pCi/L in the Puerco River near Lupton. On July 31, 1987, total gross alpha and gross beta activities at this site were 3,200 and 3,500 pCi/L, respectively (Chris Shuey, Southwest Research and Information Center, written commun., 1987).

Sampling of surface and ground water in the Puerco River basin between 1975 and 1986 has indicated that MCL's and MAL's for radionuclides were exceeded many times. The source of the elevated concentrations may be traced either to mining or to natural sources. Natural activities of radionuclides in Black Creek, a tributary to the Puerco River that was unaffected by uranium mining (fig. 128), did not exceed MCL's of Arizona for surface water (Arizona Department of Health Services, 1986a; Chris Shuey, Southwest Research and Information Center, written commun., 1987). No radionuclide data are available for surface water for the period before the mining activities of the 1950's or before the resumption of mining in 1969. The Arizona Department of Health Services (1986b) used a statistical model of flood-recurrence interval and suspended-sediment concentrations to suggest that ^{226}Ra activities in mine effluents alone were sufficient to cause violations of MAL's of Arizona for surface water in the Puerco River.

Lower Little Colorado River Basin

Little is known about radionuclide releases in the lower Little Colorado River basin. Arizona Department of Health Services (1986a) has measured elevated activities of gross alpha, gross beta, and ^{226}Ra in the Little Colorado River at Holbrook, just below the mouth of the Puerco River (fig. 128). Most radionuclides in the Little Colorado River probably emanate from the Puerco River. At Rare Metals (fig. 127), a contaminated mound of ground water underlies an unstabilized tailings pile near Moenkopi Wash, which is a tributary to the Little Colorado River (Carol Boughton, Navajo Nation, oral commun., 1987). Elevated activities of radionuclides near the mouth of the Little Colorado River may enter the river as springflow, originate in the Cameron Uranium Mining Belt (John McKlveen, Arizona State University, oral commun., 1988), or emanate from farther upstream. At least 56 springs enter the Little Colorado River within 30 mi of its mouth and may discharge over 200 ft^3/s (Loughlin, 1983). Gross alpha plus gross beta activities in five samples collected in 1985 and 1986 at Blue Spring ranged from 5 to 48 pCi/L. Radium-226 activities ranged from 0 to 1.6 pCi/L (Errol L. Montgomery and Associates, Inc., 1987).

RADIONUCLIDE ACTIVITIES AT U.S. GEOLOGICAL SURVEY GAGING STATIONS

Puerco River Near Church Rock, New Mexico

The U.S. Geological Survey gaging station near Church Rock (09395350) has a drainage area of 205 mi^2 and is 5 mi downstream from the United Nuclear Corporation facility and 9.5 mi northeast of Gallup. A water-quality sample was collected on July 17, 1979, and again on July 19, 1979, at this station (table 58). Discharges at times of sample collection were 7.1 and 8.0 ft^3/s, and pH was 3.6 and 5.2, respectively. Significantly smaller activities for dissolved gross alpha, dissolved gross beta, and dissolved ^{226}Ra on July 19 correspond with flushing of tailings-pond effluent from the reach by mine dewatering (table 58).

Puerco River at Gallup, New Mexico

The U.S. Geological Survey gaging station at Gallup (09395500) has a drainage area of 558 mi^2 and is in the city limits (fig. 128). Seventeen water-quality samples were collected by the U.S. Geological Survey at the gage from 1975 to 1984, and selected analytical results are shown in table 58. Maximum activities of both dissolved and suspended gross alpha and gross beta were measured on July 17, 1979, concomitant with a pH of 3.4 (table 58). Minimum activities measured for dissolved gross alpha and gross beta were 48 and 22 pCi/L, respectively, on August 3, 1983, and were associated with the maximum discharge (116 ft^3/s), which was measured when radiochemical data were collected. Minimum activities measured for suspended gross alpha and gross beta were 240 and 300 pCi/L, respectively. Dissolved ^{226}Ra activities ranged from 0.07 to 0.95 pCi/L. Dissolved uranium activities ranged from 18 to 2,100 pCi/L. Maximum levels of dissolved ^{226}Ra and uranium activities were both measured on July 17, 1979. Dissolved-solids concentrations measured on July 17 and 19, 1979, were 8,300 and 4,300 mg/L, respectively, or an order of magnitude larger than typical dissolved-solids concentrations for this site.

Puerco River at Chambers, Arizona

The U.S. Geological Survey gaging station at Chambers (09396100) has a drainage area of 2,160 mi^2 and is 1.5 mi southwest of Chambers (fig. 128). Twenty-nine water-quality samples were collected from 1979 to 1985 (Webb and others, 1987b), and selected analytical results are shown in table 58. Dissolved and suspended gross alpha and gross beta activities have fluctuated widely with time (fig. 131). Concentrations of total uranium, chloride, and sulfate fluctuated in 1979 apparently as a result of the tailings-pond spill (fig. 131). After 1980, changes in concentrations of these constituents are not well known because only three samples were measured for these constituents between May 1980 and May 1985. Concentrations of total uranium average about 0.1 mg/L between 1980 and 1985. The amount of dissolved uranium is not definitively known, but data for 1985 (Arizona Department of Health Services, 1986a) indicate that the concentration of dissolved uranium is 30 to 70 percent of the total uranium concentration. Activities of radionuclides, particularly those in the sus-

Table 58. Radionuclide activities and associated data of selected samples from U.S. Geological Survey gaging stations on the Puerco and lower Little Colorado Rivers, Arizona and New Mexico

[ft^3/s, cubic feet per second; μS/cm, microsiemens per centimeter; pCi/L, picocuries per liter; U-NAT, uranium natural; Cs-137, cesium-137; <, less than indicated value; ±, precision; dashes indicate no data available]

Date	Discharge (ft^3/s)	pH	Specific conductance (μS/cm)	Gross alpha		Gross beta		Radium-226, dissolved (pCi/L)	Uranium, dissolved (pCi/L)
				Dissolved (pCi/L as U-NAT)	Suspended (pCi/L as U-NAT)	Dissolved (pCi/L as Cs-137)	Suspended (pCi/L as Cs-137)		
Puerco River near Church Rock, N. Mex (09395350)									
July 17, 1979	7.1	3.6	3,450	560±110	5,400±1,100	150±30	1,800±360	1.0±.10	—
July 19, 1979	8.0	5.2	2,300	<18±4	2,800±560	<9.9±2	1,100±220	.47±.05	—
Puerco River at Gallup, N. Mex. (09395500)									
October 6, 1976	—	8.2	800	[1]1,900±380	[1]950±190	460±90	520±100	.84±.08	1,200±180
July 17, 1979	11	3.4	8,300	5,200±1,000	33,000±6,600	1,800±360	9,200±1,840	.95±.10	[2]2,100±320
July 19, 1979	11	7.2	4,300	<58±12	12,000±2,400	34±7	4,200±840	.40±.04	[2]55±8
December 18,1980....	4.8	8.5	800	750±150	240±50	110±20	300±60	.68±.07	[2]540±80
May 4, 1981.........	9.6	8.9	955	1,400±280	590±120	370±70	690±140	.33±.03	[2]1,100±160
December 7, 1982 ...	22	8.5	840	620±120	520±100	230±50	600±120	.13±.01	[2]820±120
August 3, 1983	116	7.8	—	48±10	1,800±360	22±4	1,700±340	.22±.02	[2]18±3
July 18, 1984	5.1	8.1	—	[1]680±140	[1]2,200±440	100±20	1,400±280	.15±.02	[2]630±94
Puerco River at Chambers, Ariz. (09396100)									
July 26, 1979	—	6.2	4,730	99±20	—	—	—	.20±1	—
May 18, 1982	.04	8.7	1,220	160±30	68±14	41±8	140±30	—	—
August 15, 1983	39	—	914	29±6	[1]<1,500±300	17±3	1,000±200	.20±1	[2]25±4
March 7, 1984	4.2	8.4	1,190	160±30	68±14	54±11	92±18	<.10±1	—
March 28, 1984	60	8.4	1,180	140±30	820±160	29±6	1,200±240	<.10±1	—
Little Colorado River at Cameron, Ariz. (09401200)									
June 6, 1980	1.4	8.4	1,280	<6±1	2±.40	7±1	2±.40	—	—
July 15, 1981........	231	8.0	1,190	13±3	3,900±780	12±2	3,200±640	—	—
March 1, 1982	1,340	8.2	445	20±4	1,700±340	10±2	1,700±340	—	—
November 30, 1983 ..	44	8.4	903	[1]<9±2	520±100	<8±2	560±110	—	—
April 9, 1984	16	8.4	1,750	54±11	820±160	<16±3	860±170	—	—

[1] Micrograms per liter were converted to picocuries per liter by multiplying micrograms per liter by 0.68. This conversion is based on the assumptions that uranium-234 is in equilibrium with uranium-238 and that all alpha emission is from uranium.

[2] Micrograms per liter were converted to picocuries per liter by multiplying micrograms per liter by 0.68. This conversion is based on the assumption that uranium-238 is in equilibrium with uranium-234.

pended phase, are related statistically to suspended-sediment concentrations at Puerco River at Chambers (Arizona Department of Health Services, 1986b; Chris Shuey, Southwest Research and Information Center, written commun., 1987).

Variations in measurable activities of radionuclides in streamflow in the Puerco River likely are caused in part by areal variability of tributary inflow. Tributary basins contain varying amounts of exposed uranium-bearing rock. Quality of runoff from tributaries depends partly on the amount of exposed uranium-bearing rock and amount and rate of precipitation or snowmelt. Variability of precipitation in the Puerco River basin, in conjunction with variability in amounts of exposed uranium-bearing rocks, results in highly variable discharges of radionuclides from tributaries to the main channel. Also, streamflow may cause remobilization of sediments deposited after the tailings-pond dam spill of 1979 (Arizona Department of Health Services, 1986b). This mechanism also may produce large fluctuations in the activity of radionuclides carried by the Puerco River into Arizona. Extensive monitoring of surface water in the Puerco River indicates that water recharging the alluvial aquifer in Arizona is of widely fluctuating quality. For example, dissolved gross alpha plus dissolved gross beta activities measured in samples from runoff in the Puerco River basin at Chambers, Ariz., ranged from 18 pCi/L to 273 pCi/L.

Little Colorado River at Cameron, Arizona

The Little Colorado River at the sampling site at Cameron has an approximate drainage area of 24,000 mi^2. Seventeen water-quality samples were collected and analyzed for gross alpha and gross beta activities from 1979 to 1984. Selected analytical results are shown in table 58, and

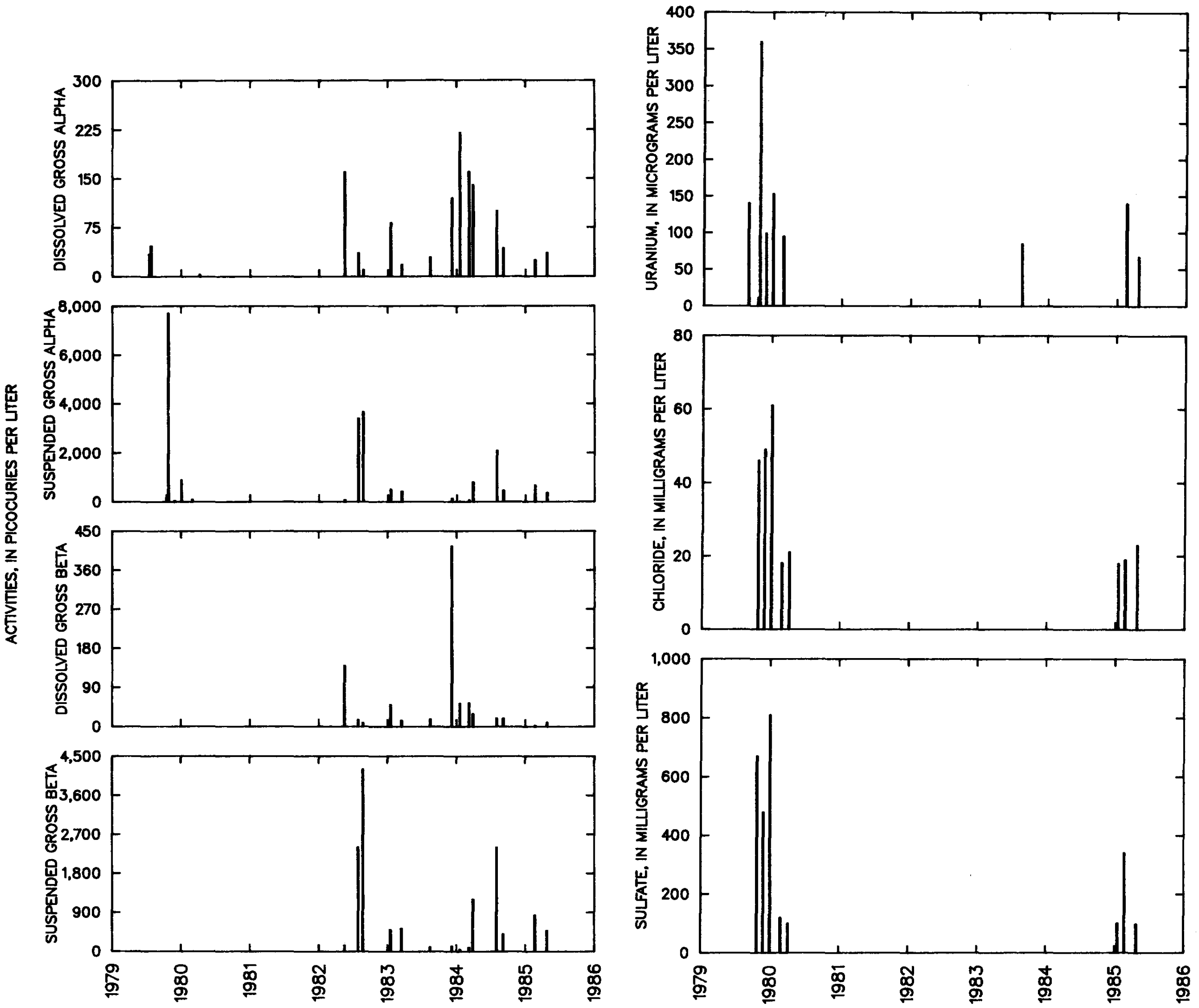

Figure 131. Temporal variations of selected chemical constituents in the Puerco River at Chambers, Arizona.

figure 132 shows dissolved and suspended gross alpha and gross beta activities measured from the 17 samples. Activities of individual radionuclides were not analyzed. Activities of dissolved gross alpha ranged from less than 6 to 54 pCi/L, and dissolved gross beta activites ranged from 7 to at least 12 pCi/L. Unlike data from the Puerco River, however, the suspended fraction of both gross alpha and gross beta was typically one to two orders of magnitude larger than the associated dissolved fraction (fig. 132). All pH measurements were 7.6 or larger. The relatively small ratio of dissolved activities to suspended activities in the Little Colorado River at Cameron suggests that the effects of mining in the Grants Mineral Belt, more than 200 mi upstream, either are muted or are masked by suspended radiochemical inflows from other sources.

CHARACTERIZATION OF QUALITY OF GROUND WATER

Puerco River Basin

On the basis of 14 well samples collected in December 1986 (Webb and others, 1987b), quality of water in the alluvium was characterized by dissolved-solids concentrations that were larger than 500 mg/L, sulfate concentrations that were larger than 250 mg/L, and some activities of radionuclides that exceeded MCL's. The median concentration of dissolved solids is 698 mg/L; alkalinity, sodium, and sulfate constitute most of this total. Concentrations of dissolved solids, sulfate, and chloride are similar to concentrations in samples measured in the 1930's (Harrell and Eckel, 1939).

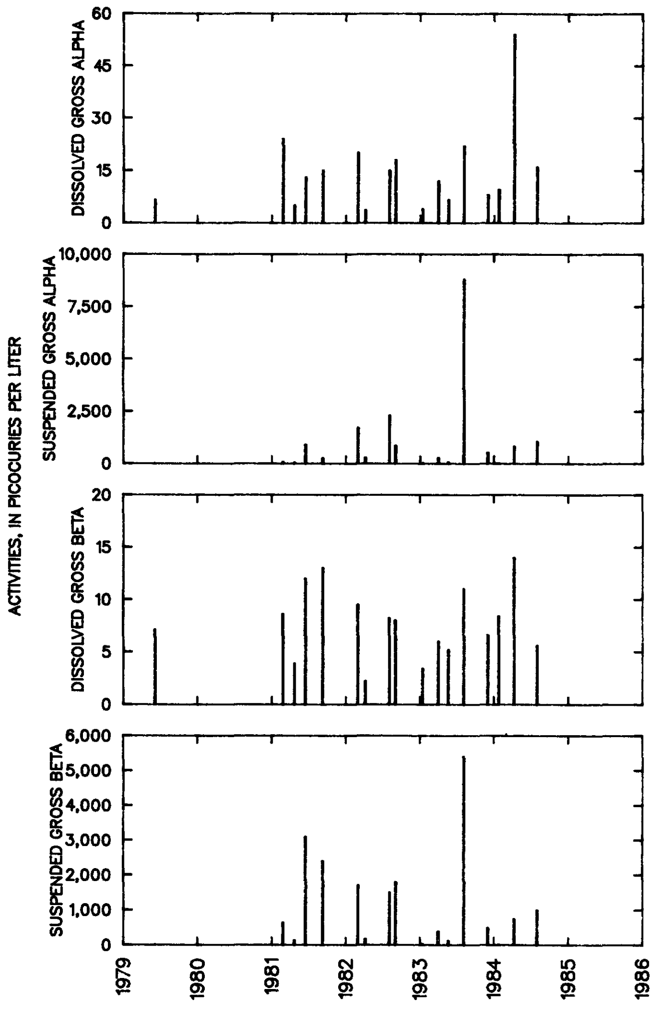

Figure 132. Temporal variations of selected chemical constituents in the Little Colorado River at Cameron, Ariz. Arizona maximum allowable limits are 15 picocuries per liter of gross alpha minus uranium and 30 picocuries per liter of gross alpha plus gross beta. Gross alpha activities are not corrected for uranium activity.

Median total gross alpha activity is 27 pCi/L, and activities range from 4 to 42 pCi/L. Median total gross beta activity is 6 pCi/L (table 59), and activities range from 1 to 19 pCi/L. Uranium concentration normally is about 4.5 µg/L in ground water in the United States (Cothern and others, 1983). In samples from the Puerco River basin and surrounding areas, normal concentrations of uranium ranged from 0.1 to 6.1 µg/L (Scott and Barker, 1962). Uranium concentrations in 14 wells and a spring in the Puerco River basin ranges from 1 to 38 µg/L with a median value of 19 µg/L. Other radionuclides, including ^{226}Ra, ^{228}Ra, ^{210}Pb, and ^{230}Th, generally have activities less than 1 pCi/L. Only one well had a ^{226}Ra plus ^{228}Ra activity exceeding 5 pCi/L (Webb and others, 1987b). These data indicate that uranium activities account for most of the gross alpha activity measured in samples from the 14 wells.

Table 59. Average and median concentrations or activities of selected constituents measured in 15 ground-water samples from the Puerco River basin, December 1–6, 1986

[Values in parentheses are medians; includes values less than the detection limit that were considered equal to the detection limit in the arithmetic mean. The average and median pH values are 7.7 and 7.8, respectively. The average and median temperatures are 15 °C and 14 °C, respectively]

Common ions (milligrams per liter, dissolved)		
Alkalinity	345	(341)
Calcium	80	(75)
Chloride	91	(61)
Fluoride	.8	(.8)
Magnesium	19	(15)
Potassium	2.8	(2.0)
Silica	14	(14)
Sodium	230	(230)
Sulfate	340	(280)
Selected trace elements (micrograms per liter, total recoverable)		
Aluminum	260	(20)
Arsenic	<2.2	(1)
Barium	<110	(<100)
Boron	400	(380)
Cadmium	<2	(<1)
Copper	18	(9)
Iron	540	(360)
Lead	<6	(<5)
Manganese	<240	(140)
Selenium	<44	(<39)
Strontium	1,000	(840)
Vanadium	<44	(<39)
Zinc	85	(70)
Radionuclides (picocuries per liter, except as noted, total)		
Gross alpha	22	(27)
Gross beta	7	(6)
Lead-210	.5	(.4)
Radium-226	.4	(.1)
Radium-228	<.7	(.3)
Thorium-230	.1	(.0)
Uranium (micrograms per liter)	19	(19)

Lower Little Colorado River Basin

Few data are available on ground-water quality in the lower Little Colorado River basin. Don Payne (Navajo Nation, written commun., 1987) described water-quality data collected in 1984, 1985, and 1986 at 49 locations in the Cameron Uranium Mining Belt region. Water samples from mining pits and nearby springs were considered indicative of ground-water conditions near the pits (Don Payne, Navajo Nation, written commun., 1987). All samples collected were analyzed for total concentrations or activities of constituents.

Analyses of water samples collected from 21 abandoned uranium mines yielded gross alpha activities that ranged from 24 to 9,400 pCi/L. Most of the gross alpha activity was attributed to ^{226}Ra and uranium (Don Payne, Navajo Nation, written commun., 1987). Radium-226 activities ranged from near below detection level to 760 pCi/L. Uranium activities ranged from 24 pCi/L to 4,020 pCi/L.

Gross alpha activities in water from six wells ranged from 3 to 10 pCi/L in the Cameron Uranium Mining Belt (Don Payne, Navajo Nation, written commun., 1986). Gross beta activities ranged from near 2 to 22 pCi/L. Radium-226 activities were below 0.8 pCi/L, whereas uranium activities ranged from 2 to 10 pCi/L.

Water from 14 developed springs had gross alpha activities that ranged from 4 to 200 pCi/L; gross beta activities ranged from 3 to 90 pCi/L; and all ^{226}Ra activities were below the detection limit. Uranium activities ranged from 11 to 159 pCi/L. The maximum value for gross alpha activity was measured concomitant with a uranium activity of 98 pCi/L at one of three springs where "obvious signs" indicated that "local people used the spring...as a source of potable water" (Don Payne, Navajo Nation, written commun., 1987).

AREAL AND TEMPORAL VARIATIONS IN WATER QUALITY

Puerco River Basin

When distance from the river is compared with radionuclide activities, the wells having the largest activities are not necessarily the wells closest to the river. For example, water from a well located approximately 1,000 ft from the Puerco River had a gross alpha plus gross beta activity of 7 pCi/L. Water from a well across the river and approximately 2,100 ft from the channel had a gross alpha plus gross beta activity of 27 pCi/L (Webb and others, 1987b). Similar differences were observed in wells at varying distances from the Puerco River at other study sites. The data also indicate that radionuclide activity was not related to the distance from the headwaters of the Puerco River (Webb and others, 1987b).

In addition to the 14 wells and 1 spring sampled in December 1986, historical water-quality data were available for 7 wells and 1 spring (Webb and others, 1987b). Ground-water samples from 1969, 1974, 1975, and December 1986 were collected and analyzed by the U.S. Geological Survey. The remaining samples were collected by other agencies—notably the Environmental Protection Agency— and the Indian Health Service and were analyzed by several different laboratories.

Data collected for Begay Well near the State line of Arizona and New Mexico include 32 water-quality measurements made before December 1986. Temporal variations of gross alpha, chloride, and sulfate indicate that activities and concentrations have fluctuated with time. For example, concentrations of sulfate rose from about 500 mg/L to as much as 1,600 mg/L after the spill of 1979 and declined thereafter to at least 840 mg/L in December 1986. Gross alpha activities also fluctuated during the same period. The gross alpha activity measured in 1986 was similar in magnitude to the largest historical activities, although the activity in 1986 is the largest measured between 1979 and 1986. The difference may result from the fact that the 1986 sample was the only sample collected immediately after the well had been pumped (Don Payne, Navajo Nation, written commun., 1988).

Historical variations in ground-water quality reflect the fluctuations in water chemistry of recharge to aquifers from the Puerco River. The data indicate that activities of radionuclides in ground water have fluctuated with time but have increased in some wells following the failure of the tailings-pond dam in 1979 near Church Rock, N. Mex. Concentrations of other constituents, such as chloride, appear to have changed little with time except at Begay Well, which is 50 ft from the Puerco River and may withdraw some of its water supply from the river through the alluvium.

PROCESSES CONTROLLING THE MOVEMENT OF RADIONUCLIDES

Radionuclide activities in surface water of the Puerco River at Chambers and Little Colorado River at Cameron are large compared with historical activities in ground water from wells. Total uranium concentrations have been approximately 100 μg/L in the Puerco River; however, concentrations of dissolved uranium are unknown. The median concentration of total uranium in water from wells near the Puerco River sampled in December 1986 is 19 μg/L. Activities of ^{230}Th and ^{226}Ra have been large in surface waters and suspended sediments of the Puerco River (table 58; Weimer and others, 1981; Arizona Department of Health Services, 1986a), but these radionuclides do not occur in significant activities in the ground water of that region. The median activity of total uranium in wells sampled by Don Payne (Navajo Nation, written commun., 1986) is 12 pCi/L. Uranium activities in surface water of the Little Colorado River are unknown.

Geochemical processes related to sediments in the surface water, clays, and grain coatings in the aquifer, in addition to chemistry of the ground water, may effectively reduce the radionuclide activities before recharge water reaches the wells. Two of the processes that remove radionuclides from water as it moves through the aquifer are mineral/water interactions and sorption. One chemical process capable of removing a radionuclide from solution is the precipitation of a mineral containing the radionuclide (Stumm and Morgan, 1970). Certain radionuclides, especially radium isotopes, may coprecipitate by substitution for a different cation, such as barium (Ba^{+2}), in a mineral that is precipitating (Langmuir and Riese, 1985). Sorption processes involve surface chemistry of certain minerals with water, such as cation exchange of radionuclides with other cations on clays (Beard and others, 1980), adsorption on particles, and complex cation reactions with organic compounds that adsorb with other cations or are taken up by plants (Landa, 1980). Sayre and others (1963) and Beard and others (1980) provide a more detailed description of the sorption process. Other processes, such as volatilization or biological activity, also may remove natural radionuclides from the water, although there is no documentation of these processes occurring in the alluvium of the Little Colorado River basin.

To test the importance of chemical reactions in ground water, a solution-equilibrium computer model (Kharaka and Barnes, 1973) that includes the reactions involving uranium (Y.K. Kharaka, U.S. Geological Survey, written commun., 1987) was used to determine the solubility of uranium minerals. Thermodynamic data for uranium are given by Langmuir (1978). The concentrations for uranium, measured as total concentrations, were assumed to be dissolved concentrations. The suspended and colloidal concentrations, therefore, are assumed to be negligible. Finally, the redox state was assumed to be positive, or oxidizing, owing to measurable concentrations of dissolved oxygen.

The solution-equilibrium model distributes measured concentrations of an element or compound among different chemical species. These species include cations, or positively charged ions; anions, or negatively charged ions; and complexes, which can be positively charged, negatively charged, or electrically neutral compounds. The ability of the chemical species to remain in solution is calculated from the saturation index of the minerals that could precipitate as a result of ionic activities of the necessary chemical species. The saturation index (SI) is defined as

$$SI = \log(AP/K(T)), \qquad (1)$$

where

AP =ionic activity product and

K(T)=equilibrium constant at absolute temperature, T (Lee and Strickland, 1988).

If the value saturation index exceeds 0 for a chemical reaction, precipitation will occur; if the saturation index is negative, dissolution of the mineral will occur if it is present in the aquifer (Kharaka and Barnes, 1973). Stumm and Morgan (1970), Kharaka and Barnes (1973), and Hem (1985) give more information on the chemical reactions.

The concentrations of constituents shown in table 59 for all 15 water samples collected in December 1986 were used in the solution-equilibrium model. Uranium-bearing minerals that could possibly precipitate from the ground water have negative saturation indices in all 15 ground-water samples, and these values indicate that uranium precipitation should not occur. To test whether the waters were close to saturation, the average concentrations of the 15 samples (table 59) also were used in the solution-equilibrium model. For modeling purposes, the uranium concentration was increased in the average sample from 19 μg/L to 100 μg/L, which is the average total concentration in surface water, and to 360 μg/L, which is the largest total concentration measured in the Puerco River at Chambers (fig. 131). Concentrations are large because 30 to 70 percent of the total uranium is either in suspended sediments or in colloids. The negative saturation indices for all samples having the largest concentrations of uranium (table 60) indicate that uranium precipitation should not occur. The dominant species containing uranium in the carbonate-rich aquifer are $UO_2(CO_3)_2^{-2}$ and $UO_2(CO_3)_3^{-3}$.

Sorption decreases the amount of uranium in solution. Uranium is strongly adsorbed by organic compounds or clays, with maximum adsorption occurring at pH levels between 5 and 6 (Ames and Rai, 1978). Uranium complexes change from positively charged to negatively charged at a pH of about 6 (Ames and Rai, 1978; Langmuir, 1978). Uranium can be adsorbed onto particles that have amorphous iron-oxyhydroxide coatings, and the amount of adsorption is largest between pH levels of 5.5 and 8.5 (Langmuir, 1978). In the alluvial aquifer of the Puerco River, where pH is generally between 7.5 and 8.5, uranium should not precipitate from ground water under oxidizing conditions but may be adsorbed on iron-oxyhydroxide-coated particles. The data collected in this study suggest that uranium is mobile in the alluvial aquifer under oxidizing conditions.

Other radionuclides have a different behavior. At pH levels of 7.5 to 8.5, thorium exists predominantly as thorium hydroxide (Langmuir and Herman, 1980), which is a mineral of low solubility (Ames and Rai, 1978). On the basis of thermodynamic data given by Langmuir and Herman (1980), the equilibrium activity of ^{230}Th is 0.2 pCi/L in alluvial ground water containing the average concentrations of constituents shown in table 60. Thorium is readily adsorbed on clays (Ames and Rai, 1978; Beard and others, 1980) and is almost completely adsorbed at pH levels greater than 6.5 (Langmuir and Herman, 1980). Thorium, therefore, is nearly immobile in a neutral or alkaline environment except when transported on sediment in surface water. The lack of ^{230}Th activities that are significantly greater than the detection limit further indicates that thorium is immobile in ground water of the alluvial aquifer of the Puerco River.

Radium-226 and ^{228}Ra are chemically similar to barium (Ames and Rai, 1978) and form sulfate and carbonate complexes (Langmuir and Riese, 1985). Calculations for the equilibrium concentrations (if all free carbonate ions are bound in radium-carbonate complexes) indicate that 180 mg/L of total radium (about 1.8×10^{11}pCi/L of ^{226}Ra) could dissolve in the water.

Table 60. Values of saturation indices for minerals of uranium and lead in an average sample of ground water from the Puerco River basin with uranium concentrations of 0.019, 0.100, and 0.360 milligrams per liter

[Thermodynamic data for uranium minerals are from Langmuir (1978). Saturation indices for concentrations of uranium (milligrams per liter) are calculated from equation 1 using the average concentrations of constituents in ground water given in table 59]

Solid-phase mineral		Saturation indices for concentrations of uranium (milligrams per liter)		
Formula	Name	0.019	0.100	0.360
UO_2CO_3	Rutherfordine	−3.0	−2.3	−1.7
UO_3	Gummite	−7.8	−7.0	−6.5
$UO_2(OH)_2$	—	−5.7	−5.0	−4.5
$K_2(UO_2)_2(VO_4)_2$	Carnotite	−6.7	−6.0	−5.5
$Ca_2(UO_2)_2(VO_4)_2$	Tyuyamunite	−5.2	−4.5	−4.0
$Ca_2(UO_2)_2(HSiO_4)_2$	Uranophane	−11.2	−9.7	−8.6
$UO_2(OH)_2 \cdot H_2O$	Schoepite	−4.4	−3.7	−3.1
$MgUO_4$	—	−11.9	−11.2	−10.6
$CaUO_4$	—	−2.7	−2.0	−1.5
$BaUO_4$	—	−8.9	−8.0	−7.6
UO_2F_2	—	−16.7	−16.1	−15.5
$PbCl_2$	Cotunnite	−8.1	—	—
$PbCO_3$	Cerussite	.3	—	—
PbO	Litharge	−5.2	—	—
PbO	Massicot	−5.4	—	—
$PbSO_4$	Anglesite	−2.6	—	—
$Pb_3(VO_4)_2$	—	4.3	—	—

Radium activities in natural waters are regulated by coprecipitation in other minerals and adsorption (Langmuir and Riese, 1985). Coprecipitation may occur in barite ($BaSO_4$), which has a saturation index of 0.9 in the average concentrations measured in the Puerco River basin. Radium may be sorbed on clays (Landa, 1980), although physical and chemical factors governing the adsorption are not well known (Beard and others, 1980). Ames and Rai (1978) report a correlation between the cation-exchange capacity of clays and the adsorption of radium.

Published data on radium are inconclusive with respect to mobility in ground water in the Little Colorado River basin. Radium may be adsorbed too strongly on clays to be transported as dissolved species. The magnitude of adsorption is evident in a significant correlation in increased ^{226}Ra activity and increased suspended sediment in the Puerco River (Arizona Department of Health Services, 1986b) compared to small radium activity in surface water and ground water near uranium mines or mills (Kaufmann and others, 1976; Yang and Edwards, 1984). Although radium is dissolved in water as $(RaSO_4)^\circ$ and $(RaCO_3)^\circ$, radium occurs principally in the solid phase in the alluvial aquifer of the Puerco River basin because of coprecipitation and adsorption processes.

Lead-210 is expected to have the same chemical properties as stable lead, has a half-life of 22 years, and can produce an activity of 1 pCi/L at a concentration of 1.3×10^{-8} µg/L. Saturation indices calculated from the average concentration suggest that two of six mineral phases are supersaturated at a lead concentration of 6 µg/L (table 60). Cerussite ($PbCO_3$), one of the two supersaturated minerals (table 60), controls the chemistry of lead in carbonate-rich waters at a pH of 8 (Stumm and Morgan, 1970). Lead also can be adsorbed by clay particles. The net effect of the chemical processes on the mobility of ^{210}Pb in the alluvial aquifer of the Little Colorado River basin is unknown.

DISCUSSION

The presence of uranium ores in the Little Colorado River basin of Arizona and New Mexico probably contributes to elevated background levels of radionuclides in surface water and ground water. Impacts of mining superimposed on generally large background levels of radionuclides and other trace elements probably increased levels of radionuclides in some water resources of the region. The quality of both surface water and ground water in the Puerco River basin varies both temporally and areally, and the same probably is true for the lower Little Colorado River basin.

Radionuclide transport in surface water and ground water is poorly understood. As of 1987, there are few reliable data on the presence and activities of radionclides in the Little Colorado River basin. Additionally, the lack of understanding of the processes that govern radionuclide movement and storage in the basin limits assessments of trends in the occurrence of contamination. This knowledge needs to be attained to develop and manage safe and reliable water supplies in the region.

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URANIUM, RADIUM, AND RADON IN DEEPLY BURIED SEDIMENTS OF THE U.S. GULF COAST

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Abstract

Uranium, radium, and radon were examined in the water, rock, and gases of deeply buried clastic sediments of the U.S. Gulf Coast. Results of these investigations are summarized in this paper. Uranium in solution in all cases examined seemed to be at or near saturation with a uranium mineral phase, either uraninite or coffinite. The presence of radium isotopes in solution in excess of supporting parent nuclides indicates a continuous removal of radium from the solid phase to the aqueous phase. The efficiency of this removal is proportional to the total dissolved-solids concentration of the aqueous phase. Radon was present in excess of radium in the aqueous phase in formation waters of less than 95 grams per liter total dissolved solids, whereas radon and radium were in secular equilibrium in waters above 95 grams per liter total dissolved solids.

INTRODUCTION

Conditions deep in large sedimentary basins are radically different from those of surface and near-surface environments; water temperature is much higher, pressure is greater, and the water usually is much more saline than surface or shallow ground waters. As a result, many changes take place that produce diagenetic reactions, mineralogic transformations, and changes in elemental distributions between water, rock, and gaseous phases present in the system. Examination of uranium- and thorium-series radioelements in this three-phase system can reveal a great deal about physical and chemical processes taking place in this environment. This report summarizes results of examinations of the natural radionuclide content of water, rock, and gas phases present in the thick clastic sequence of sediments deposited in the northern Gulf of Mexico basin.

Samples of rock, water, and natural gas were collected from these formations. The sources of the samples were from oil and gas wells and "geopressured-geothermal" energy test wells. These latter wells, which were basically research wells, were drilled by the U.S. Department of Energy in Louisiana and Texas to test the possibility of recovering commercial amounts of dissolved methane from hot, highly pressured aquifers. Care was given to drilling, completion, and production techniques to maximize scientific usefulness. They yielded large volumes of brine that were accurately measured, as was any gas or gas condensate produced. In several cases, the wells also were cored through the productive sandstone, and these cores were the sources of the rock samples. These wells, though few in number, provided excellent samples of water, rock, and gas.

The hydrocarbon-producing wells, drilled and owned by petroleum companies and also located in Louisiana and Texas, generally produced much less water than the geopressured-geothermal wells and presented sampling problems, in many cases. For example, at many wells that yielded only a small amount of water, the water was stored in tanks where cooling and oxidation could occur. At these wells, it was much more difficult to collect samples that were representative of in situ conditions, especially for uranium and radium, but the data reported in this summary are felt to be representative. Specific sampling techniques can be found in Kraemer and Reid (1984), Kraemer (1986), and Kraemer and Kharaka (1986).

GEOLOGIC SETTING

The study area encompasses the coastal section of Texas and Louisiana, where thick sequences of clastic sediments have been accumulating in the Gulf Coast geosyncline, a subsiding basin of deposition which presently is estimated to contain more than 15,000 m (meters) of water-saturated sediments at its center (Hardin, 1962). The major sedimentary rock types in this basin are sandstones and shales, the result of deposition of sediment carried by rivers draining the interior of the North American continent.

Basinal subsidence, salt tectonics, and differential compaction of sand and shale layers resulted in the formation of an extensive system of faults in the Gulf Coast sediments. This subsidence heated the permeable sandstone aquifers, and the faulting compartmentalized them. As a result, high temperatures and high fluid pressures developed in water occupying the pore spaces. During the subsidence and compaction sequences, the contained pore fluids commonly accumulated large amounts of dissolved methane from the breakdown of indigenous kerogen. Salinity of the pore fluids commonly increased above that originally trapped at time of initial deposition of the sediment. The most often cited processes causing salinity increase are ion filtration by shale beds (White, 1965; Jones, 1969) and dissolution of Jurassic salt beds underlying the area (Kharaka and others, 1978). Salinity decreases also occur, probably as a result of clay-mineral diagenesis (Burst, 1969) as temperatures rose above 110 °C in the sediment.

URANIUM

Uranium is present in solution in deep sedimentary aquifers in very low concentrations, ranging from 0.003

Table 61. Uranium content of geopressured-geothermal formation-water samples from the U.S. Gulf Coast

[From Kraemer and Kharaka, 1986, table 1; ^{234}U, uranium-234; ^{238}U, uranium-238; F, filtered; N, nonfiltered; μg/L, micrograms per liter]

Well	Uranium (μg/L)	^{234}U/^{238}U activity ratio	Filtering
2 Zellerbach	0.007±.001	1.23±.10	N
2 Sutter	.030±.002	1.21±.10	N
1 Sweezy	.003±.002	1.15±.30	N
1 Girouard	.011±.001	1.35±.20	F
2 Simon	.009±.001	1.21±.12	N
2 Pleasant Bayou	.003±.001	1.42±.14	N
1 Delcambre, Planulina 3.	.010±.001	1.62±.15	F
1 Delcambre, Planulina 1.	.005±.001	1.06±.10	F
2 Saldana	.044±.003	1.35±.15	F
1 Koelemay	.054±.010	1.38±.10	F
1 Prairie Canal	.030±.005	1.28±.10	N
1 Amoco Fee (zone 5)	.003±.001	1.69±.17	F

μg/L (micrograms per liter) to 0.054 μg/L (table 61). This low concentration is consistent with the reducing nature of the formation water, where uranium is in a reduced, insoluble oxidation state. Kraemer and Kharaka (1986) have shown by use of chemical equilibrium modeling that the uranium concentrations presented in table 61 are very close to what would be expected for these formation waters in contact with solid uraninite (UO_2) or coffinite ($USiO_4$) at the presumed in situ pH and Eh conditions within the aquifers.

Data on the nature or distribution of uranium minerals in geopressured-geothermal aquifers are not available, but it seems reasonable that the early diagenetic history of the sediments could be responsible for the deposition of small amounts of uranium on matrix grains, probably as amorphous UO_2. This uranium would have come from interstitial sea water present at the time of deposition. Sea water contains 3 μg/L of uranium under open-ocean conditions (Krauskopf, 1967). As conditions in the sediment became reducing, U^{+6} would have been converted to a lower oxidation state and precipitated. As temperatures rose in the sediment because of subsidence, amorphous UO_2 probably became converted to crystalline UO_2. Silica (SiO_2) from dissolution of quartz grains and silicate minerals also increased in the interstitial solution as a result of the rising temperature. According to Langmuir (1978), thermodynamic data suggest that under such conditions, $USiO_4$ should be the stable phase, so the final reaction would be conversion of crystalline UO_2 to $USiO_4$ on the matrix grain surface. The kinetics of this final reaction are not well known, and, for lower temperatures, UO_2 and $USiO_4$ are commonly found to coexist in sedimentary uranium deposits (Brookins, 1975). It would appear from the results of the geochemical modeling that both solid phases are close to equilibrium with dissolved uranium; hence, either mineral could be present (Kraemer and Kharaka, 1986).

As shown in table 61, all uranium activity ratios (UAR) are within the range 1.06 to 1.69. Kraemer (1981) and Andrews (1983), among others, have shown, however, that the UAR of ground waters should be significantly affected by a process known as alpha-particle recoil, where uranium-234 (^{234}U) becomes enriched in solution through the ejection of uranium-238 (^{238}U) daughter atoms from the matrix as a result of alpha decay. Such a process can, theoretically, result in UAR of formation waters in excess of 20, and high values have been reported in the literature (Osmond and Cowart, 1976, 1982). The slightly elevated UAR in these deep formation waters probably result from the equilibration of dissolved uranium with uranium minerals and uranium recoiled from the matrix grains.

These released ^{234}U atoms would enter the formation water, but, before the large excess activities of ^{234}U predicted by theory could build up, the ^{234}U would participate in the equilibrium exchange between solid-phase and dissolved-phase uranium. The equilibrium UAR for the formation water then would be controlled not only by the ^{234}U being supplied by alpha recoil effects but also by the total amount of uranium in the uranium-mineral phase and the liquid phase. Isotopically, this reequilibration would keep the UAR lower than if no solid-phase uranium were present, because much of the solid-phase uranium is ^{238}U. The greater the amount of precipitated-phase uranium in relation to the ^{234}U being added to solution by recoil effects, the nearer the UAR of the solution will be to unity.

It appears that enough alpha-recoil-generated ^{234}U is being introduced into the dissolved uranium-precipitated uranium system to cause a slight elevation of the UAR. It also appears that very high UAR are not likely to be found in deep, high-temperature formation waters of this type, because of the unusual circumstances that would be required, such as the presence of uranium-rich sand grains, extremely small grain size, or the absence of a solid uranium mineral with which to reequilibrate.

Kraemer and Kharaka's (1986) finding of slightly elevated UAR and near equilibrium of waters with UO_2 or $USiO_4$ in the formation water produced by every well sampled indicates that alpha-recoil reequilibration with solid uranium minerals is widespread in the Gulf Coast. Formation water from oil and gas wells similarly has slightly elevated UAR (Osmond and Cowart, 1982). The overall process described above also may be common to other geothermal aquifers. For example, Osmond and Cowart (1976) report finding low uranium concentrations and slightly elevated UAR in other geothermal systems, including the Yellowstone, Imperial Valley, and Mt. Lassen geothermal areas.

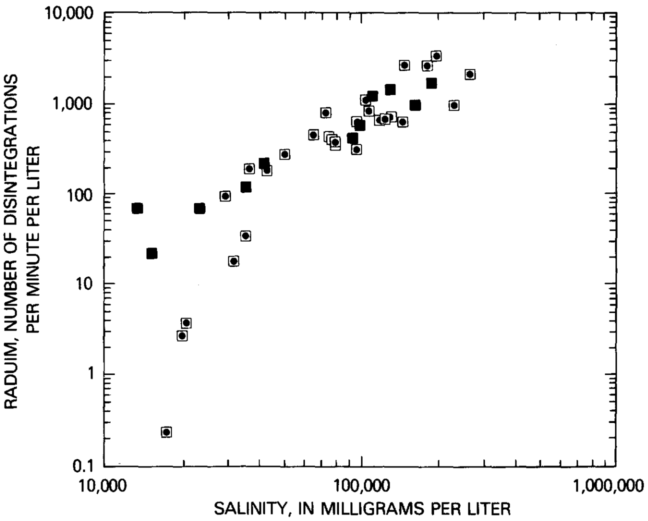

Figure 133. The radium and salinity relation in formation water from the U.S. Gulf Coast. Solid squares represent samples from geopressured-geothermal well tests conducted by the Department of Energy, and the other squares represent samples from aquifers that have coproduced oil or gas or both (from Kraemer and Reid, 1984).

RADIUM

Radium in Gulf Coast formation water is present in variable amounts, depending on the salinity of the formation water. Kraemer and Reid (1984) have shown that a correlation exists between salinity of formation water and radium activity that persists over a range of dissolved solids from 10,000 mg/L (milligrams per liter) to greater than 250,000 mg/L (fig. 133). The best correlation is for the geopressured-geothermal wells, which, as described earlier, are the best sources of samples; however, the correlation remains strong even if samples from oil and gas wells are included.

Because all radium isotopes are radioactive and the bulk of the radium in formation water is unsupported by its longer lived parents in solution, the formation water must continually be replenished in radium to replace the radium that decays. Because of its chemistry, radium is soluble in formation waters as long as the sulfate ($SO_4^=$) concentration is low. When formed by decay of previously existing nuclides within the matrix grains, radium can undergo a transfer from the solid to the liquid in accordance with the new chemistry. The radium also can be transferred to the liquid phase by alpha-recoil processes similar to those affecting uranium.

It can be demonstrated convincingly that radium is being leached from the aquifer matrix into solution if the ^{228}Ra/^{226}Ra (radium-228/radium-226) activities of the formation water are compared with the Th/U (thorium/uranium) activity ratios of the matrix, as shown in table 62. As thorium and uranium decay to ^{228}Ra and ^{226}Ra, respectively, the formation water will remove some fraction of the radium, but the relative activities of the two radium isotopes removed will be the same as the relative activities of their ultimate parents. Therefore, radium isotopic activities of formation water should reflect the Th/U activity ratios of the aquifer. In most cases, the two activity ratios agree (table 62), although there is a tendency for the formation-water ratios to be less than the parental ratios in the matrix. This tendency may be the result of the alpha-recoil process, because during decay, the ^{238}U atoms undergo three alpha decays before becoming ^{226}Ra, whereas the ^{232}Th atoms undergo only one alpha decay before becoming ^{228}Ra. Thus, the ^{226}Ra atom has three times as many chances as ^{228}Ra to end up in the water phase.

Table 62. Comparison of radium-228/radium-226 activity ratios of formation water with thorium/uranium activity ratios of the host matrix

[From Kraemer, 1985; ^{228}Ra, radium-228; ^{226}Ra, radium-226; Th, thorium; U, uranium]

Well	Formation water ^{228}Ra/^{226}Ra activity ratio	Matrix Th/U activity ratio
2 Pleasant Bayou	0.9	1.4
1 Sweezy	1.2	1.7
1 Amoco Fee (zone 3)	.8	.9
1 McCall (zone 8)	.6	1.3

The relation that exists between radium content of the formation water and salinity of the water must be chemical. Kraemer and Reid (1984) suggest the following possible controls on the relation between radium and salinity.

1. Ion exchange. In this process, the clay minerals in the formation control the radium content of the water by adsorbing more divalent cations (including radium) at lower salinity. The clay minerals are less efficient at adsorbing divalent cations at higher salinities, where Na^+ and other univalent cations are more abundant and compete more effectively for the clay exchange sites.
2. Barite equilibration. In formation waters of low ionic strength, the concentration of barium is usually low. Radium, because of its close chemical similarity to barium, could become involved in the equilibration process with solid-phase barium to a greater extent than in solutions of higher ionic strength, where barium is generally in much higher concentration. This process, however, was not considered likely, because in most cases, no barite ($BaSO_4$) is reported in Gulf Coast sediments, and formation waters from these formations are usually undersaturated with respect to $BaSO_4$.
3. Chloride-ion complexing. At the time Kraemer and Reid (1984) published their data, no estimates of chloride (Cl^-) complexing of radium were known, although they suggested this complexing as a possibility in explaining the relation between radium and

salinity. Langmuir and Riese (1985) subsequently published a paper that discussed the thermodynamic properties of radium and estimated formation constants of various radium complexes. If their data are used for the $RaCl^+$ complex, it can be estimated that the complexing of radium by the chloride ion, although possible, is not sufficient to explain satisfactorily the relation shown in figure 133.

4. Chloride-ion influence on silicate-solution equilibration. The continuous reequilibration of silica between solution and grain surface is capable of freeing radium atoms from the solid matrix and delivering them to solution. The greater the salinity of the formation water, the more effective the solution is at removing radium from the solid matrix material. The result is represented in figure 133. Kraemer and Reid (1984) hypothesize that the process responsible for the liberation of radium from matrix to solution is the continuous exchange of SiO_2 between silicate grains and solution. As the outer shell around a matrix grain is continually rehomogenized with SiO_2 from solution, any radium atoms exposed during the process are free to escape into and remain in the solution. Kraemer and Reid (1984) hypothesize that the presence of Cl^- in the formation water accelerates the rate at which SiO_2 exchanges between solution and matrix. The higher the Cl^- content, the more rapid the exchange, the result of which is a faster rate of "uncovering" of radium atoms. Radium atoms are allowed a faster rate of entry into solution, so a larger "standing crop" of radium exists in solution (not a continuously increasing amount, inasmuch as the radium atoms are also decaying in solution) in higher salinity water than in lower salinity water.

RADON

Radon is a decay product of radium. One isotope, ^{222}Rn, has a half-life of 3.8 days and is the daughter of ^{226}Ra. It commonly is found in subsurface voids, such as pore spaces and fractures, where it accumulates because of its noble-gas character. Two other isotopes of radon exist naturally, ^{220}Rn and ^{219}Rn, but their half-lives are so short that it is difficult to measure them or interpret their distribution patterns.

Radon (as ^{222}Rn) was measured in natural gas wells (table 63) and in gas produced from geopressured-geothermal wells (table 64) where the gas was extracted from solution at the surface (Kraemer, 1986). The radon in free gas from the natural-gas wells is quite low, averaging only 1.8 pCi/L (picocuries per liter) at standard temperature and pressure (STP). In fact, this figure is well below the average of 37 pCi/L found by Johnson and others (1973) in a survey of gas produced from the various gas-producing regions of the United States. Radon in gas extracted from solution from the formation waters of the geopressured-geothermal wells (table 65) was considerably higher and had a larger range (13–233 pCi/L at STP) than did the free gas. Even this range, however, is well within the range found by Johnson and others (1973).

Table 63. Radon activity in conventional gas at well head in the U.S. Gulf Coast

[From Kraemer, 1986; ^{222}Rn, radon-222; STP, standard temperature and pressure]

Well	^{222}Rn, in picocuries per liter at STP	Depth, in meters[1]	Louisiana parish
1 Gayle	0.7	4,973	Vermilion.
1 Hardee	.8	4,878	Vermilion.
1 Broussard	1.0	5,180	Vermilion.
S.L. 3057 no. 30	2.2	4,546	Vermilion.
S.L. 3057 no. 22	.8	4,512	Vermilion.
S.L. 3055 no. 5	2.9	4,489	Vermilion.
1 Dartez	3.4	5,930	Vermilion.
1 Hulin	.5	6,425	Vermilion.
1 Landry	2.2	4,396	Lafayette.
2 McConnel	5.8	3,424	Lafayette.
1 McConnel	1.2	3,575	Lafayette.
1 Melancon	3.4	3,891	Acadia.
1 Guilbeau	3.5	3,000	Lafayette.
B-1 Hebert	3.6	4,725	Lafayette.
B-1 Blanchard	<.5	4,021	Lafayette.
White Lake 40	1.0	4,553	Vermilion.
2 Humble Fee	2.5	4,935	Vermilion.
4 Hardee	.8	4,860	Vermilion.
LeJeune	.7	4,960	Vermilion.
3 Hardee	.5	4,805	Vermilion.
1 Green	1.0	5,146	Vermilion.
3 McConnel	2.8	4,725	Lafayette.
Average	1.8		

[1] Depths below land-surface datum.

The difference in radon activity between solution gas and free gas can be explained by the differing nature of the gas in the subsurface. As explained by Kraemer (1986), conventional gas deposits occur as free gas occupying space between matrix grains. Most of the water has been displaced, with usually only a small amount (30 percent or less of pore volume) of water remaining. Radon originating from the decay of radium enters this gas space and establishes a concentration dependent on the rate of supply from the liquid and solid phases and the rate of decay of unsupported radon within the gas. When the natural gas is produced from deep, hot reservoirs, it undergoes expansion in the range 500 to 1,000 times its in situ volume. Because the amount of radon is constant (neglecting decay) during expansion, its concentration (in terms of activity per unit of constant volume) in the gas decreases by a factor of 500 to 1,000 during this expansion to surface temperature and pressure, so the result is low radon activity per unit volume of gas.

Table 64. Radon activity in geopressured solution gas at well head in the U.S. Gulf Coast

[From Kraemer, 1986; ^{222}Rn, radium-222; pCi/L; picocuries per liter; STP, standard temperature and pressure; La., Louisiana; Tex., Texas]

Well	^{222}Rn, in pCi/L at STP	Depth, in meters[1]	Parish or county	State
1 Sutter	233	4,834	St. Mary	La.
2 Simon	96	4,450	Vermilion	La.
2 Pleasant Bayou	158	4,473	Brazoria	Tex.
1 Girouard	46	4,511	Lafayette	La.
1 Prairie Canal	58	4,511	Calcasieu	La.
2 Zellerbach	34	5,105	Livingston	La.
1 Saldana	13	2,984	Zapata	Tex.
1 Koelemay	30–119	3,567	Jefferson	Tex.
1 Amoco Fee (zone 5)	127	4,694	Cameron	La.
1 Amoco Fee (zone 3)	114	4,654	Cameron	La.
1 Sweezy	95	4,079	Lafayette	La.
1 McCall (zone 9)	55	4,738	Cameron	La.
1 McCall (zone 8)	41	4,665	Cameron	La.
Average	96			

[1] Depths below land-surface datum.

Table 65. Radon and radium activity in formation water produced from geopressured-geothermal test wells

[From Kraemer, 1985; ^{222}Rn, radon-222; ^{226}Ra, radium-226; mg/L, milligrams per liter; pCi/L, picocuries per liter]

Well	Approximate salinity (mg/L)	^{226}Ra, dissolved (pCi/L of water)	^{222}Rn, dissolved (pCi/L of water)	^{222}Rn/^{226}Ra activity ratio
1 Sutter	190,000	784	945	1.20
1 Amoco Fee (zone 5)	165,000	631	452	.72
1 Amoco Fee (zone 3)	165,000	509	405	.80
2 Pleasant Bayou	132,000	692	619	.89
1 McCall (zone 9)	95,000	233	235	1.01
1 McCall (zone 8)	95,000	201	209	1.04
1 Sweezy	95,000	190	338	1.78
1 Prairie Canal	42,600	101	465	4.61
2 Zellerbach	36,000	56	180	3.23
1 Girouard	23,750	32	368	11.4
1 Koelemay	15,300	10	637	61.5
2 Saldana	12,800	32	71	2.26

In a geopressured solution-gas reservoir, however, the same amount of radon is "stored" in the liquid phase. When the brine is withdrawn from the aquifer and brought to the surface, the radon is scavenged by the comparatively small amounts of natural gas exsolving from solution as a result of pressure and temperature reduction. Hence, the final activity of radon is much higher in this type of gas at surface conditions than in conventional gas.

Table 65 shows radium and radon activity in geopressured-geothermal wells in a unit volume of formation water. It can be seen from this table that, for formation waters containing less than about 95,000 mg/L total dissolved solids, the activity of ^{222}Rn in the water is greater than the activity of its parent, ^{226}Ra, in the water. The explanation is probably that most of the radium is physically located on or within the solid matrix grains and is, therefore, immobile, whereas the daughter product of the decay, ^{222}Rn, a noble gas, migrates with ease from the surfaces or even interior of the grains to the interstitial fluid. There it builds up to some equilibrium amount on the basis of its rate of supply from the solid material.

At or above a salinity of 95,000 mg/L total dissolved solids, the situation changes. At these salinities, in most cases, the amount of ^{226}Ra in the formation water is sufficient to supply the total amount of ^{222}Rn found. In other words, the radon is supported by the radium in solution, a rather unusual case for ground waters. The values of less than 1.0 for the ^{222}Rn/^{226}Ra activity ratios for some wells probably reflect incomplete extraction of radon from the brine or slightly inaccurate gas- or water-production measurements, which, of course, could affect the other wells as well.

Apparently, in these high-salinity formation waters, leaching of the matrix grains is sufficient to remove radium from a depth within each grain equal to or greater than that which radon could diffuse had the radium remained in the matrix. Thus, all of the radium that would have supported the radon in solution is now in solution itself. It is surmised that a very active process is occurring between the aqueous and solid-phase surfaces that continually and rapidly extracts radium from the outer regions of the matrix grains to a depth of several micrometers. The process cannot be a simple dissolution of the sand grains, because all these waters are in equilibrium with SiO_2, and dissolution would have long since ceased. The process is not understood at this time. Kraemer and Reid (1984) suggest that Cl^- may act as a catalyst in the movement of silica between solid phase and solution by preventing the hydrolysis product $Si(OH)_4$ from building up on the surfaces of the mineral grains. This action of the ion would tend to reduce the silica exchange rate.

CONCLUSIONS

1. Uranium is present in low concentrations in formation waters of clastic sediments of the U.S. Gulf Coast. Uranium in solution seems to be in chemical equilibrium with a solid phase of either uraninite or coffinite. The uranium isotopic-activity ratios support this conclusion.
2. Radium in solution in these formation waters is present in activities related to the salinity of the formation water. The higher the salinity, the greater the activity of ^{226}Ra. This relation indicates a continuous interaction of formation water with the matrix, and the result is the extraction of radium. The intensity of the reaction, and hence the activity of radium in solution, is proportional to the salinity of the formation water.
3. Radon is present in these formation waters and natural gases. At low salinities, ^{222}Rn is present in excess of

the radium in solution. The excess radon in solution is the result of exhalation from the aquifer rock. In formation waters having greater than 95,000 mg/L total dissolved solids, however, the amount of ^{222}Rn activity present in solution is nearly equal to the amount of ^{226}Ra activity present in solution, a situation that once again indicates continuous removal of radium from the matrix grains. The exact mechanism for this removal is not known.

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RADON, HELIUM, AND OTHER GASES IN SHALLOW GROUND WATERS OF URANIFEROUS HOLOCENE ALLUVIUM, FLODELLE CREEK, STEVENS COUNTY, NORTHEAST WASHINGTON

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Abstract

Shallow ground waters from highly uraniferous late Pleistocene and Holocene valley fill sediments along the north fork of Flodelle Creek in Stevens County, Wash., were sampled in shallow holes dug in the valley floor and permitted to fill with water drained from the sediment. Springs on the hillslope above the valley floor and on the bottom of the valley floor were sampled also. Dissolved gases in the water samples were analyzed for radon, helium, oxygen, carbon dioxide, and methane.

Radon concentrations in the shallow ground waters of the valley floor are 2 to 3 orders of magnitude less than those expected for uranium deposits of similar grade because the uranium has not been in place long enough for secular equilibrium to be established between it and its decay products in these sediments. Radon concentrations in the ground waters in contact with the bedrock and overlying glacial till of the hillslopes of the catchment are greater than those in the uraniferous sediment of the valley floor, except where bedrock ground waters appear to be upwelling into the valley-floor sediment column. Measuring radon in the shallow ground water of drainages such as these is not a good method for searching for surficial uranium deposits but may be useful in determining ground-water flow paths in the catchment basin.

The ground waters in the drainage basin show characteristic values of radon, helium, oxygen, carbon dioxide, and methane controlled by the environment in which the waters occur or have recently traveled through. Stagnant waters from the sediment of a beaver pond in the upper part of the study area are high in carbon dioxide and methane because the organic matter in the sediment column has been oxidized and biogenically degraded. These waters are low in helium and radon as a result of dilution of these gases by addition of the biogenic gases and depletion by bubbling of carbon dioxide and methane through the sediment column. Ground waters from springs on the hillslopes are high in oxygen but relatively low in carbon dioxide and methane because the waters have limited contact with thin soil horizons and organic matter. Such waters are high in radon and helium derived from recent contact with uranium-rich weathered bedrock (where uranium approaches secular equilibrium with its decay products) and sandy glacial till derived largely from bedrock material. "Normal" ground waters in the sediment filling the valley floor contain modest amounts of radon, helium, oxygen, carbon dioxide, and methane because the waters have recent contact with biologically active, uraniferous sediment in the upper part of the sediment column of the valley floor. Because ground waters in a zone of upwelling along the valley floor are very low in oxygen, carbon dioxide, and methane, but very high in radon and helium, a bedrock source is suggested, possibly a fracture zone that intercepts the valley floor.

INTRODUCTION

Late Quaternary organic-rich alluvium along the valley floor of the north fork of Flodelle Creek in eastern Stevens County, Wash. (fig. 134), is highly enriched in uranium. Mining of a section of the valley floor began in September of 1983, and about 1,000 pounds of uranium were recovered prior to cessation of operations in 1985. Since 1983, scientists of the U.S. Geological Survey have studied the uraniferous sediment, the ground and surface waters, the bedrock, and the vegetation within the drainage basin of the north fork of Flodelle Creek in order to understand the source, movement, and entrapment of uranium in this modern, natural system. This work has resulted in a series of reports describing the geologic setting and stratigraphy of the uranium deposit (Johnson and others, 1987); the aqueous geochemistry of the shallow ground and surface waters (Zielinski and others, 1987); uranium series disequilibrium of the sediments (Zielinski and others, 1986); mechanisms of entrapment of uranium in the sediment (Otton and Zielinski, 1985); and the biogeochemistry of uranium in Douglas fir in the drainage basin (Zielinski and Schumann, 1987).

This earlier work strongly suggests that uranium is leached from the uranium-rich quartz monzonite bedrock of the drainage basin, is carried by oxidized ground waters, and then is trapped by adsorption on organic matter in the shallowest parts of the sediment column. Uranium mineralization has occurred continuously since ice departed the area about 12,500 years ago and organic-rich sediment first began to accumulate. Once the organic-rich sediment becomes compacted and less permeable (below about 1 m (meter) in depth), the sediment appears to be closed with respect to uranium addition or loss except where more permeable layers, such as coarse sands at the base, occur. Gross disequilibrium occurs within these sediments: the content of radioactive decay products (specifically radium-226) increases from 1 percent of that expected under conditions of secular equilibrium at the surface to about 10 percent near the base of the mineralized sediment column. The sediment column near the spring pool (S4) near site WR14 (see fig. 135C) shows excess activity of lead-210

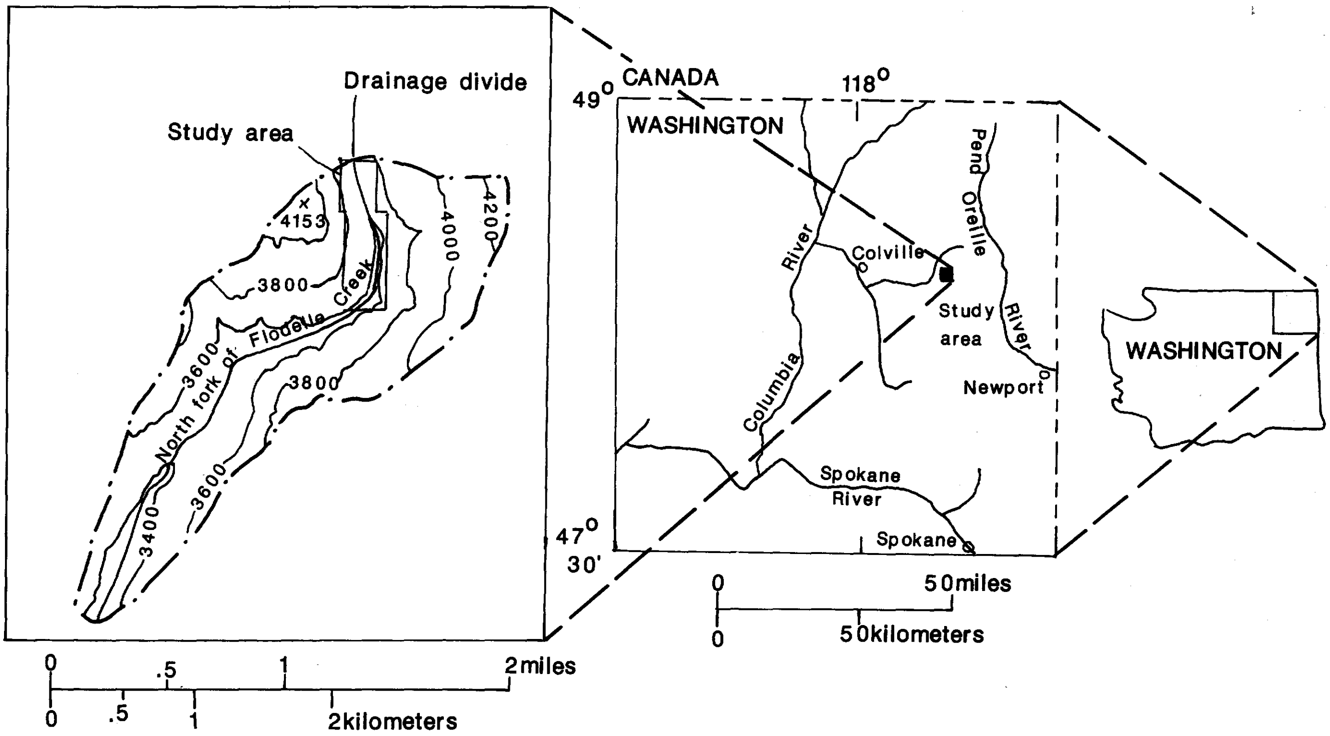

Figure 134. Study area in the upper part of the drainage basin of the north fork of Flodelle Creek. Contour interval is 200 feet.

compared with radium-226. Addition of radon-222 at that site from a source external to the sediment column is suggested.

Surface outcrop samples of the quartz monzonite in the drainage average 5.9 ppm (parts per million) uranium, whereas bedrock core from the drainage averages 15.8 ppm uranium (J.K. Otton, unpub. data, 1986). The near surface rocks have lost about 10 ppm uranium evidently attributable to physical and chemical weathering since deglaciation. Spring waters, surface waters, and shallow ground waters from the sediment column contain 17 to 318 ppb (parts per billion) uranium because the uranium has been leached from the rocks and is mobile in these waters.

In the summers of 1985 and 1986, we sampled shallow ground waters in the upper 0.9 km (kilometer) of the drainage basin in reconnaissance fashion and measured the dissolved radon, helium, carbon dioxide, oxygen, and methane contained in those waters (Sites WR1 to WR16, fig. 135). The objective of the study was to test a method for sampling dissolved gases in shallow ground water, to characterize the radiogenic and other gases dissolved in the shallow ground waters of this uranium deposit, to compare the dissolved gas content of the various types of waters found along the drainage, and to compare helium and radon content of the shallow ground waters to the uranium content of the subjacent sedimentary column. We wanted to understand the source and movement of radiogenic gases in the ground-water system and the probable pathways of ground-water movement.

CHEMISTRY OF RADON AND HELIUM

Radon-222 has a 3.823 day half-life, whereas its immediate precursor, radium-226, has a 1,623 year half-life. Radium and its precursor, thorium-230, are not very soluble in most natural waters. Where thorium-230 is formed by radioactive decay of uranium-234 in the water, it is readily sorbed onto various solid components of rocks and organic-rich sediment. Radium-226 also is sorbed readily or is coprecipitated with various mineral species. Thus the radon concentrations observed in waters are generally proportional to the radium content of the rocks and sediments in recent contact with the water and not of the water itself.

Radon readily degasses from water with agitation (during turbulent stream flow, for example) or with gas streaming through a sample (which forms the basis for the sampling technique used here and sampling techniques developed by others). Under equilibrium conditions, the ratio of the radon concentration in the water phase to the radon concentration in the gas phase of a gas-water system ranges downward from about 0.5 at 0 °C to 0.15 near 45 °C (Rogers, 1958).

The present-day atmospheric concentration of helium is 5,240 ppb. The helium content of water in equilibrium with the atmosphere at STP (standard temperature and pressure, 25 °C and 1 atmosphere) is 45×10^{-9} cm^3 of helium per cubic centimeter of H_2O. For shallow ground waters recently in contact with the atmosphere, values lower than this indicate additions of other gases to the dissolved gas load. Gases derived from the oxidation of organic matter or the respiration of biota in the soil of the hillslope and sediment of the wetland, such as carbon dioxide, methane, and oxygen, may lower the helium content of the dissolved gases by simple dilution. Losses of helium also may occur by bubbling of gases through a water-saturated sediment column. Those losses may be greater for helium than for other gases because helium has such a low atomic weight. Additions of helium from decay of radionuclides in rocks and soils in close contact with the water increase the helium content of the dissolved gases.

Because helium is small and chemically inert, it is a highly mobile element. It diffuses at a rate 7 times that of radon. Helium readily degasses from water upon agitation and has a strong affinity for the gas phase in a two-phase (air-water) system.

SITE DESCRIPTION

This study was confined to the upper 0.9 km of the stream valley occupied by the north fork of Flodelle Creek (figs. 134 and 135), and this description will focus on that area. For a more complete description of the entire drainage basin, see Johnson and others (1987). This upper part of the drainage basin was selected because it is essentially undisturbed by mining development, contains the highest grade of uraniferous sediment observed by us (as much as 0.9 percent uranium by dry weight near Site WR14), and contains several valley-slope and valley-bottom springs.

Adjacent hillslopes are underlain by quartz monzonite bedrock mantled by sandy glacial till; the valley floor is underlain by postglacial fluvial and marsh sediments. Thin volcanic ash-bearing loess and thin, organic-rich, poorly developed soils rest on the sandy till. Volcanic ash also occurs within the sediment column of the valley floor at depths of a few tens of centimeters to almost 3 m. Accumulations of organic-rich sediment extend essentially continuously along the valley floor in the study area from a small marsh a few hundred feet south of the drainage divide to a major turn in the valley about 0.9 km downstream (fig. 135).

In the upper 180–210 m of the drainage basin (fig. 135*A*, Site WR1 to just south of Site WR4), the valley bottom is about 15 to 23 m wide, slopes gently down valley (1 to 2½ percent), and is occupied by continuous wetland. The valley initially trends south-southeast but doglegs to the south near Site WR3. In the lower part of this section, where continuous streamflow develops (about halfway between Sites WR2 and WR3), beavers have dammed the stream and ponded the valley floor. The valley bottom is characterized by organic-rich, highly uraniferous sediment 0.9 to 1.5 m in thickness. The water table is at or near the surface throughout this part of the valley floor.

South of this section, for a distance of about 90 m, the valley narrows to about 9 m, changes orientation to the southeast, and the gradient steepens (fig. 135*A*, from below Site WR4 to Site WR6). Surface flow is confined to a narrow channel cut in the valley floor a few tens of centimeters. The valley is choked with deadfall trees. Scattered boulders poke up through the valley-floor sediment. Organic-rich sediment 0.6 to 0.9 m thick persists in this interval, but the abundance of organic matter decreases between Sites WR5 and WR6. These sediments are only slightly to modestly enriched in uranium (less than 1,000 ppm).

South of Site WR6, the valley floor abruptly widens (21 to 36 m), and the slope decreases. Between Sites WR6 and WR16 (fig. 135 *B,C*), the valley forms a broad arc and changes trend gradually from south-southeast to south-southwest. The character of the valley floor alternates from a flat-bottomed wetland with a meandering shallow stream channel to gently west-sloping valley bottom with a channel 15 to 45 cm deep cut into the valley floor by the present stream. These latter areas lie near the mouths of side valleys near Sites WR8, WR9, and WR15. At these localities, small alluvial fans that formed at the mouth of the tributary drainages force the stream channel to be displaced to the west side of the valley floor. These alluvial fans appear to have partially dammed the valley floor and thus have created ponded zones upstream. Auger holes near these fans are dominated by clastic sediments (Sites WR8 and WR9, fig. 135*B*), whereas auger holes upstream contain thick sections of organic-rich sediment (Site WR7, fig. 135*B*; Sites WR13 and WR14, fig. 135*C*). The alluvial fans have subsequently been cut by the channel in which the modern stream flows. The sediments in this part of the valley floor are variably enriched in uranium and have no particularly good correlation between the presence of organic matter and the presence of uranium.

Previous hydrologic studies of watershed areas (Freeze, 1972; Freeze and Cherry, 1979) and studies of Douglas fir that sample shallow ground waters in this basin (Zielinski and Schumann, 1987) suggest that the principal movement of ground water on the hillslopes in this drainage basin occurs as interflow along the contact between the highly permeable sandy glacial till and the underlying much less permeable quartz monzonite bedrock. Logging roadcuts show that the depth of the sandy till is in the range of 0 to 1.5 m on the west side of the drainage and 0 to 0.9 m or more on the east side. These roadcuts also show that the upper 2 or 3 m of the quartz monzonite has undergone rapid physical and chemical weathering since deglaciation. Outcrops and core show that the bedrock is highly fractured

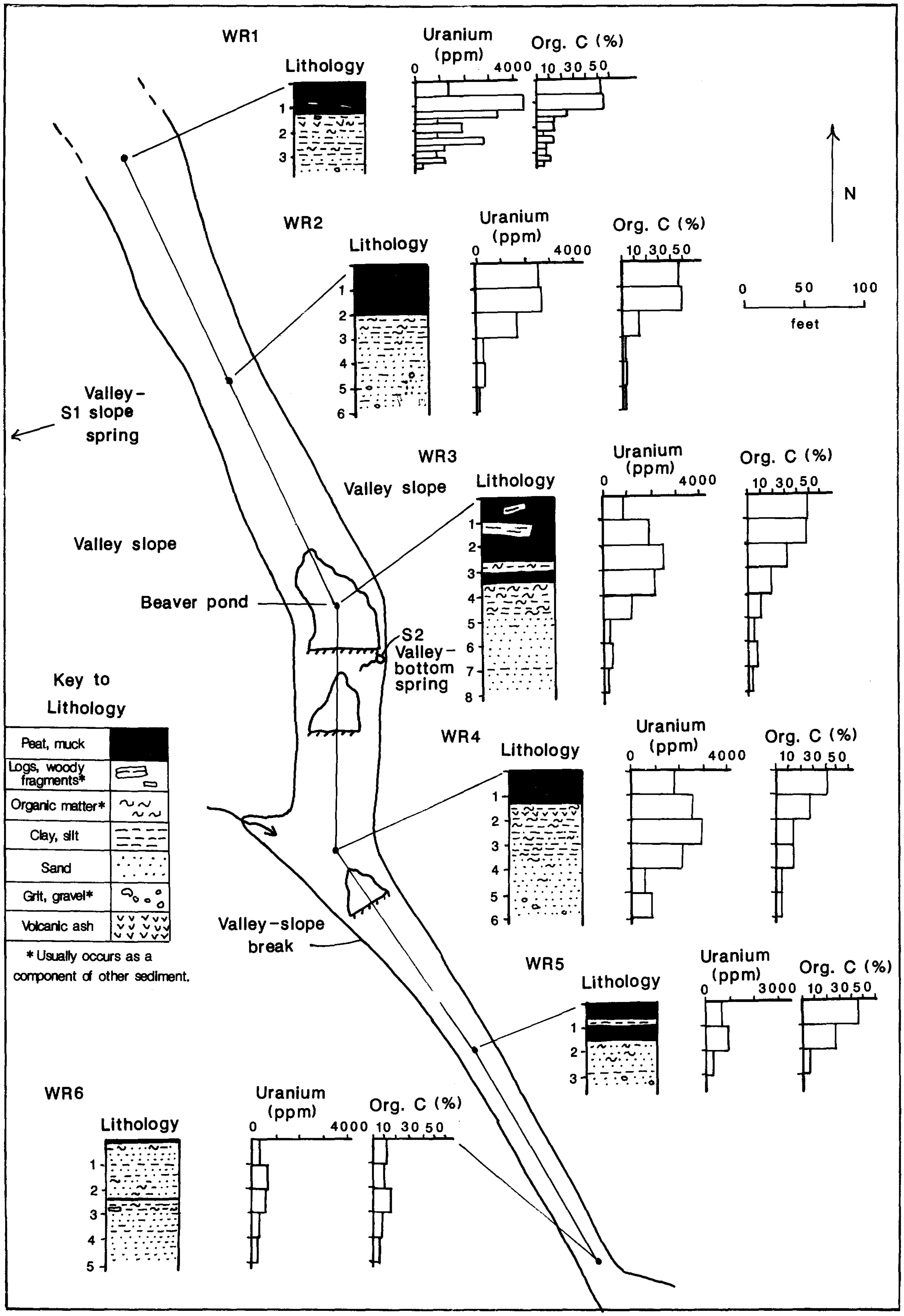

Figure 135. Plan maps of the study area in the upper part of the drainage of the north fork of Flodelle Creek. See figure 134 for location. Maps show location of shallow holes sampled for ground water and lithologic logs and geochemical data for auger holes taken at the same sites. Maps also show sample sites for the slope springs, the valley bottom spring, and the spring pool. All depths are in feet. Uranium values are in parts per million; organic carbon (Org. C) values are in percent. *A*, Upper section; *B*, Middle section; *C*, Lower section.

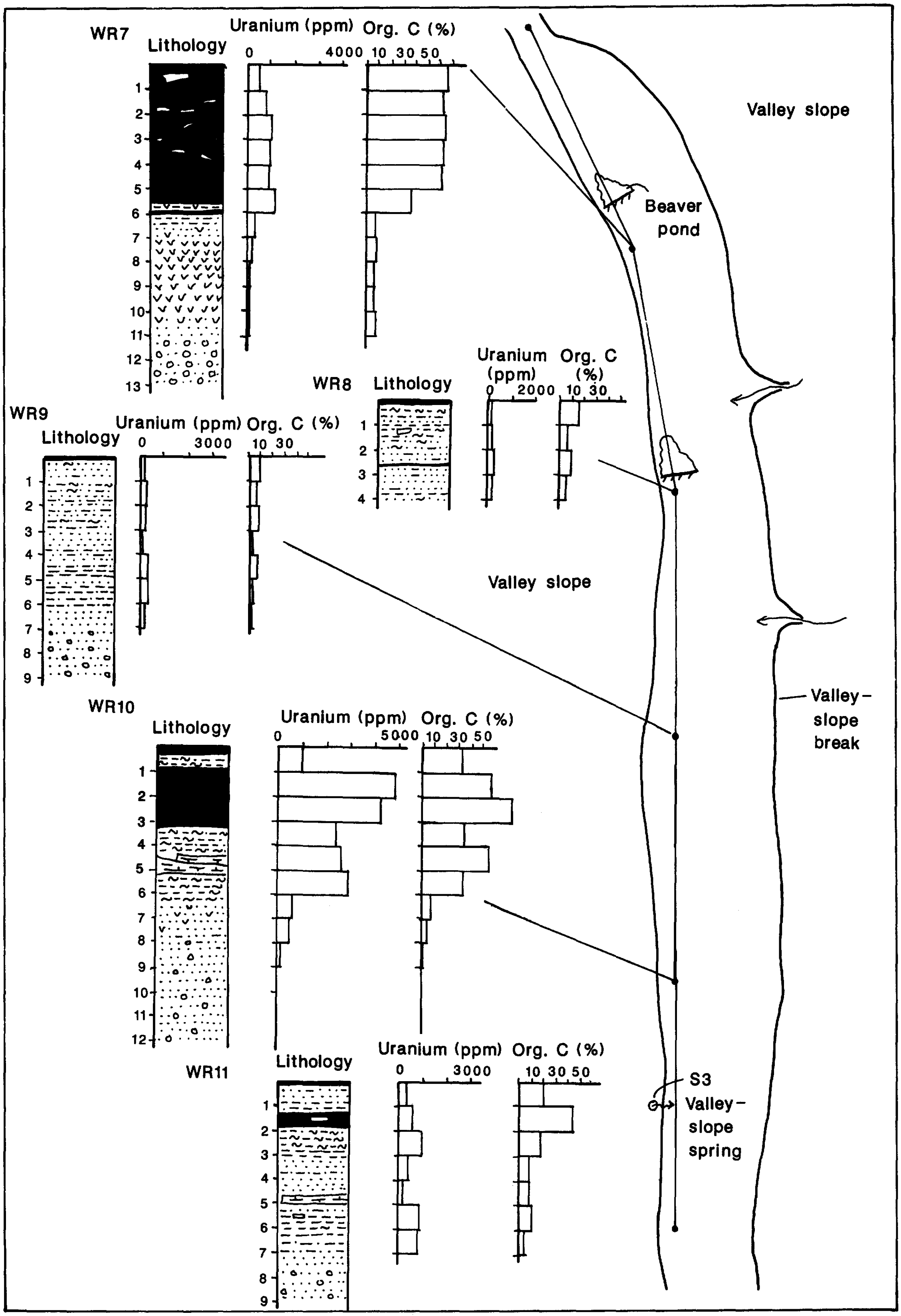

Figure 135.—Continued.

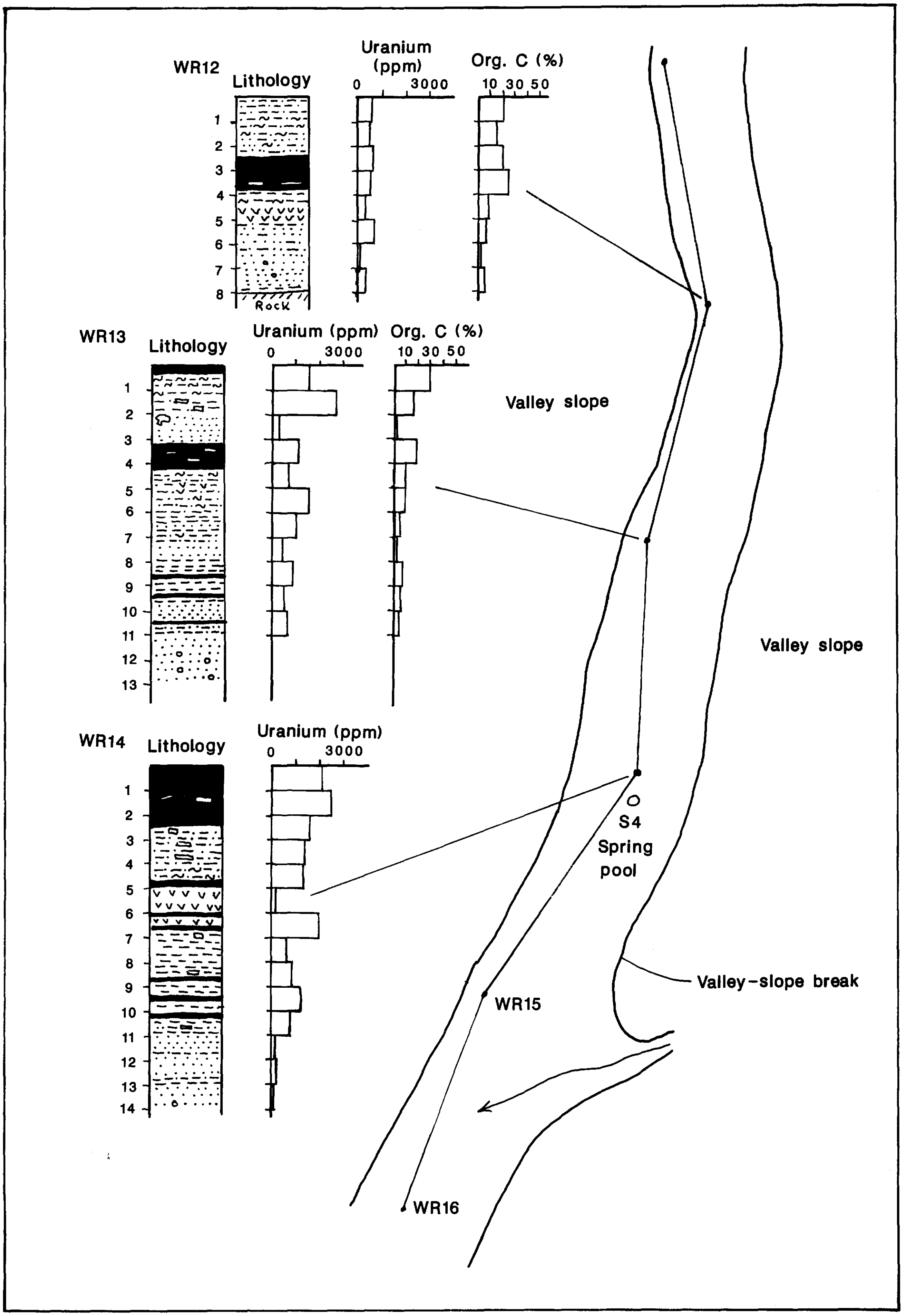

Figure 135.—Continued.

locally. Significant ground-water movement likely occurs within zones that have any significant fracture permeability. On the basis of the pattern of elevated uranium in Douglas fir twigs, the position of a slope spring, orientation of fracture and shear zones observed in the bedrock, and the north to northwesterly alignment of topographic elements in this basin and nearby basins, Zielinski and Schumann (1987) suggest that a major fracture zone parallels and lies directly west of this upper part of the drainage basin and intersects it in the lower part of the valley studied here.

METHODS

Sixteen water sample sites (Sites WR1 to WR16, fig. 135) were established at 61-m intervals down the axis of the valley floor along the upper 0.9 km of the north Flodelle drainage basin starting in the wetland along the valley floor several tens of meters south of the drainage divide. Each site was selected as close to the topographic low point in the valley bottom as was practical, typically adjacent to flowing streams on the valley floor. At each site, a hole 45 cm (centimeters) deep and 20–30 cm in diameter was dug. Shallow ground water seeped in, and the site was left undisturbed overnight for 18 to 24 h (hours) to permit the hole to fill and for the water in the hole to equilibrate with the water-saturated sediment. The following day each site was revisited, a water sample taken, and dissolved gases retrieved for radon measurements. A split of each gas sample also was taken for later measurement of helium, carbon dioxide, oxygen, and methane. A hole was subsequently augered at each site, and samples on 0.3-m intervals were recovered for lithologic descriptions, uranium analyses (by delayed neutron activation), and loss on ignition (after drying 24 h at 95 °C to estimate organic-carbon content). At Sites WR1 and WR14, lithologic descriptions and uranium data from auger holes sampled during a previous summer's fieldwork were used. Geochemical data from auger holes at Sites WR15 and WR16 were not received in time to be included in the results reported here.

Diffusive loss of dissolved gases from the surface of the water in the sample holes was expected to affect the gas measurements to some degree. Sequential readings of radon in waters of the same hole over 2 days showed a 20 percent drop in the second day (Site WR8, see table 66). This amount is within the analytical and sampling error of the technique. Sampling error is discussed later in this section. Because the holes were sampled uniformly 18 to 24 h after they were dug, diffusive losses to the atmosphere were likely to be consistent and not affect the interpretation.

Dissolved gases were recovered from the water at each site in a 1-L (liter) polyethylene bottle dipped into the water-filled hole, filled (with the minimum possible turbulence) to 0.75 L, capped, shaken for 1 min (minute), and allowed to rest for 1 min so all the gas bubbles could be exsolved. Then a gas sample was drawn from the headspace above the water with a 50-cm^3 syringe through a septum fitted into the cap.

Methods for determining the radon content were similar to but differed in detail between the 1985 and 1986 sample visits. During the 1985 sampling, a 20-cm^3 gas sample from the headspace was injected into an alpha-sensitive scintillometer (Lucas cell) counting device and counted for three 1-min intervals with a 1-min delay between counts. The scintillometer had a modified inlet system to permit the sample to be injected by syringe into the evacuated cell (Reimer, 1977). Background counts taken prior to sample injection were then subtracted from the sample count. Next, a second 20-cm^3 sample was injected into a 10-cm^3 metal cylinder for later analyses for helium, carbon dioxide, oxygen, and methane. During the 1986 sampling, we used three consecutive 2-min counts of the sample and a 50-cm^3 sample size to get greater precision. In 1986, no samples were taken for analysis of other gases. The counts per minute for radon in the gas sample were then converted to radon dissolved in the water in picocuries per liter. The conversions assume total degassing of the dissolved gases from the water in the bottle. Sequential degassing and radon measurements on the same sample (G.M. Reimer, unpub. data, 1975) show that about 10 percent of the radon remains dissolved in the water at low radon concentrations (100 pCi/L (picocuries per liter)) when this procedure is used, whereas only 1 percent remains in the water at higher concentrations (1,000 pCi/L). The scintillometer had been calibrated previously at the U.S. Bureau of Mines radon generation facility at Denver, Colo. The calibration established a sensitivity of 25 pCi/L for the unit for use with our sample size and field counting technique in the summer of 1985 and 5 pCi/L for our procedure in the summer of 1986.

Samples of a valley-bottom spring (S2, adjacent to Site WR3, fig. 135*A*), a valley-bottom spring pool (S4, adjacent to Site WR14, fig. 135*C*), and two slope springs (S1, on the valley slope west of Site WR2 (fig. 135*A*) and S3, just above the valley slope break between Sites WR10 and WR11, fig. 135*B*) were retrieved by a procedure in which the mouth of the 1-L polyethylene bottle was held as close to the source as possible and the water sample was allowed to flow into the bottle with a minimum of turbulence. Retrieval of dissolved gases followed a procedure similar to that of water from the shallow holes described above.

All 16 sample localities were visited between August 19 and August 24, 1985 (table 66), and revisited July 23 and July 24, 1986. In 1985, some sample splits were lost because the metal cylinders failed, and some samples were insufficient for complete analysis of all gases. We sampled the study area in late summer so that the diluting effect of spring runoff would be minimized; however, during the July 1986 visit, the water table was noticeably higher along

Table 66. Multiple measurements of radon taken at the same site in northeast Washington

Date	Time	Radon, in picocuries per liter
S4—Spring pool near WR14		
Aug. 19, 1985	5:06 p.m.	2,175
Aug. 20, 1985	9:32 a.m.	1,875
Aug. 23, 1985	3:51 p.m.	1,825
Aug. 24, 1985	12:01 p.m.	2,700
		2,150±400 average
WR2—Shallow ground water		
Aug. 23, 1985	9:23 a.m.	775
Aug. 23, 1985	9:33 a.m.	825
WR8—Shallow ground water (hole dug Aug. 22, 1985, about 6 p.m.)		
Aug. 23, 1985	12:47 p.m.	1,825
Aug. 24, 1985	9:25 a.m.	1,425

the valley floor than during the August 1985 sampling. The higher water table affected our sampling in ways described in the following section.

Helium analyses were made in laboratory facilities in Denver, Colo., by use of a mass spectrometer. The helium data are reported in parts per billion helium for the helium content of the gas in the headspace above the water in the sample bottle after the bottle was shaken, rather than for helium content of the water itself. For the sampling method used here, an increase of 20 ppb above the background of 5,240 ppb in the helium content of the air in the headspace corresponds to a sevenfold increase in the helium content of the water in the bottle. The other gases were analyzed on a gas chromatograph. The gas chromatograph uses a thermal conductivity detector for determining the permanent gases and a flame ionization detector for determining hydrocarbons. Data are reported as ratios with respect to nitrogen in the sampled gas. Ratios for a reference gas are also reported for comparison.

Sampling and operator errors in our sampling method may include (1) diffusive losses from the surface of the sampling holes (discussed previously); (2) gas loss or gain during agitation of the water sample while the sample bottle was being filled; (3) incomplete degassing of the water sample in the bottle during agitation (discussed previously); (4) inconsistent filling of the bottle to the same 750-mL volume for each water sample; (5) variations in the gas pressure in the headspace after the sample was degassed; and (6) inconsistent extraction of the same volume of gas sample. The percent error introduced likely varies from gas to gas. Some errors are systematic and produce values that are consistently lower than expected, while others are random. We estimate the random sources of sampling and operator error to be about 20 to 30 percent.

RESULTS AND DISCUSSION

Radon in the Shallow Ground-Water Samples

The radon content of shallow ground-water samples along the upper part of the north fork of Flodelle Creek ranged from about 50 pCi/L for Site WR7 to 8,700 pCi/L for Site WR13 (both in 1985, table 67, fig. 136). This range is in the same order of magnitude as those values observed in studies of springs (but not streams) draining unmineralized Paleozoic sedimentary and Precambrian crystalline rocks in the Wasatch Mountains of Utah (43 to 4,730 pCi/L in spring waters, Rogers, 1958) and for ground waters in two south-central Utah basins (70 to 8,800 pCi/L in wells and springs, McHugh and Miller, 1982). But the range along Flodelle Creek is 1 to 2 orders of magnitude lower than ranges observed in private and small public water supplies derived from granitic rocks in Maine (Brutsaert and others, 1981) and 2 to 3 orders of magnitude lower than radon measured in water-supply wells in the Helsinki, Finland, metropolitan area (10,000 to 880,000 pCi/L, Asikainen and Kahlos, 1979). Moreover, these values are 2 to 3 orders of magnitude less than those observed in waters in contact with uranium deposits of this grade, 23,000 to 1,600,000 pCi/L in uranium mine waters at Elliot Lake, Ontario (Dyck and Jonasson, 1977). These overall lower-than-expected values are due to the extreme youth and gross disequilibrium found in this uranium deposit (Zielinski and others, 1986). Thus, measurement of radon in shallow ground waters is not expected to be an effective exploration tool in searching for this type of uranium occurrence.

Observed radon levels for the sampling in 1986 were consistently lower than those in 1985. Precipitous drops in the year-to-year sampling were observed at Sites WR1, WR3, WR4, WR5, and WR14; for example, WR1 dropped from 3,500 pCi/L in 1985 to 960 pCi/L in 1986. The 1986 sampling occurred earlier in the summer than the 1985 sampling, the water table was noticeably higher, and at these holes the water levels were above the surface of the ground in 1986. We attribute these precipitous drops in radon content in 1986 to the draining of the surface layer of radon-poor water into the hole after digging. We attribute the overall lower 1986 readings to greater dilution of more radon-rich base flow by radon-poor surface runoff from the previous spring snowmelt. At Site WR6, the 1985 hole was dry when revisited the following day, so we took a stream water sample instead, and it contained 200 pCi/L radon. The following year we dug a hole in the adjacent stream-channel bottom to obtain a sample, and it contained 1,095 pCi/L. Because of such sampling problems in 1986, we consider the 1985 dataset to be a more reliable estimate of the shallow ground-water radon concentrations at the time of sampling.

Table 67. Measurements of radon and helium in shallow ground waters in northeast Washington

[All radon data are in picocuries per liter; all helium data are in parts per billion; NS, not sampled; ND, analytical data not received in time for this study; lost, sample lost from container prior to analysis]

Sample site	1985 Rn	1986 Rn	Calculated Rn yield	1985 He	1986 He	Calculated He yield
WR1	3,500	960	3,900	5,320	5,100	10,000
WR2	800	1,055	2,950	5,250	5,110	8,840
WR3	425	75	1,350	5,010	4,960	6,920
WR4	1,700	255	3,850	5,230	5,260	9,950
WR5	800	225	1,000	5,290	5,140	6,470
WR6	[1]200	1,095	5,850	5,310	5,180	12,410
WR7	50	160	150	5,250	5,310	5,440
WR8	1,825	1,500	2,200	5,330	4,980	7,930
WR9	2,050	1,295	2,750	5,210	5,260	8,600
WR10	1,125	1,040	2,150	5,310	5,310	7,890
WR11	300	330	1,850	lost	5,100	7,510
WR12	4,600	4,505	3,650	lost	5,050	9,770
WR13	8,700	7,520	6,950	5,300	5,190	13,710
WR14	3,700	1,705	1,350	6,860	6,580	6,900
WR15	3,875	NS	ND	5,730	6,170	ND
WR16	2,375	NS	ND	5,270	5,140	ND

[1] This sample was taken from surface water in the stream adjacent to the hole when the hole proved to be dry.

Highest radon concentrations in the shallow ground water along the valley bottom occur from Site WR12 to Site WR16 (fig. 136, top graph). Within this section, upwelling ground waters are inferred from the abundance of seeps in the valley floor near Sites WR14 and WR15 and the presence of an open spring pool (S4, near Site WR14). Bedrock spring waters from the valley slopes (see fig. 136, 1985, S1 and S3; table 68) have consistently higher radon values than adjacent valley floor waters. These observations suggest that ground waters in contact with the weathered bedrock and the overlying sandy till contain more radon than shallow ground waters in contact with the highly uraniferous valley fill. Several factors may be responsible for this difference:

1. The uranium in the valley fill sediments is in gross disequilibrium with its radioactive decay products (the concentration of radioactive decay products is 1–4 percent of that expected for the amount of uranium present in the shallowest parts of the sediment column); thus, far less radon is produced than that which would be expected for the uranium present in the sediment.
2. The uranium content of the bedrock in this drainage basin is relatively high (5 and 15 ppm average for surface and core materials, respectively).
3. The uranium in the weathered bedrock and sandy till probably approaches secular equilibrium with its decay products. With the very recent loss of uranium from the near surface, there may be an excess of radium with respect to uranium.
4. Uranium (and radium) in the bedrock and till sources may tend to be concentrated on fracture and grain surfaces in contact with the water. This contact with concentrated uranium would tend to increase the apparent uranium content of the bedrock in a surface-area analysis and increase the fraction of radon (emanating power) that can escape to the water in the pores.

Without some detailed investigations of the distribution of uranium in the bedrock and uranium series radionuclides in the bedrock and till and without comparisons of the emanating power of the bedrock, till, and valley-floor sediment, the relative effect of these factors cannot be evaluated.

Radon levels in the shallow ground waters do not follow a pattern related in any simple way to the uranium content of the underlying sediment column. In order to evaluate this relation, we made grade × thickness (*GT*) calculations for the sample sites along the valley floor (fig. 136). The *GT* calculation was developed by uranium geologists working with drill hole samples and gamma logs on the Colorado Plateau as a means of estimating and comparing uranium content. A 10-ft intercept of ore averaging 0.1 percent yields a *GT* of 1.0. In contrast to the findings of these geologists where the density of the sandstone ore varied only slightly, the *GT* calculations we made here have been weighted for the varying density of the sediment.

Steady increases in the *GT* calculation from Site WR1 to Site WR4 are not followed by steady increases in the radon content of the shallow ground water. Note also that high *GT* calculations for Sites WR10, WR13, and WR14 (all close to 1.0) are marked by low, moderate, and high observed radon levels, respectively. This lack of correlation suggests that the radon content of the shallow ground water is not directly affected by the uranium (radium) present in

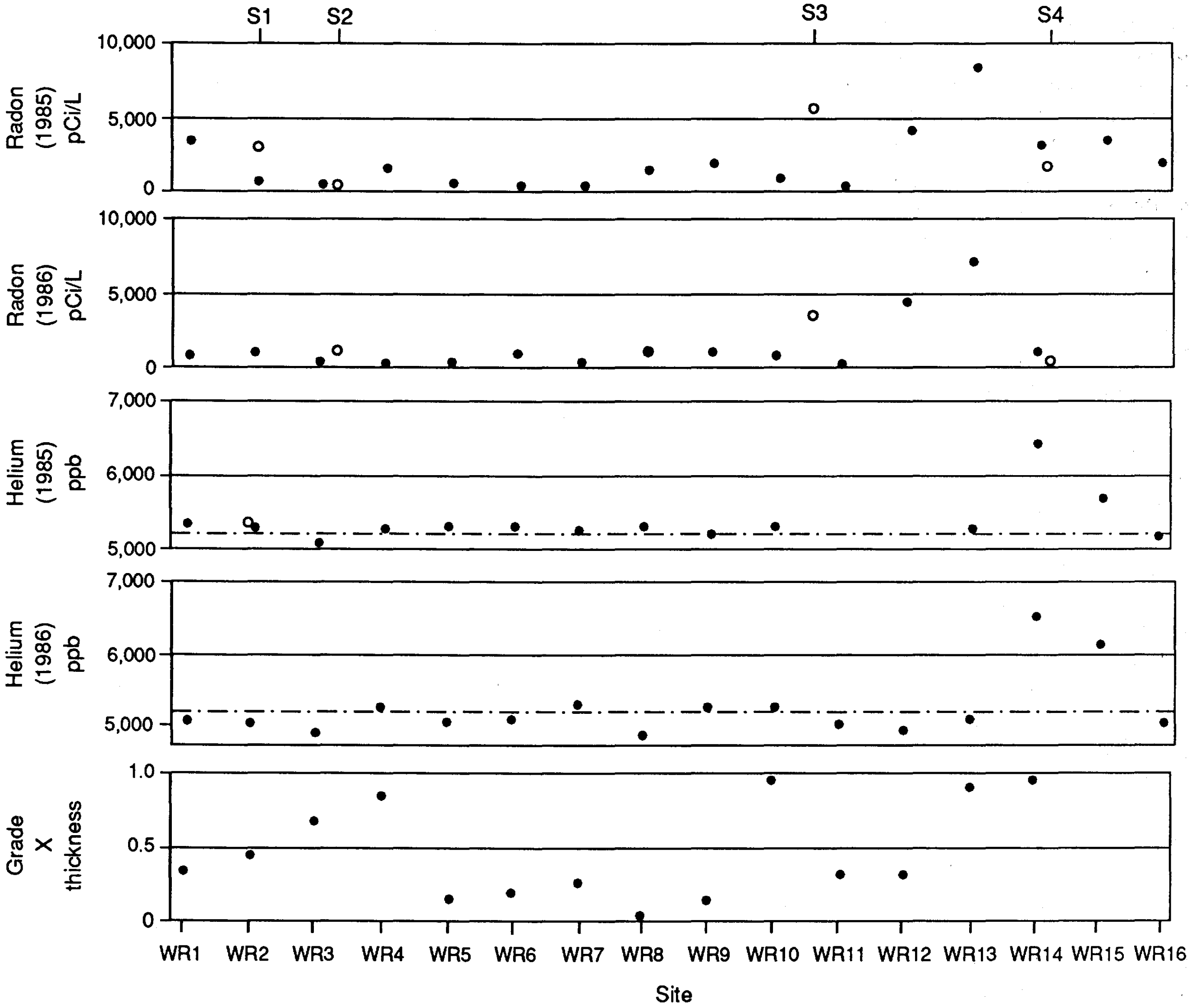

Figure 136. Radon and helium data for shallow ground waters (Sites WR1 through WR16 represented by dots) and spring waters (springs S1 through S4 represented by open circles) from the 1985 and 1986 sampling in the study area. For sample site locations see figure 135. The relative positions of spring sample sites (S1–S4) are marked along the upper edge of the series of graphs, whereas the relative positions of the shallow ground-water sample sites (WR1–WR16) are marked along the lower edge of the series of graphs. The dash-dot line on the helium graphs marks the helium content of air. Refer to text for explanation of grade X thickness calculations. pCi/L, picocuries per liter; ppb, parts per billion. Please refer to table 67 for an explanation of missing data from Sites WR1–WR16. Only one helium measurement for a spring (S1, 1985) was made.

Table 68. Radon measurements of selected spring and surface-water sample sites in northeast Washington
[pCi/L, picocuries per liter]

Site	Description	Date	Time	Radon (pCi/L)
WR3	Surface water from beaver pond	Aug. 23, 1985	9:49 a.m.	25
WR6	Surface water from stream adjacent to hole	Aug. 23, 1985	11:38 a.m	225
S1	Slope spring on hillslope above Site WR2	Aug. 21, 1985	No time recorded	3,075
S2	Spring water from valley floor spring just below beaver dam at Site WR3	Aug. 23, 1985	10:16 a.m.	775
S3	Spring water from valley wall spring between Sites WR10 and WR11; seep about 3 or 4 feet above valley floor	Aug. 24, 1985	10:27 a.m.	5,900

the underlying sediment column. We had previously noted in core sampling along this drainage basin that the near-surface sediment is the least compact and most water saturated or, by inference, the most permeable. Zielinski and others (1986) also noted apparent closed-system behavior of uranium and its decay products through radium-226 in a 10-ft core through the sediment column near the spring pool just downvalley from Site WR14 (fig. 135*C*). This closed-system behavior requires that little or no contact of the lower layers of sediment with uraniferous ground waters occur after initial uranium entrapment and subsequent burial. Compaction of the peaty sediment and reduction in hydraulic conductivity most likely prevent movement of uranium-rich waters into the sediment below the surface layers. Clymo (1983) observed that the bulk density of Sphagnum peat increases by a factor of 3 to 4 between 10 and 30 cm below the surface and that the hydraulic conductivity of peaty sediment decreases by 3 to 5 orders of magnitude during compaction and humification. These observations suggest that the flow path of the shallow ground waters is, in general, not downward (or upward) through the sediment column but rather horizontally through the near-surface layer, except near zones of upwelling waters marked by seeps and spring pools. In these areas, continuous upward flow of water has maintained flow paths through the sediment column.

These observations lead us to conclude that the near-surface sediment should be an important contributor to the radon in the shallow ground water except where upwelling waters from the bedrock invade the sediment column. In order to evaluate the relative contribution of radon from the near-surface sediment in the immediate vicinity of the hole to radon in water from bedrock sources, we calculated the expected radon yield of the upper 45 cm of sediment from each site by use of the auger hole data (see appendix A for methods and assumptions; table 67 and fig. 137 for the calculated radon yields and comparisons with measured radon levels for 1985 and 1986, and fig. 138 for the ratio of observed to expected radon for the 1985 samples). The ratio of observed to expected radon levels is below 1.0 from Sites WR1 to WR11 but goes over 1.0 from Sites WR12 to WR14 (in fig. 138, no calculations were possible for Sites WR15 and WR16). The zone from Sites WR12 to WR14 is also the zone that has the highest absolute radon levels. This observation suggests to us that excess radon is being contributed to the shallow ground waters from Site WR12 downstream to Site WR14. Upwelling ground waters recently in contact with bedrock are a likely source for the excess radon. Seeps and a spring pool are present near Sites WR14 and WR15 but such obvious *surface* evidence of upwelling waters is not present near Sites WR12 and WR13. However, we would suggest that upwelling bedrock-derived ground waters are likely in the subsurface near Sites WR12 and WR13.

Helium in the Ground-Water Samples

The helium content of the shallow ground waters in our sample sites along the valley floor ranged from 4,960 ppb at Site WR3 in 1986 to 6,860 ppb at Site WR14 in 1985 (table 67 and fig. 136). Waters taken from the spring pool near Site WR14 (S4) yielded 7,062 ppb in 1985 and 6,584 ppb in 1986. Waters taken from the slope spring above Site WR2 (S1) in 1985 yielded 5,327 ppb. Shallow ground waters along the valley floor yielded helium values close to or slightly below atmospheric levels except at Sites WR14 and WR15 which, in both 1985 and 1986, were considerably above atmospheric levels. Helium levels in 1986 were generally lower than those in 1985, except at Sites WR14 and WR15, where the values were similar overall. The lower 1986 values may result from a higher water table and thus greater contact of ground waters with biologic activity in soils on the hillslopes and in the surface layers of the valley-floor alluvium. In detail (fig. 137), the pattern of rising and falling values does not correspond from 1985 to 1986 except at Site WR3 (both years low) and Sites WR14 and WR15 (both years very high). In a fashion similar to that of radon, the observed helium values do not correspond in any simple way to calculated *GT* values for the sample sites (fig. 136), nor do observed values correspond with calculated helium yields for the near-surface (upper 45 cm) sediment layers (fig. 137; Helium yields were calculated from methods similar to those in appendix A for radon except that the emanating power for helium was assumed to be 1.00 on the basis of the surface adsorption suggested by the studies of Zielinski and others (1987) and on the basis of the small size and inertness of the helium atom. The calculated values shown in figure 137 are the helium yielded by the radioactive decay in the shallow sediment added to atmospheric helium at 5,240 ppb.).

Helium levels are significantly depressed at Site WR3, the beaver pond. At this site, we noted bubbling of gases from the sediment column, especially when it was disturbed during digging of the hole. High levels of carbon dioxide and methane, the highest observed in the study, occur in this water sample. Low helium levels at this site are likely due to dilution of the helium by biogenic carbon dioxide and methane produced by degradation of the organic matter in the sediment that has accumulated on the floor of the pond and by depletion of helium from the water by bubbling of carbon dioxide and methane through the water.

Highest helium values occur at Sites WR14 and WR15 and at the spring pool (fig. 136). This region is within the zone of upwelling ground waters inferred from the radon data. We are uncertain why the zone of elevated helium values seems more restricted than the zone of elevated radon values except that other gases are elevated at

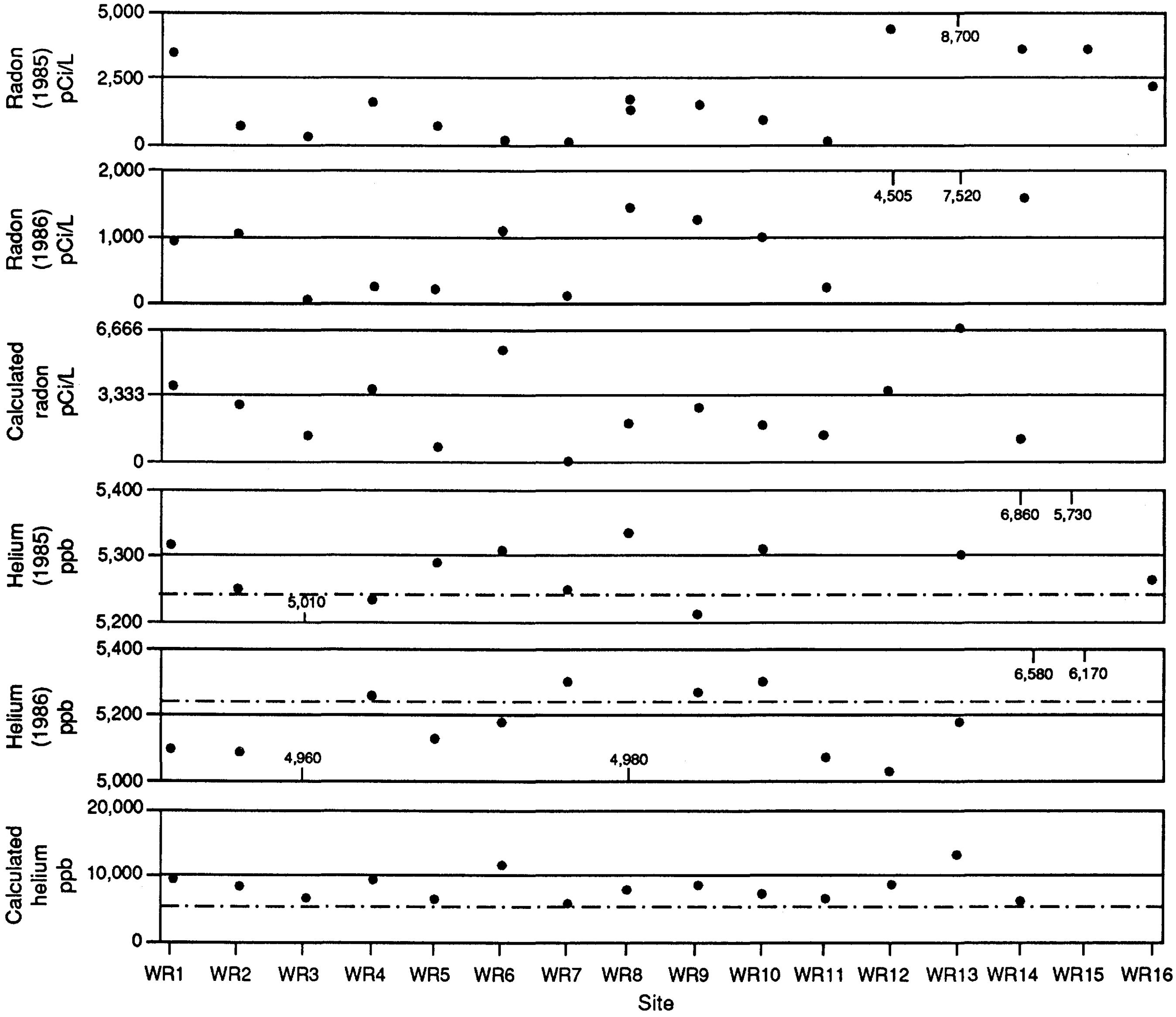

Figure 137. Radon and helium data for shallow ground waters from the 1985 and 1986 sampling in the study area, shown with an expanded scale for comparison with calculated radon and helium yields for the shallow sediment. Calculated radon and helium yields are not given for Sites WR15 and WR16 because uranium data were not available for these sites. Radon and helium values out of the range of the graphs are shown with leaders and numbers. Other missing data represent unsampled sites or lost samples.

Sites WR12 and WR13 but not at Sites WR14 and WR15 and the spring pool. Dilution or depletion of helium may occur at the first two sites.

Because helium, in contrast to radon, does not decay to other products while entrained in the water, the helium values observed are affected by gains and losses along the entire flow path of the water since first falling in the drainage basin as rainfall and becoming isolated from the atmosphere. From the present data, we cannot estimate the amount of helium entrained in the water from the hillslopes before it entered the valley-floor environment or entrained along its pathway through the valley floor, nor can we estimate the amount diluted by added biogenic and other gases along the way. Thus, we do not necessarily expect observed helium levels to be controlled by the uranium content of the sediment immediately surrounding the sample site. Specific environmental conditions like those at Sites WR3 (stagnant pond), WR14, and WR15 and the spring pool site (upwelling waters) may permit the observed helium values to correspond to the radon values (unusually low values at Site WR3 or unusually high at the latter sites). Dilution by added gases seems to be an important phenomenon overall, however, because about 30 percent of our

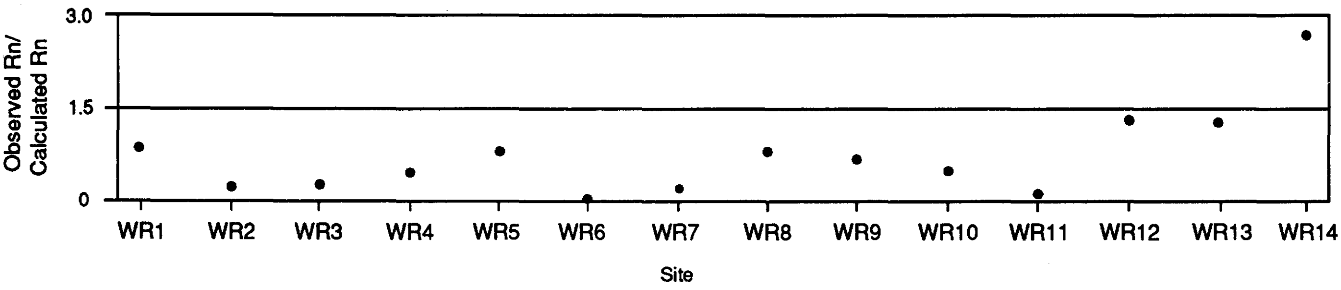

Figure 138. Ratio of observed to calculated radon levels for the shallow ground waters in the 1985 sampling. Sites 15 and 16 are not shown because uranium data were not available for these sites.

samples yielded values less than those expected for waters in equilibrium with the atmosphere.

Table 69. Oxygen, carbon dioxide, and methane data for selected sample sites and springs in northeast Washington, 1985

[Measured as a ratio to N_2]

Sample site	O_2/N_2	CO_2/N_2	CH_4/N_2
WR1	0.26535	0.0200	0.0019
WR2	.25910	.0307	.0017
WR3	.26253	.0847	.0234
WR5	.25879	.0128	.0037
WR7	.25883	.0187	.0051
WR8	.25868	.0202	.0051
WR9	.25669	.0217	.0016
WR10	.26360	.0122	.0018
WR11	.26296	.0055	.0000
WR12	.25690	.0593	.0011
WR13	.25883	.0252	.0033
WR14	.25290	.0055	.0000
WR15	.25513	.0085	.0000
WR16	.25729	.0237	.0019
S1 (valley slope spring) .	.27483	.0026	.0000
S4 (spring pool)	.25522	.0065	.0000
Reference gas	.27324	.0177	.0111
Reference gas[1]	.27287	.0175	.0101
Reference gas[1]	.27351	.0179	.0111

[1] Repeat measurements from the same reference gas cylinder.

Oxygen, Carbon Dioxide, and Methane in Ground Water

Oxygen, carbon dioxide, and methane were measured for 14 of 16 shallow ground-water sample sites and for the slope spring (S1, on the hillslope above Site WR2) and the spring pool (S4, near Site WR14). These data are reported in table 69 and figure 139 as ratios with respect to nitrogen (N_2) in the sample. If we assume that the nitrogen in the samples varies only slightly, we can measure the relative variation in the three other species.

Oxygen to nitrogen ratios varied within a narrow range for the shallow ground waters along the valley floor (0.253 to 0.265). The slope spring (S1) was the most oxygenated (0.275) of all the samples collected, whereas Site WR14, the spring pool (S4), and Site WR15 were the least oxygenated (0.253 to 0.255). Consumption of oxygen during oxidation of organic matter and by other reactions may deoxygenate waters. Deeply circulating ground waters in granitic rocks typically are very oxygen poor. The relatively low oxygen levels at Sites WR14 and WR15 and the spring pool may indicate a bedrock source.

Carbon dioxide to nitrogen ratios along the valley floor varied from 0.00550 to 0.08468; however, the slope spring was the least carbon dioxide rich (0.00260). Carbon dioxide values were highest at Site WR3 (the beaver pond) and at Site WR12 and lowest at Sites WR11 and WR14, the spring pool, and Site WR15. Carbon dioxide levels in this system are likely raised by the oxidation of organic matter in contact with the shallow ground water. The relatively low carbon dioxide values at Sites WR11 and WR14, the spring pool, and Site WR15 are atypical of the other shallow ground waters and may reflect a different origin for these waters.

Methane to nitrogen ratios along the valley floor varied from 0.00000 (not detected) to 0.02344. The highest methane was measured at Site WR3 (the beaver pond) at a level more than 4 times that of the next highest sample, whereas methane was not detected at the slope spring site (S1), Sites WR11 and WR14, the spring pool, and Site WR15. The high methane level at the beaver pond site is not surprising, because these stagnant waters and the underlying sediment at depth are likely hosts to methanogenic bacteria. A low methane level at the slope spring site is also not surprising, because these waters are flowing, are oxygenated, and have been in recent contact with the atmosphere and bedrock. Zero methane values for Sites WR11, WR14, and WR15 and the spring pool are surprising, because they are much lower than at other sites along the valley floor, yet the sediment column at these sites contains abundant organic matter. These four waters may represent ground waters upwelling through bedrock that have only limited contact with the organic-rich sediment because the

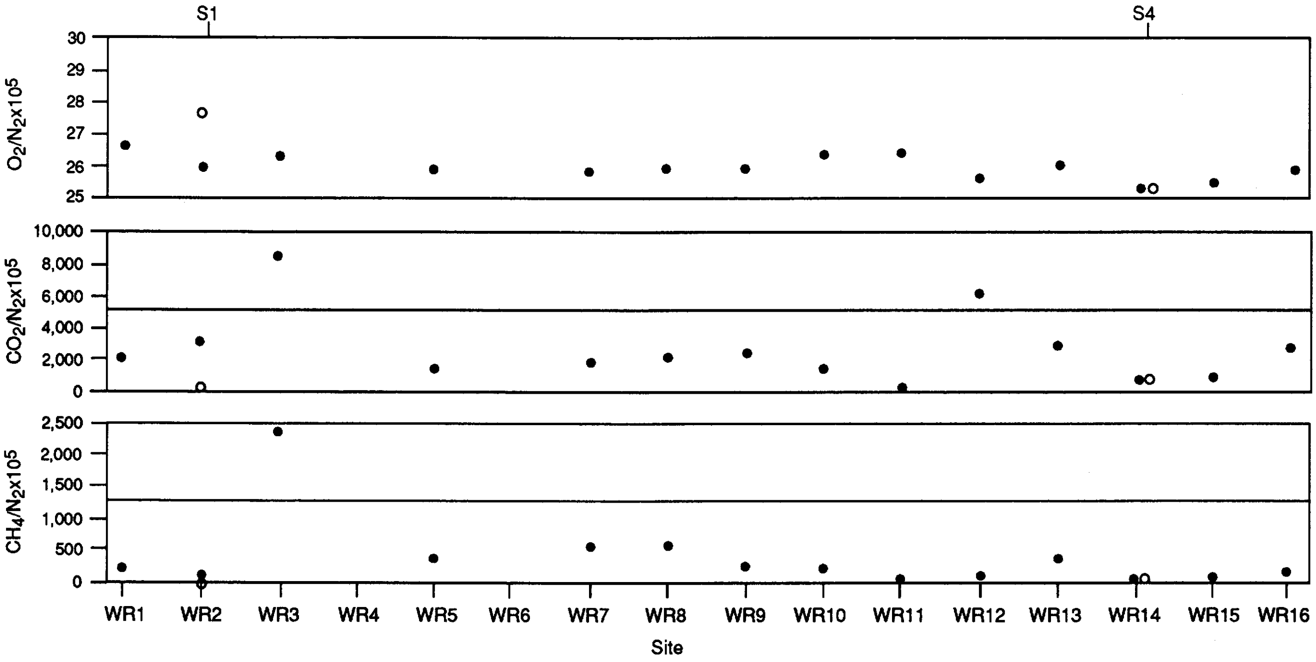

Figure 139. Oxygen, carbon dioxide, and methane data for the 1985 samples. Data are reported as ratios with respect to nitrogen. Samples WR4 and WR6 were lost from sample cylinders in transit from field to laboratory.

transit time through the sediment column is rapid. Sites WR14 and WR15 and the spring pool are in a zone of waters suspected of upwelling through bedrock, and Site WR11 is immediately downvalley from a slope spring. The bedrock source interpretation for Sites WR14 and WR15 and the spring pool is compatible with our interpretation of the oxygen and carbon dioxide data.

Four end-member assemblages of dissolved gases seem to be present in these shallow ground waters: (1) beaver pond waters that are methane and carbon dioxide rich; (2) slope spring waters that are oxygen rich and methane and carbon dioxide poor; (3) "normal" valley-floor waters that have modest levels of methane, oxygen, and carbon dioxide varying over a limited range; and (4) valley-bottom upwelling waters that are oxygen, carbon dioxide, and methane poor. The chemistry of the beaver pond waters is controlled by the intense biologic activity in the surface layers (carbon-dioxide generating) and subsurface layers (methanogenic). The chemistry of the slope spring waters may indicate limited interaction with soil and shallow interflow along the bedrock-till interface.

CONCLUSIONS

Sampling dissolved gases from waters in springs and shallow holes dug in a small drainage basin is an effective method for characterizing the shallow ground water of the basin. The holes must be dug so that standing water from above the ground surface is not admitted, and these holes must intercept a water-bearing interval. Sites should be selected carefully late in the dry season. The holes should be checked to confirm that they are seeping; if not, a deeper hole must be dug.

The radon and helium concentrations in the shallow ground waters of the valley-floor sediment along the upper part of the north fork of Flodelle Creek are much lower than those expected for uranium ore deposits of this grade. The lower values are largely the result of the gross disequilibrium between uranium and its decay products in the very young surficial uranium deposit hosted by these sediments. Because of this disequilibrium, measurement of radon or helium in shallow ground waters is not likely to be effective in the search for surficial uranium deposits.

Ground waters apparently derived from the bedrock have higher levels of radon associated with them than ground waters whose most recent contact has been with the valley-floor sediment alone. If so, measurement of radon in ground water, and, to a lesser degree, measurement of helium in ground water, may be an effective technique for examining the flux of ground water from the underlying bedrock into the sediment column of the valley floor. This technique is analogous to measuring the flux of radon in streams as a measure of the ground-water input to the surface flow (Lee and Hollyday, 1987). Sampling of ground waters for radon could have broader application to the study of the ground-water budget of wetlands in other areas where this contrast in radon levels exists.

The ground waters in the drainage basin show radon, helium, oxygen, carbon dioxide, and methane concentrations characteristic of the environment in which the waters occur or have recently traveled through. Stagnant waters from the sediment of a beaver pond in the upper part of the study area are high in carbon dioxide and methane, because the organic matter in the sediment column has been oxidized and biogenically degraded. These waters have low levels of helium and radon because these gases have been diluted by addition of the biogenic gases and depleted by bubbling of carbon dioxide and methane through the sediment column. Ground waters from springs on the hillslopes are high in oxygen but relatively low in carbon dioxide and methane because the waters have had limited contact with the thin soil horizons, and these horizons are low in organic matter. Such waters are high in radon probably because they have had recent contact with uranium-rich weathered bedrock and sandy glacial till; the waters are slightly below atmospheric levels in helium. "Normal" ground waters in the sediment filling the valley floor contain modest amounts of radon, helium, oxygen, carbon dioxide, and methane because these waters have had recent contact with biologically active, uraniferous sediment in the upper part of the sediment column of the valley floor. Ground waters in a zone of upwelling along the valley floor are very low in oxygen, carbon dioxide, and methane but are very high in radon and (or) helium. A bedrock source is thus suggested for the upwelling water, possibly a fracture zone intercepting the valley floor, as inferred by Zielinski and Schumann (1987).

APPENDIX A. CALCULATION OF EXPECTED RADON YIELD

For calculation of the expected radon in the shallow ground water at the sample site, we assume that the sediment contributing to the water in the sample hole lies within the upper 45 cm of the surface (average depth of our sample holes) and that the nearby auger hole is representative of that interval. The radon content of water in a representative 1-L (1,000 cm^3) volume of water-saturated sediment is given by

$$\text{Rn (in pCi/L)} = \frac{1{,}000\ \text{cm}^3/\text{L} \times d \times c \times a_{\text{U}} \times e \times p}{f}$$

where d is the dry density of the sediment (in grams per cubic centimeter, estimated by comparison with measurements of organic-rich sediment here and elsewhere (Otton, unpub. data, 1985); c is the uranium content (in micrograms per gram) as determined in the nearby auger hole; a_{U} is the specific activity of uranium at secular equilibrium (0.33 pCi/μg); e is the fractional equilibrium factor estimated by comparison with a study site near Site WR14 (Zielinski and others, 1987) where the fractional equilibrium ranged from 0.01 to 0.04 for the near surface layers; p is the emanating power (the ratio of radon escaping to the pores to radon produced by the decay of radium in the sample) taken as 0.33 (emanating power for "lignitic" soils reported by Barretto and others, 1975); and f is the fraction of water making up the 1-L volume of water-saturated sediment.

Note that weighted averages for dry density, uranium content, and fractional equilibrium were estimated or calculated for the 45-cm interval from the descriptions of and analytical data for the sediment at each site. Although extensive measurements of emanating power of mineral matter in air have been made, the emanating power of water-saturated sediment is not well known.

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