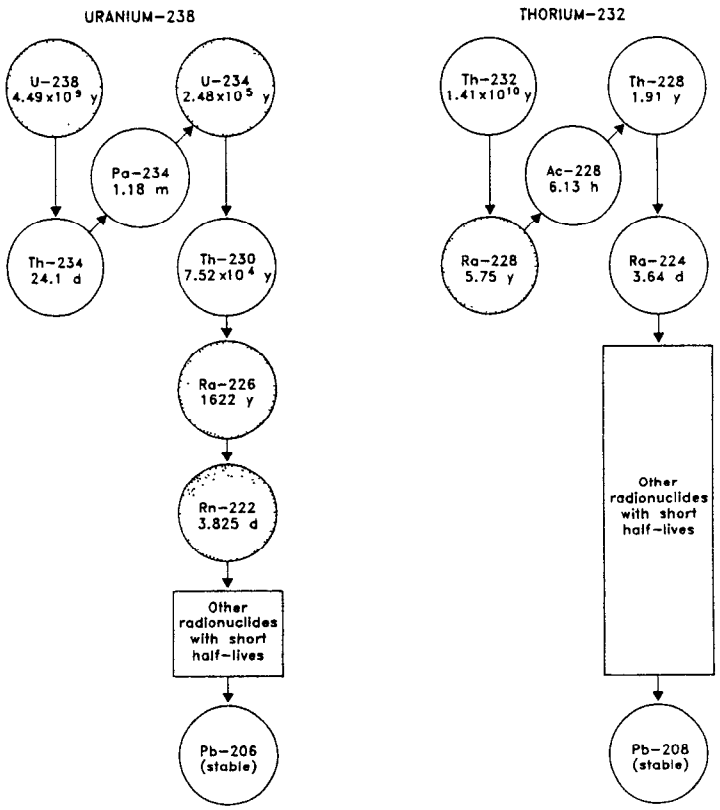
Water-Resources Investigations Report 92-4092

GEOCHEMISTRY OF AND RADIOACTIVITY IN GROUND WATER OF THE HIGHLAND RIM AND CENTRAL BASIN AQUIFER SYSTEMS, HICKMAN AND MAURY COUNTIES, TENNESSEE



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GEOCHEMISTRY OF AND RADIOACTIVITY IN GROUND WATER OF THE HIGHLAND RIM AND CENTRAL BASIN AQUIFER SYSTEMS, HICKMAN AND MAURY COUNTIES, TENNESSEE

By Gregg E. Hileman and Roger W. Lee

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Nashville, Tennessee

1993

U.S. DEPARTMENT OF THE INTERIOR
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CONVERSION FACTORS, VERTICAL DATUM, AND ABBREVIATED WATER-QUALITY UNITS

Multiply	By	To obtain
foot (ft)	0.3048	meter
mile (mi)	1.609	kilometer

Sea level: In this report "sea level" refers to the National Geodetic Vertical Datum of 1929—a geodetic datum derived from a general adjustment of the first-order level nets of the United States and Canada, formerly called Sea Level Datum of 1929.

Water-quality units

mg/L	milligrams per liter
pCi/L	picocuries per liter
μS/cm	microsiemens per centimeter at 25 degrees Celsius

Other Abbreviations

R^2	coefficient of correlation squared

GEOCHEMISTRY OF AND RADIOACTIVITY IN GROUND WATER OF THE HIGHLAND RIM AND CENTRAL BASIN AQUIFER SYSTEMS, HICKMAN AND MAURY COUNTIES, TENNESSEE

By Gregg E. Hileman and Roger W. Lee

ABSTRACT

A reconnaissance of the geochemistry of and radioactivity in ground water from the Highland Rim and Central Basin aquifer systems in Hickman and Maury Counties, Tennessee, was conducted in 1989. Water in both aquifer systems typically is of the calcium or calcium magnesium bicarbonate type, but concentrations of calcium, magnesium, sodium, potassium, chloride, and sulfate are greater in water of the Central Basin system; differences in the concentrations are statistically significant. Dissolution of calcite, magnesium-calcite, dolomite, and gypsum are the primary geochemical processes controlling groundwater chemistry in both aquifer systems. Saturation-state calculations using the computer code WATEQF indicated that ground water from the Central Basin system is more saturated with respect to calcite, dolomite, and gypsum than water from the Highland Rim system. Geochemical environments within each aquifer system are somewhat different with respect to dissolution of magnesium-bearing minerals. Water samples from the Highland Rim system had a fairly constant calcium to magnesium molar ratio, implying congruent dissolution of magnesium-bearing minerals, whereas water samples from the Central Basin system had highly variable ratios, implying either incongruent dissolution or heterogeneity in soluble constituents of the aquifer matrix.

Concentrations of radionuclides in water were low and not greatly different between aquifer systems. Median gross alpha activities were 0.54 picocuries per liter in water from each system; median gross beta activities were 1.1 and 2.3 picocuries per liter in water from the Highland Rim and Central Basin systems, respectively. Radon-222 concentrations were 559 and 422 picocuries per liter, respectively. Concentrations of gross alpha and radium in all samples were substantially less than Tennessee's maximum permissible levels for community water-supply systems. The data indicated no relations between concentrations of dissolved radionuclides (uranium, radium-226, radium-228, radon-222, gross alpha, and gross beta) and any key indicators of water chemistry, except in water from the Highland Rim system, in which radon-222 was moderately related to pH and weakly related to dissolved magnesium. The only relation among radiochemical constituents indicated by the data was between radium-226 and gross alpha activity; this relation was indicated for water from both aquifer systems.

INTRODUCTION

Much of the ground water used for domestic and municipal supply in Middle Tennessee is withdrawn from two regional aquifer systems: the Highland Rim aquifer system and the Central Basin aquifer system. Both systems are comprised of sedimentary rock that includes phosphatic limestone and shale. Low-level radioactivity commonly is associated with phosphatic limestones and black shales, and results from the incorporation of uranium and other radionuclides into the rock matrix. Because the radioactive constituents are soluble, ground water percolating through these rocks has acquired small concentrations of radionuclides.

Despite awareness that ground water from these aquifer systems contains low concentrations of radionuclides and that the long-term ingestion of water containing radioactive matter presents potential health risks, little study has been made of the identity and concentrations of radionuclides in ground water or of the geochemical environments in which they are either gained or removed from solution. To provide water managers with data necessary to ensure the suitability of the water

source for public and domestic supply, and to enhance knowledge of the geochemistry of these systems, the U.S. Geological Survey (USGS), in cooperation with the Tennessee Department of Environment and Conservation (formerly called the Tennessee Department of Health and Environment), conducted a reconnaissance of the quality of ground water from the two regional aquifer systems during spring and summer 1989. The study was limited to Hickman and Maury Counties (fig. 1), which were selected as representative of areas where each aquifer system occurs within a few hundred feet of land surface.

Objectives of the study were to identify and measure concentrations of selected radionuclides in ground water from each of the two aquifer systems, to characterize the geochemical environment of ground water flowing through the two aquifer systems, and to determine if a relation exists between water geochemistry and radionuclide concentrations. The first objective was met with the publication of data collected during the investigation (Hileman, 1990).

Purpose and Scope

The purpose of this report is to present interpretive results of the study described above. The report describes the hydrogeology of the area, summarizes results of the chemical analyses, discusses major geochemical processes, and examines possible relations between radionuclides and some of the constituents for which analyses were made.

To characterize water quality in the Highland Rim and Central Basin aquifer systems, 17 wells and 3 springs from Hickman County and 20 wells from Maury County were sampled and the water analyzed for selected water-quality constituents and characteristics. Chemical and radiochemical analyses of ground water from the two aquifer systems are summarized and compared. Geochemical environments are characterized by calculating saturation indexes of key minerals, and major geochemical processes are discussed. Relations between radiochemical constituents and key indicators of water chemistry and relations among radiochemical constituents in ground water are investigated.

Description of the Study Area

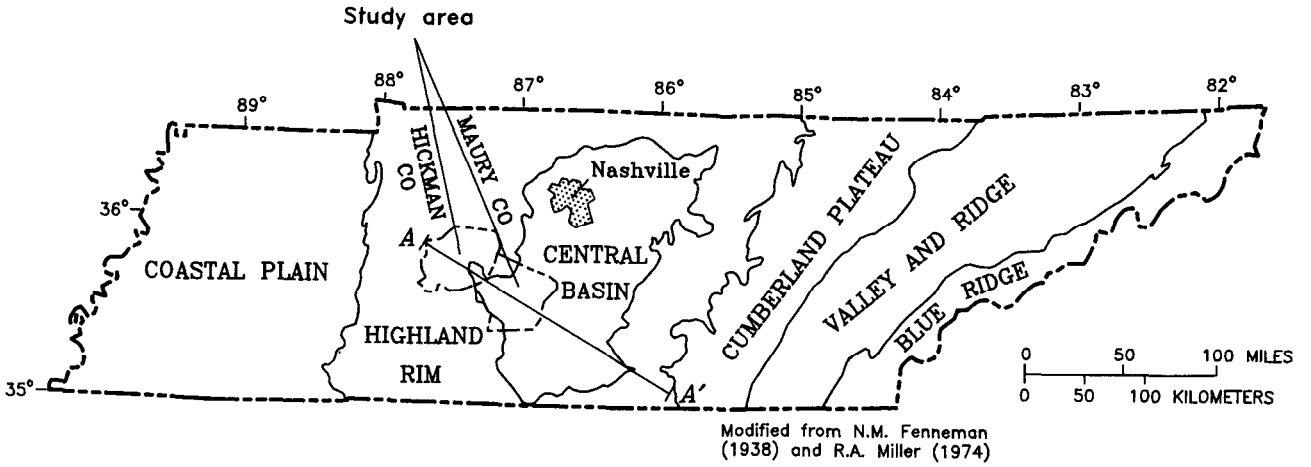
Hickman and Maury Counties are located about 25 to 50 miles southwest of the Nashville city limit in Middle Tennessee. Most of Hickman County and the northwestern and southwestern corners of Maury County are included in the Highland Rim physiographic province, whereas much of Maury County and part of the Duck River valley in Hickman County are included in the Central Basin province (figs. 1 and 2). The Highland Rim province within the study area is a rolling upland that stands about 300 feet above the Central Basin. Most of the isolated hills in western Maury County are erosional remnants of the Highland Rim. The Central Basin in this area is characterized by gently rolling hills drained by the Duck River. The boundary between the two provinces is marked by an escarpment of minor relief. Elevations range from about 400 feet above sea level along the Duck River in western Hickman County to 1,100 feet above sea level along the drainage divide between the Duck and Elk Rivers in southern Maury County.

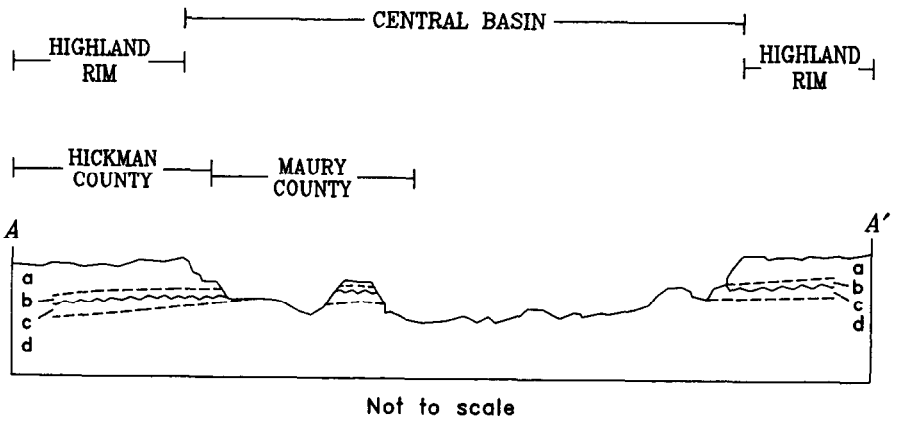
Land use is primarily agricultural except where communities have become established. The population of Hickman County in 1990 was 16,754 and 54,812 in Maury County (U.S. Department of Commerce, 1991). Mean annual rainfall of this area is about 47 inches (National Oceanic and Atmospheric Administration, 1987).

HYDROGEOLOGIC SETTING

The Highland Rim aquifer system and the Central Basin aquifer system are the primary sources of ground water in Hickman and Maury Counties. These aquifer systems have been described by Brahana and Bradley (1986a, 1986b) and are named for the physiographic provinces where they are located. Formations comprising these aquifer systems range in age from Ordovician to Mississippian (table 1).

Ground water in both aquifer systems generally is near land surface (within 300 feet). Both systems are recharged by precipitation. Ground water moves through soil and regolith, to gravel-rich zones within the regolith, then into solution openings in the underlying bedrock. Ground water





EXPLANATION

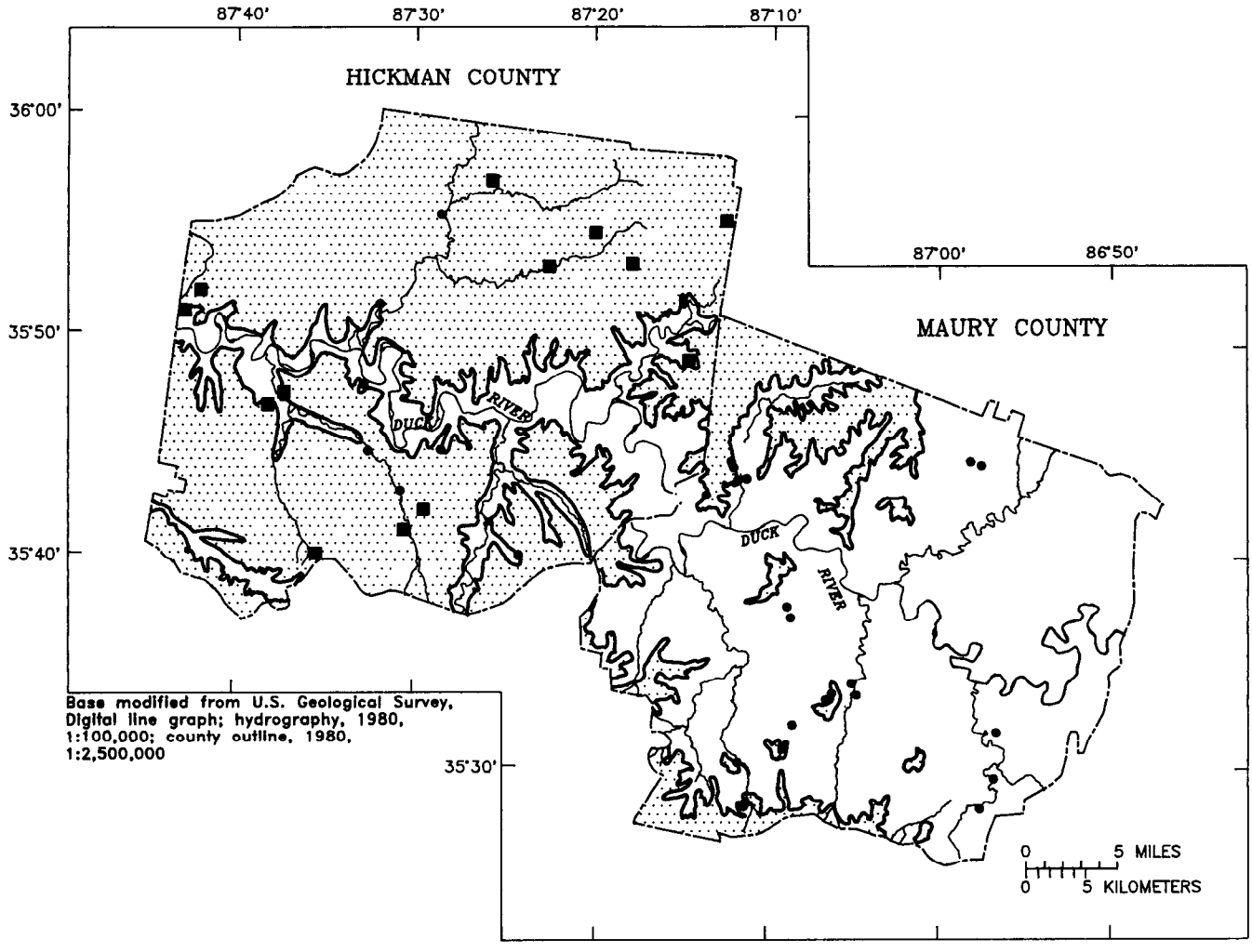
a – MISSISSIPPIAN SYSTEM
b – DEVONIAN SYSTEM
HIGHLAND RIM AQUIFER SYSTEM

c – SILURIAN SYSTEM
d – ORDOVICIAN SYSTEM
CENTRAL BASIN AQUIFER SYSTEM

CONTACT BETWEEN SYSTEMS

A – A′ TRACE ON MAP AND LINE OF SECTION

Figure 1. Generalized physiographic provinces of Tennessee, study area, and idealized section through part of the Highland Rim and Central Basin provinces.



EXPLANATION

OUTCROP AREA OF THE HIGHLAND RIM AQUIFER SYSTEM--Corresponds to Highland Rim physiographic province within the study area

OUTCROP AREA OF THE CENTRAL BASIN AQUIFER SYSTEM--Corresponds to Central Basin physiographic province within the study area

■ WELL OR SPRING OPEN TO THE HIGHLAND RIM AQUIFER SYSTEM

• WELL OPEN TO THE CENTRAL BASIN AQUIFER SYSTEM

Figure 2. Location of wells and springs sampled for study.

Table 1. Bedrock units underlying the study area

[Modified from Brahana and Bradley, 1986a, 1986b]

Age	Group	Formation	Thickness (feet)	Lithology
Mississippian		St. Louis Limestone	80-175	Limestone, generally massive where unweathered.
		Warsaw Limestone	100	Limestone, massive; shale; sandstone.
		Fort Payne Formation	100-350	Limestone, with dolomite, siltstone, and chert stringers. Evaporites present locally.
		Maury Shale	1-4	Shale, mudstone, and siltstone; glauconite, with abundant phosphate nodules.
		Chattanooga Shale	5-20	Shale, black, fissile, uraniferous.
Devonian		Brownsport Formation	0-80	Limestone and shale in alternating thin beds.
Silurian	Wayne	Lego Limestone	0-45	Limestone.
		Waldron Shale	0-5	Shale and shaly limestone.
		Laurel Limestone	0-30	Limestone.
		Osgood Limestone	0-15	Shale and limestone, argillaceous.
Ordovician		Brassfield Limestone	0-130	Limestone, cherty and locally glauconitic.
		Mannie Shale	0-20	Shale and argillaceous limestone.
		Fernvale Limestone	0-30	Limestone and shale.
		Leipers Formation	0-75	Limestone with interbedded shale; phosphatic.
	Nashville	Catheys Formation	125-400	Limestone with shale, locally phosphatic.
		Bigby Limestone	60-150	Limestone, sandy; has phosphatic facies.
		Hermitage Formation	50-100	Limestone, shaly, sandy, phosphatic.
	Stones River	Carters Limestone	65-250	Limestone; contains four thin beds of bentonite.
		Lebanon Limestone	115	Limestone.
		Ridley Limestone	100	Limestone, dolomite.
		Pierce Limestone	25	Limestone, shaly.
		Murfreesboro Limestone	425	Limestone, massive, cherty.
		Pond Spring Formation	0-150	Limestone, silty dolomite, dolomitic limestone.

from the aquifer systems discharges through seeps and springs and as base flow to area streams.

The Highland Rim aquifer system within the study area includes the Mississippian St. Louis Limestone, Warsaw Limestone, Fort Payne Formation, and Maury Shale (Brahana and Bradley, 1986a). For the purposes of this report, the Chattanooga Shale, a regional confining unit, also is assigned to the Highland Rim system. The St. Louis Limestone generally is represented only by residual clay with blocks of siliceous limestone. The Warsaw Limestone is typically a fine- to coarse-grained fossil fragmental limestone. The Fort Payne Formation consists of argillaceous and siliceous limestone and calcareous siltstone, and includes much chert, both nodular and bedded. In some areas, gypsum and other evaporites are present. The Maury Shale, a thin, glauconitic formation containing phosphatic nodules, lies between the Fort Payne Formation and the Chattanooga Shale. Total thickness of formations of the Highland Rim aquifer system in the study area ranges from about 300 to 600 feet.

The Warsaw Limestone and Fort Payne Formation weather deeply, producing a clay-rich, and in the case of the Fort Payne Formation, a chert-rich regolith as much as 100 feet thick. The regolith contains a reservoir of water that supplies ground water to openings along bedding planes and solution-enlarged fractures in the bedrock. These openings form by the dissolution of soluble minerals, mostly carbonates, reacting with slightly acidic recharge water. The openings represent secondary porosity and permeability in an otherwise nonporous rock mass and are highly irregular in their distribution.

The Chattanooga Shale is a black shale rich in organic matter that separates the Highland Rim aquifer system from the Central Basin aquifer system and plays a significant role in the hydrology of the area. The Chattanooga Shale, although not thick in the study area, is considered to be an effective barrier to the vertical flow of ground water between the Highland Rim aquifer system and the underlying Central Basin aquifer system. In addition, the shale might affect water chemistry. Iron sulfide, radionuclides, and other constituents in the shale might dissolve in water that comes in contact with the rock unit, either along its surface or in fractures or partings within the rock.

Sources of uranium in rocks of the Highland Rim aquifer system include phosphatic nodules of the Maury Shale, and probably the entire section of the Chattanooga Shale. The Gassaway (or upper) Member of the Chattanooga Shale contains about 0.006 percent uranium (Swanson, 1961). This uranium probably does not occur as a distinct uranium mineral, but rather is associated with organic matter - pyrite complexes (Bates and others, 1954). The Hardin Sandstone (or lowest) Member of the Chattanooga Shale is a nodular phosphatic zone, and exposures along one of the tributaries to the Duck River in Hickman County were formerly mined commercially for "blue rock" phosphate (Theis, 1936, p.110). In phosphatic rock, uranium commonly substitutes for calcium in fluorapatite [$Ca_5(PO_4)^3F$] (Altschuler and others, 1958).

The Central Basin aquifer system includes the Silurian- and Ordovician-age formations from the base of the Chattanooga Shale to the top of the lower Ordovician Knox Group. The Central Basin aquifer system includes numerous interbedded limestone, shale, and siltstone units and various admixtures of these lithotypes. Limestones in this stratigraphic interval are much less siliceous than those of the Mississippian system, but commonly are argillaceous and locally, glauconitic. Limestones of some of the Ordovician formations are massive; some are dolomitic. Although thickness of the formations in the Central Basin aquifer system in the study area is over 1,000 feet, most of the water-bearing units are within a few hundred feet of land surface.

The hydraulic properties of the Central Basin system generally resemble those of the Highland Rim system. Recharge flows through openings along bedding planes and solution-enlarged fractures that are highly irregular in distribution. The rock mass itself has little or no primary permeability. Regolith in the Central Basin is thin, generally 20 feet or less, thus storage capacity is limited.

Uranium and other radionuclides in rocks of the Central Basin aquifer system most commonly are formed in phosphate- and shale-rich zones. The Leipers Formation, Catheys Formation, Bigby Limestone, and Hermitage Formation (all of Ordovician age) contain phosphatic intervals or facies. At the time of Theis' (1936) study, the weathered Bigby Limestone was the most

important source of phosphate in Tennessee. Phosphate nodules from Ordovician rocks in Middle Tennessee are typically 0.003 percent uranium (Altschuler and others, 1958), whereas nonphosphatic rocks such as chert and "pure" limestone commonly are less than 0.0005 percent (Swanson, 1961). These phosphatic limestones and numerous shales in the aquifer system could be sources of dissolved uranium and other radionuclides in the ground-water supply.

GEOCHEMISTRY OF GROUND WATER

The Highland Rim aquifer system and the Central Basin aquifer system are both primarily limestone aquifers. However, considerable difference exists in the mineralogy and geochemistry of these two aquifer systems and between the individual formations within each system. To provide an understanding of the geochemistry and geochemical processes operating within the two aquifer systems, inorganic chemical characteristics of water from each system were analyzed, saturation indexes of certain ions were determined, and key geochemical processes were evaluated.

Major Constituents and Physical Properties

Seventeen wells and 3 springs in Hickman County and 20 wells in Maury County were sampled for this investigation (fig. 2). All of the sites are used for domestic supply. The geologic unit supplying water to each well was determined by examination of drillers' logs and information provided by well owners. Of the Hickman County samples, 13 were interpreted to be from sources tapping the Highland Rim aquifer system, and 7 from sources tapping the Central Basin aquifer system. All Maury County wells tapped the Central Basin aquifer system. Thus, the total number of sites in the Highland Rim and Central Basin aquifer systems were 13 and 27, respectively.

Samples were collected from existing plumbing lines and treated in accordance with current USGS guidelines. Alkalinity, pH, temperature, and specific conductance were measured in the field at each site. Concentrations of principal cations and anions, trace metals, and total organic carbon were determined in the laboratory. A description of sample collection and treatment is included in the data report of this study (Hileman, 1990).

Summary statistics (table 2) describe the quality of water from each aquifer system. Boxplots presented in figure 3 for several constituents in water provide a visual comparison of the quality of water from each system.

Most of the water-quality measures indicate that moderate differences exist between the aquifer systems. Median values for the specific conductance of water from the Highland Rim and Central Basin aquifer systems were 150 and 500 μS/cm, respectively; for dissolved solids, 101 and 271 mg/L; for alkalinity, 73 and 206 mg/L; and for pH, 7.2 and 7.2 standard units. Each measure showed greater variation for the Central Basin system, which might partly result from the greater number of samples representing that system and the greater number of formations sampled.

Concentrations of many ions typically are greater in water of the Central Basin system than in the Highland Rim system, as indicated by larger mean and median values for calcium, magnesium, sodium, potassium, chloride, sulfate, fluoride, phosphorous, organic carbon, boron, and most of the trace metals (table 2). Average silica and barium concentrations in water from the two systems are about equal. In contrast, the mean and median concentrates of dissolved oxygen in water from the Highland Rim system are two to three times greater than corresponding values for the Central Basin system.

All 13 of the samples collected from the Highland Rim aquifer system were of the calcium bicarbonate type. The overall similarity in major ion composition of water from this system is shown by the clustering of points on the three parts of a trilinear diagram (fig. 4). The composition of water in samples from the Central Basin system showed more variability than did samples from the Highland Rim system. Of the 27 samples collected, 18 were of the calcium bicarbonate type; 3 were of the calcium magnesium bicarbonate type; 3 were of the calcium sulfate type; 1 was a calcium magnesium sulfate type; and 2, of mixed types. Variability in the major ion composition of water from the Central Basin system is shown by the

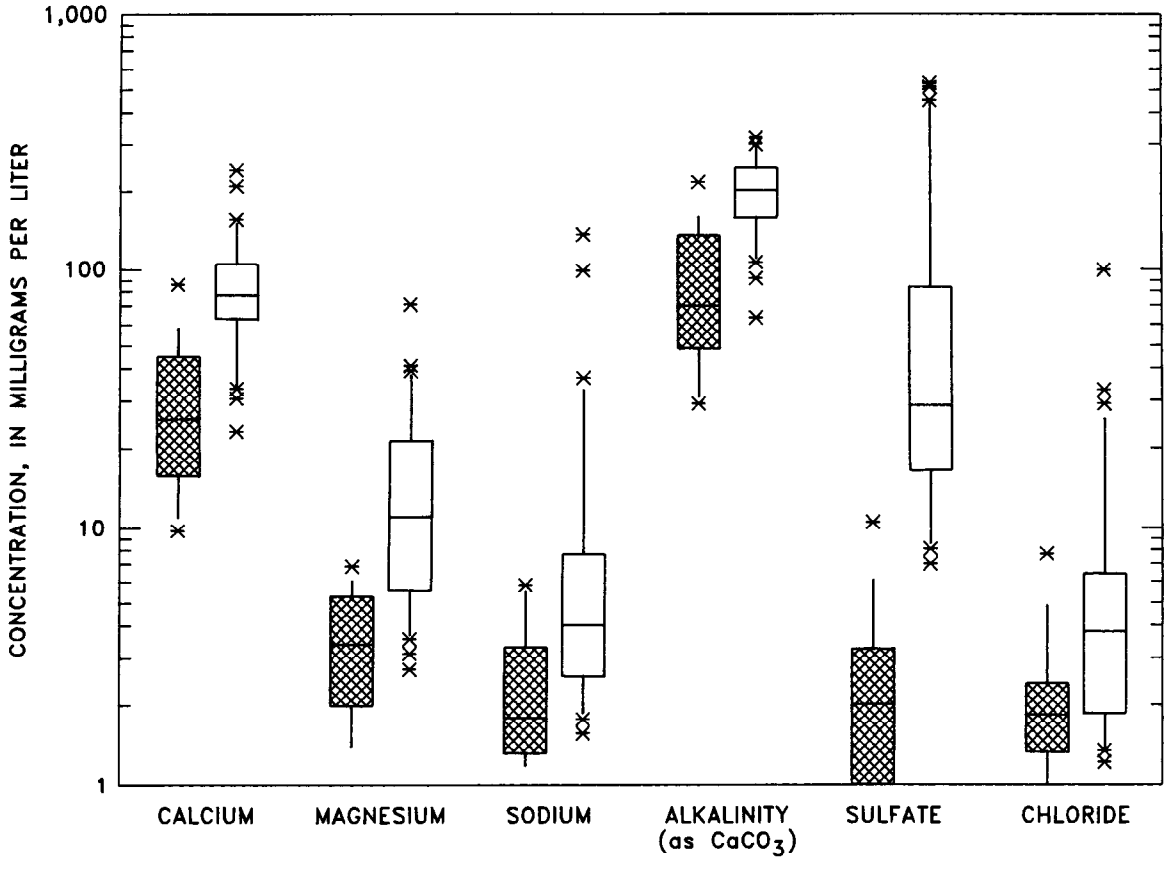
Table 2. Summary statistics of ground-water quality, exclusive of radionuclides, Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee

[All constituents are dissolved, in milligrams per liter unless otherwise specified; n, number of samples; μS/cm, microsiemens per centimeter; *, value estimated by using a log-probability regression to predict the values of data below the detection limit; °C, degrees Celsius; <, less than; μg/L, micrograms per liter]

Constituent or property	Highland Rim aquifer system					Central Basin aquifer system				
	n	Maximum	Minimum	Mean	Median	n	Maximum	Minimum	Mean	Median
Temperature (°C)	13	17	14	15.6	15.5	27	20	14.5	16.6	16.5
pH (standard units).	13	7.6	6.5	7.0	7.2	26	7.9	6.4	7.2	7.2
Specific conductance (μS/cm).	13	450	60	189	150	27	1,610	135	606	500
Solids	13	253	26	106.7	101	27	1,160	85	371	271
Total alkalinity (as $CaCO_3$).	13	224	30	92	73	27	334	64	202	206
Oxygen	13	8.0	2.0	5.6	5.5	25	7.0	.14	2.2	1.7
Total organic carbon (as C).	13	.3	<.1	.1*	.1*	26	1.4	<.1	.4*	.3*
Calcium	13	86	10	32	26	27	250	24	92	81
Magnesium	13	7.1	1.1	3.6	3.5	27	74	2.8	17	11
Sodium	13	6.0	1.2	2.6	1.8	27	140	1.1	14.7	4.1
Potassium	13	4.8	.1	.69	.4	27	5.2	.4	1.6	1.0
Chloride	13	7.6	1.0	2.3	1.8	27	99	1.0	9.6	3.8
Fluoride	13	.1	<.1	.1*	.1*	27	4.0	.1	.6	.3
Phosphate (as P).	10	.05	<.01	.02*	.02*	27	.63	<.01	.11*	.05*
Sulfate	13	10	<1	3*	2*	27	550	7	111	29

Table 2. Summary statistics of ground-water quality, exclusive of radionuclides, Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee--*Continued*

Constituent or property	Highland Rim aquifer system					Central Basin aquifer system				
	n	Maximum	Minimum	Mean	Median	n	Maximum	Minimum	Mean	Median
Nitrite plus nitrate (as N).	10	2.5	<0.1	0.69*	0.29*	27	7.8	<0.1	0.91*	0.40
Ammonia (as N).	10	.03	<.01	.02*	.02*	27	.34	<.01	.05*	.02*
Silica	13	9.2	7.9	8.6	8.7	27	11	4.9	8.6	8.7
Iron (µg/L)	13	9.0	<3	4.5*	4.0*	27	1,000	<3	67*	13*
Manganese (µg/L).	13	25	<1	3*	1*	27	51	<1	7*	3*
Strontium (µg/L).	13	350	18	96	79	27	8,100	43	1,992	880
Barium (µg/L)	13	56	11	26	25	27	130	4.0	26	19



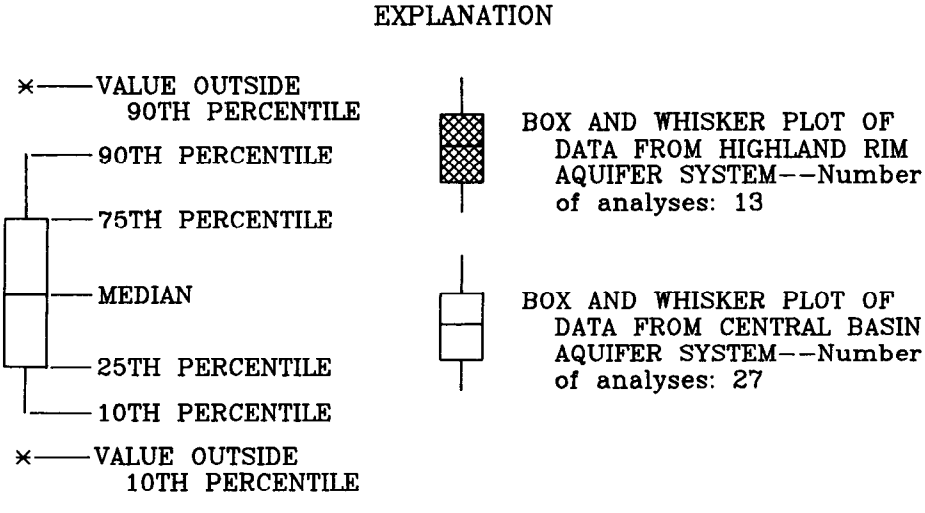


Figure 3. Major ion constituents in ground water of the Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee. Minimum reporting level for analytical methods is 1 milligram per liter.

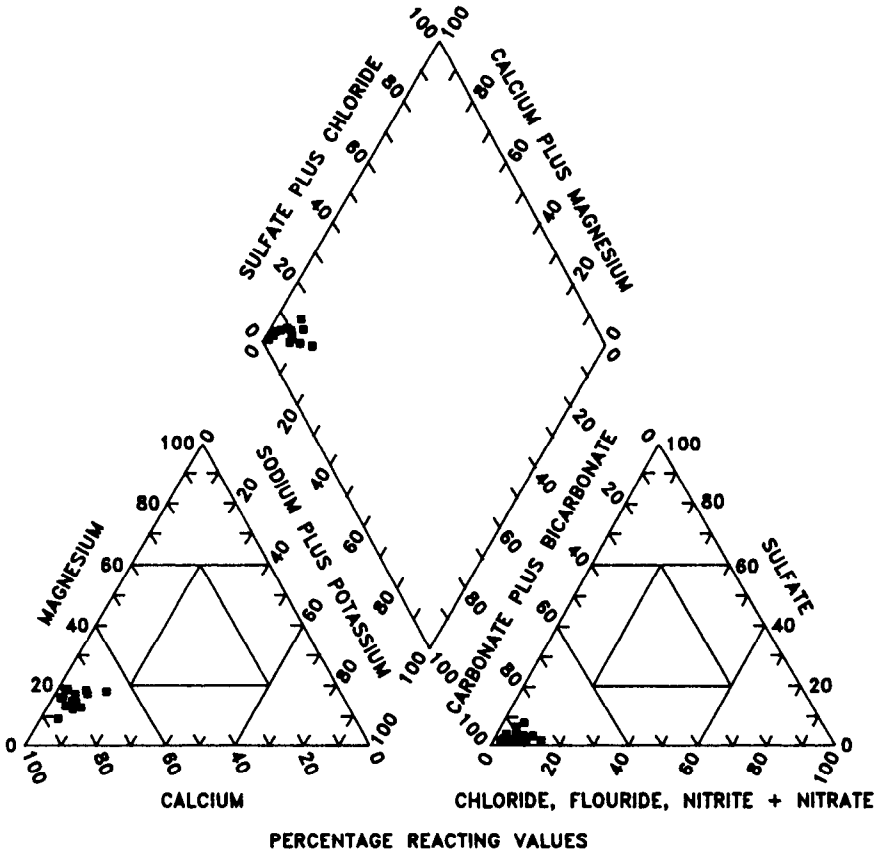


Figure 4. Trilinear diagram illustrating principal ion composition of ground water at 13 sites tapping the Highland Rim aquifer system in Hickman County, Tennessee. Percentages are computed from concentrations in milliequivalents per liter.

scattering of points on the three parts of a trilinear diagram (fig. 5).

Differences and similarities between the ground-water chemistries of the Highland Rim and Central Basin aquifer systems are supported by statistical comparison of the data sets. The Wilcoxon-Mann-Whitney rank sum test (Iman and Conover, 1983) was used to test for statistically significant differences between the two aquifer systems for the constituents and physical characteristics listed in table 2.

Results of the statistical testing indicated differences between the two aquifer systems at the 95-percent confidence level for temperature, specific conductance, dissolved solids, alkalinity, dissolved oxygen, organic carbon, calcium, magnesium, sodium, potassium, chloride, sulfate, iron, strontium, and fluoride. These results support differences observed in data presented in figure 3 and table 2. The statistical test failed to indicate any significant differences (at the 95-percent confidence level) between the two aquifer systems for pH, dissolved silica, ammonium, nitrite plus nitrate, phosphorus, manganese, and barium.

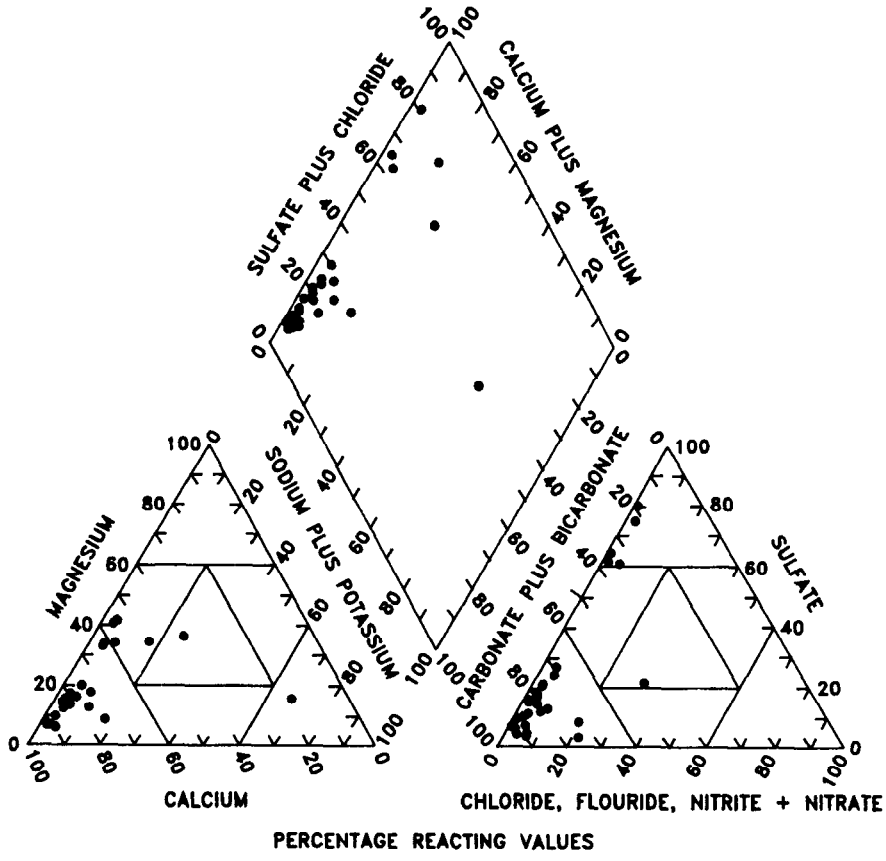


Figure 5. Trilinear diagram illustrating principal ion composition of ground water at 27 sites tapping the Central Basin aquifer system in Hickman and Maury Counties, Tennessee. Percentages are computed from concentrations in milliequivalents per liter.

Geochemical Processes

Based on lithologic descriptions and predominant ions in ground-water samples, the primary geochemical changes within both aquifer systems probably result from the dissolution or precipitation of calcite, dolomite, magnesium calcite, and gypsum. Chemical equations for these reactions are:

$$CaCO_3 + H_2CO_3 = Ca^{++} + 2HCO_3^- \quad (1)$$
(calcite)

$$CaMg(CO_3)_2 + 2H_2CO_3 = Ca^{++} + Mg^{++} + 4HCO_3^- \quad (2)$$
(dolomite)

$$Ca_{0.9}Mg_{0.1}CO_3 + H_2CO_3 = 0.9Ca^{++} + 0.1Mg^{++} + 2HCO_3^- \quad (3)$$
(magnesium calcite)

$$CaSO_4 * 2H_2O = Ca^{++} + SO_4^{--} + 2H_2O. \quad (4)$$
(gypsum)

Saturation indexes are indicators of whether a mineral is likely to precipitate or dissolve in a fluid such as ground water. The saturation index for a solid species is based on the chemical characteristics of the fluid, and is determined by comparing the actual ion activity product of the dissolved constituents of the solid species with its solubility product. A value less than zero indicates that the ground water is undersaturated with respect to the mineral and net dissolution of the mineral from solid phase should occur. If the value equals zero, the ground water is at equilibrium with respect to the mineral. When the value is greater than zero, the ground water is oversaturated with respect to the mineral, and net precipitation should occur (Matthess, 1982).

Saturation indexes of calcite, dolomite, and gypsum, the three principal soluble minerals in the aquifers, were determined using the computer code

WATEQF (Plummer and others, 1976). Most samples representing the Highland Rim aquifer system ranged from significantly undersaturated to nearly saturated with respect to calcite; one sample was slightly oversaturated (fig. 6). The water was significantly undersaturated to nearly saturated with respect to dolomite (fig. 6), and significantly undersaturated with respect to gypsum (fig. 7). Almost every sample from the Central Basin system was nearly saturated to slightly oversaturated with respect to calcite (fig. 6). Most samples were undersaturated with respect to dolomite, although three samples were slightly oversaturated (fig. 6). All samples were undersaturated with respect to gypsum (fig. 7). The plots of saturation indexes indicate that, overall, water in the Highland Rim aquifer system is less saturated with respect to these three minerals than water in the Central Basin aquifer system.

The two aquifer systems are similar in terms of primary geochemical reactions and the interactions of ground water with respect to calcite, dolomite, magnesium calcite, and gypsum. An evaluation of specific geochemical processes further supports some similarities observed and indicates that some differences exist between the two aquifer systems.

Similarities, in terms of carbonate dissolution, are indicated by the relation between dissolved calcium plus dissolved magnesium and total inorganic carbon. Least-square regression lines through the data points (fig. 8) show that approximately two moles of inorganic carbon are in water for every mole of dissolved calcium plus dissolved magnesium. Regression lines through data sets for each aquifer system have similar slopes (0.54 and 0.38 for the Highland Rim aquifer system and Central Basin aquifer system, respectively), indicating similar molar ratios in ground water from both aquifer systems. Total inorganic carbon (CO_2) in each ground-water sample was calculated from field alkalinity using part of the WATEQF code (Plummer and others, 1976). The two to one molar ratio between inorganic carbon and calcium plus magnesium is characteristic of the dissolution of calcite, dolomite, and magnesium calcite (equations 1, 2, and 3). Analyses for five sites were not included

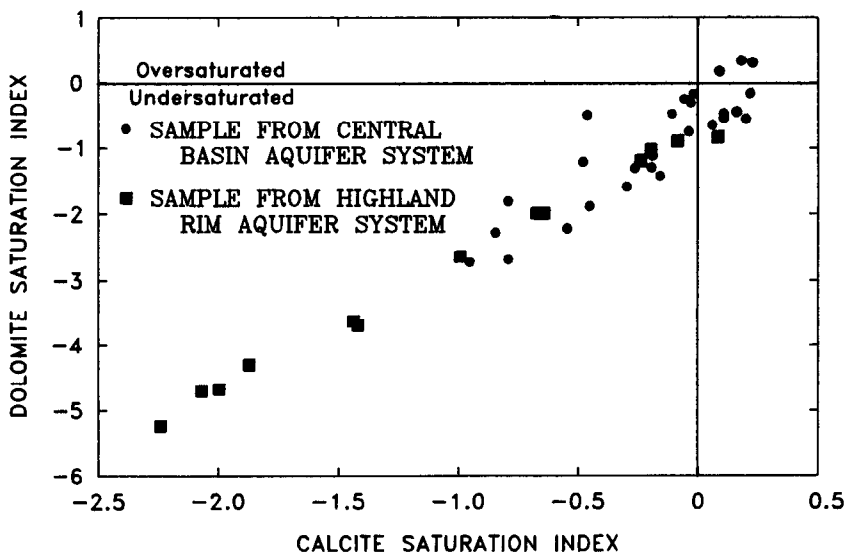


Figure 6. Calcite and dolomite saturation indexes of ground water from the Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee, calculated using the WATEQF program.

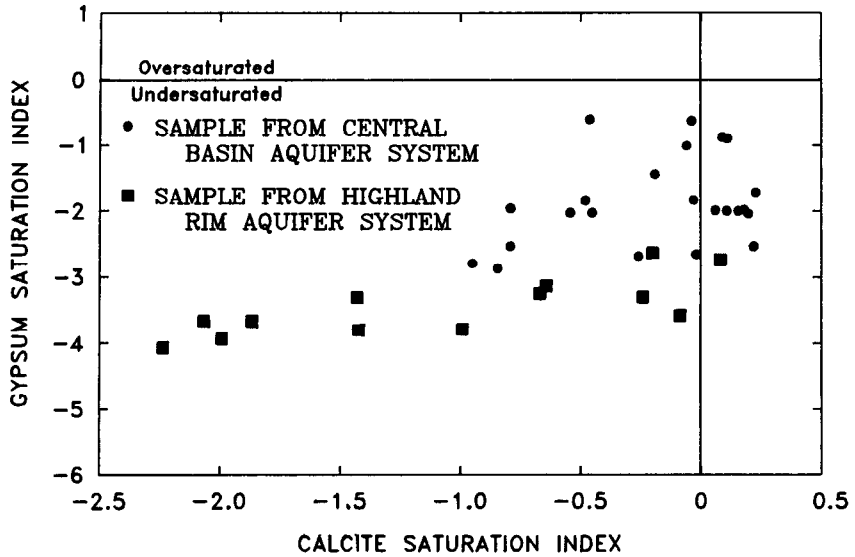


Figure 7. Calcite and gypsum saturation indexes of ground water from the Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee, calculated using the WATEQF program.

in the regression. These samples contained higher concentrations of dissolved calcium and dissolved sulfate, and might indicate areas where gypsum dissolution (equation 4) affects water composition.

Comparison of dissolved calcium to dissolved magnesium (fig. 9) indicates slightly different geochemical processes with respect to the dissolution of magnesium-bearing carbonate minerals present in each aquifer system. The regression through the data, calculated with values from the Highland Rim aquifer system only, has a correlation coefficient squared (R^2 value) of 0.87. This strong correlation, combined with thermodynamic calculations showing undersaturation of carbonates, suggests that dissolution of calcium- and magnesium-bearing carbonates from the Highland Rim aquifer system is congruent. In this process, calcite dissolves without simultaneous precipitation of calcite. Furthermore, the aquifer material that reacts with the water in the Highland Rim aquifer system has a fairly constant calcium to magnesium molar ratio of about 5.3 to 1.

Data for ground-water samples from the Central Basin aquifer system are more variable and do not plot along this same regression line. Possible explanations for the scatter of data from the Central Basin aquifer system are incongruent dissolution of calcium- and magnesium-bearing minerals or dissolution of aquifer material with variable calcium to magnesium ratios. Incongruent dissolution involves dissolution of a magnesium-rich solid phase such as dolomite (equation 2) or magnesium calcite (equation 3) followed by precipitation of a calcium-rich phase such as calcite (equation 1). Evidence for the likely precipitation of calcite is the nearly saturated to oversaturated state of calcite in most samples from the Central Basin aquifer system, as previously noted. Alternatively, variable calcium to magnesium ratios in the aquifer materials of the Central Basin aquifer system associated with changes in lithology could have a similar effect. For example, pure dolomite dissolving in water (equation 2) will result in a lower calcium to magnesium ratio than will pure calcite dissolving in water (equation 1).

Sulfate is the predominant anion in water from five sites; several processes can account for high sulfate concentrations. Dissolution of gypsum, a calcium sulfate evaporite mineral (equation 4), will increase the sulfate concentration

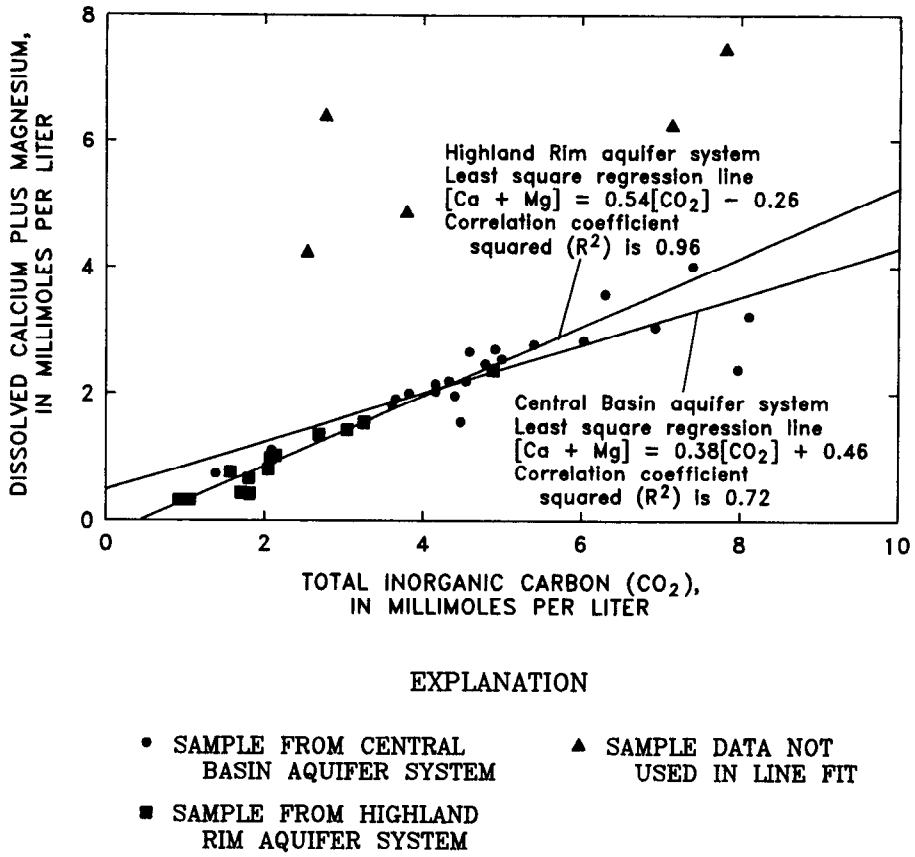


Figure 8. Relation between dissolved calcium plus dissolved magnesium and total inorganic carbon in ground water of the Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee.

in water. Pyrite oxidation also will contribute sulfate to solution, although in smaller amounts because oxygen availability often is limited in ground water. Alternatively, water containing high concentrations of dissolved sulfate might move upward from a deeper aquifer. Most of the high sulfate concentrations reported in this study probably are the result of gypsum dissolution because dissolved oxygen concentrations are low in most samples and because gypsum observed in rock outcrops confirms that the mineral is present.

RADIOACTIVITY IN GROUND WATER

The most common radionuclides in ground water are from the uranium-238 and the thorium-232 decay series (fig. 10). Uranium-238 is a naturally occurring, long-lived isotope (half life, 4.49×10^9 years) present in small concentrations in many rocks world-wide. This radionuclide slowly decays into a sequence of daughter products, ending in a stable isotope of lead. Among the daughters of uranium-238 are uranium-234 (half life, 2.48×10^5 years), radium-226 (half life, 1,622 years), and radon-222, a gas (half life, 3.825, days). Thorium-232 also is a naturally occurring, long-lived isotope (half life, 1.41×10^{10} years) present in many rocks. Among its daughters is radium-228 (half life, 5.75 years). Other radionuclides in the decay series of these two elements and all isotopes of the uranium-235 decay series are highly immobile or have short half lives, and thus are not present in significant amounts in ground water (Zapecza and Szabo, 1988).

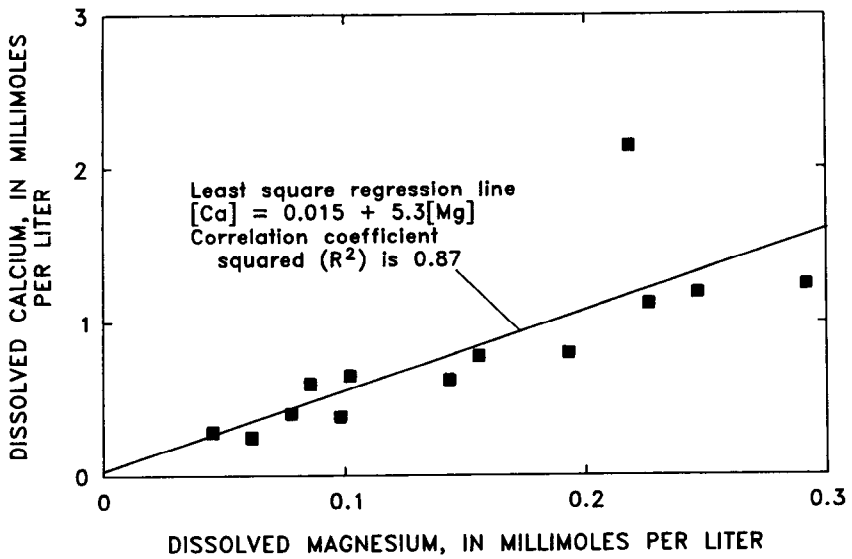


Figure 9. Relation between dissolved calcium and dissolved magnesium in ground water of the Highland Rim aquifer system, Hickman County, Tennessee.

Radiochemical Constituents

Samples of water for radiochemical analyses were collected concurrently from the same wells and springs as the samples for analyses of major constituents. Radon-222 concentrations in ground water were measured at each site using a Lucas cell technique. Gross alpha and gross beta activity, dissolved uranium, radium-226, and radium-228 concentrations were determined in the laboratory. Summary statistics of the analyses are given in table 3.

Differences in activity levels of constituents between aquifer systems generally were small (fig. 11). The median concentration of gross alpha activity in samples from each aquifer system was 0.54 pCi/L. The median concentration of gross beta activity in samples from the Highland Rim system was 1.1 pCi/L; and in samples from the Central Basin system, 2.3 pCi/L. Median concentrations of dissolved uranium (all isotopes) in water from the Highland Rim and Central Basin aquifer systems were 0.09 and 0.12 pCi/L, respectively; of radium-226, 0.09 and 0.07 pCi/L; and of radium-228, 0.04 pCi/L and 0.04 pCi/L. The largest difference was between median concentrations of radon-222. Median values were 559 pCi/L for the Highland Rim system and 422 pCi/L for the Central Basin system. The range in concentrations of each constituent was greater for samples from the Central Basin system, which might reflect the greater number of samples representing that system and the greater number of formations sampled. A Wilcoxon-Mann-Whitney rank sum test (Iman and Conover, 1983) was used to test for statistical differences between concentrations of radioactive constituents in water from each aquifer system. The test failed to reveal statistical differences at the 95-percent level of significance for any of the radioactive constituents.

None of the samples exceeded the radioactivity standards adopted by the Tennessee Department of Health and Environment (1991) for naturally occurring radioactivity in water used for community water supplies (a maximum of 15 pCi/L for gross alpha activity and 5 pCi/L for radium-226 plus radium-228 concentrations). Of the 40 samples collected, the largest gross alpha activity measured was 6.4 pCi/L, and the largest radium-226 plus radium-228 concentration was 2.7 pCi/L. Both maxima are identified with the same well, located in the Central Basin.

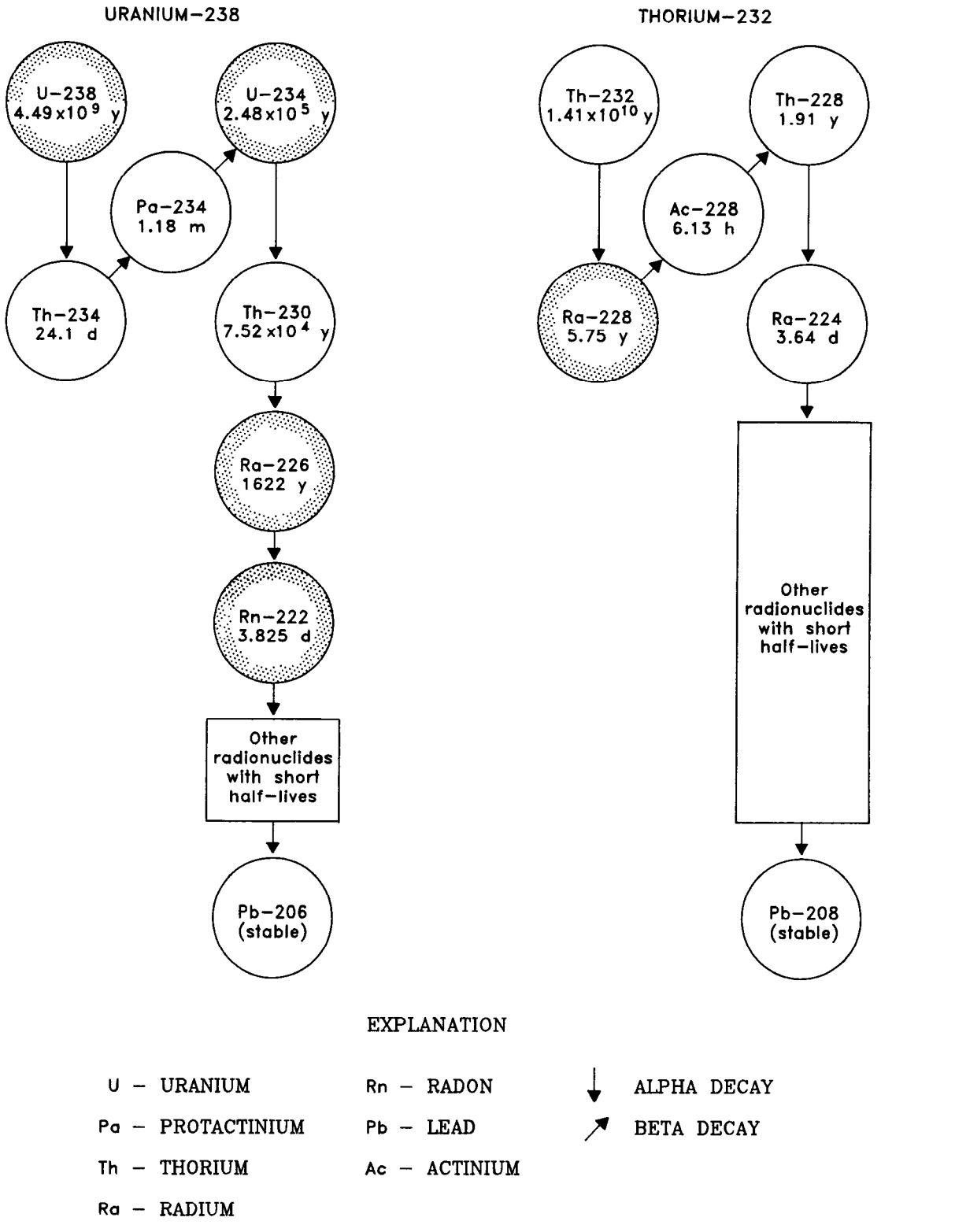


Figure 10. Uranium-238 and thorium-232 radioactive decay series. Stippling pattern indicates those radionuclides most frequently detected in ground water. Times shown are half-lives: y, years; d, days; h, hours; m, minutes. (*Modified from* O.S. Zapecza and Z. Szabo, 1988.)

Table 3. Summary statistics of selected radionuclides in ground water of the Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee

[All constituents are expressed in picocuries per liter; n, number of samples; *, value estimated by using a log-probability regression to predict the values of data below the detection limit; <, less than]

	Highland Rim aquifer system					Central Basin aquifer system				
Constituents	n	Maximum	Minimum	Mean	Median	n	Maximum	Minimum	Mean	Median
Gross alpha, dissolved, as natural uranium.	13	2.5	<0.4	0.74*	0.54*	27	6.4	<0.4	0.88*	0.54*
Gross beta, dissolved, as cesium-137.	13	5.7	<.6	1.6*	1.1*	27	10	<.6	2.9*	2.3*
Uranium, natural, dissolved.	13	1.44	.02	.21	.09	27	1.51	<.01	.20*	.12*
Radium-226, dissolved.	12	.73	.01	.16	.09	27	2.57	<.01	.25*	.07*
Radium-228, dissolved.	12	.08	.01	.04	.04	26	.46	<.01	.09*	.04*
Radon-222, total.	13	1,000	256	578.3	559	27	1,480	63	475.6	422

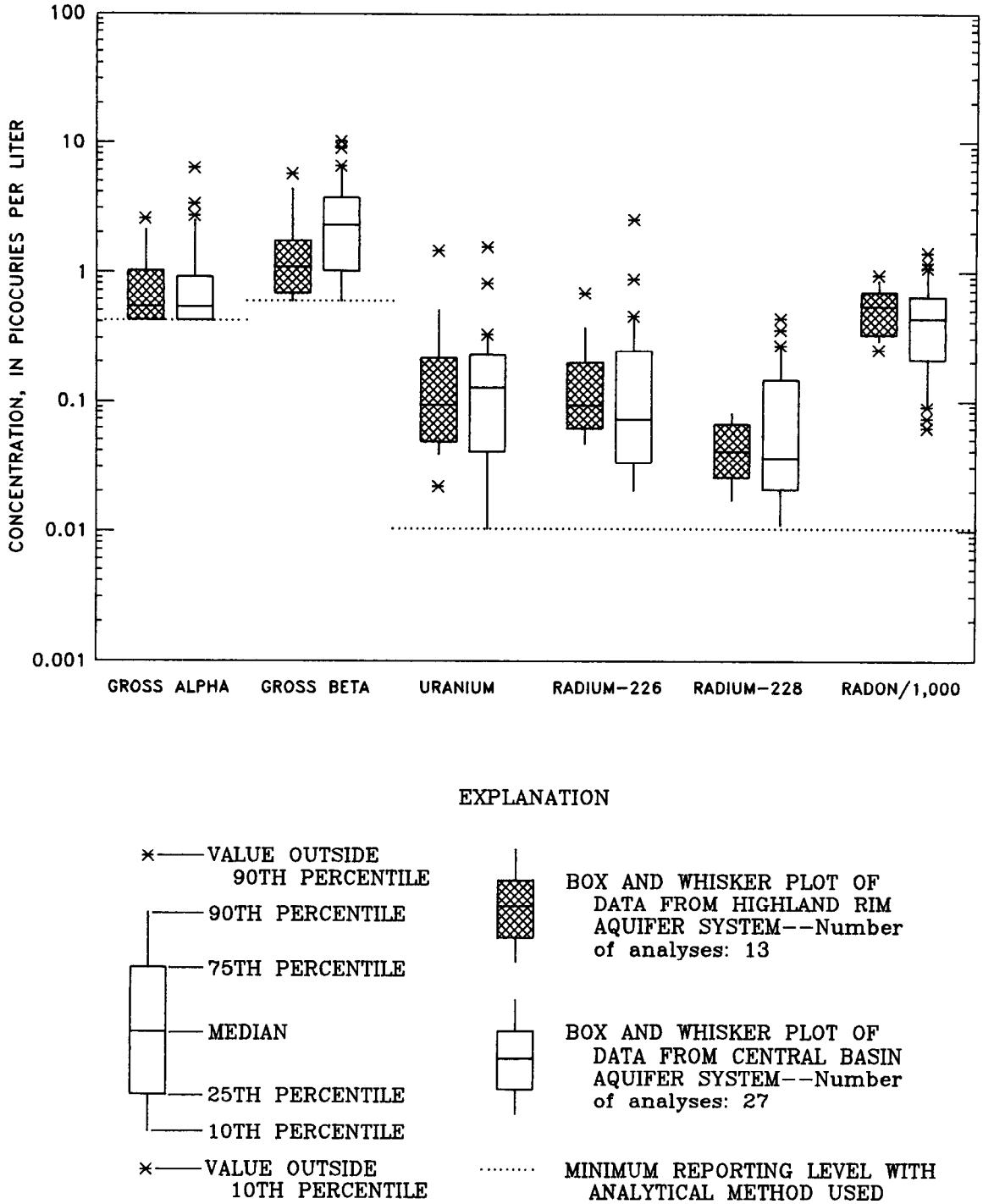


Figure 11. Selected radioactive constituents in ground water of the Highland Rim and Central Basin aquifer systems, Hickman and Maury Counties, Tennessee. Radon activities shown are 1/1,000 of actual measurements.

Concentrations in samples from most wells were substantially less than these maxima. Gross alpha activity was less than 2 pCi/L in about 90 percent of the samples from each aquifer system. The combined radium concentrations were less than 0.9 pCi/L in all samples from the Highland Rim aquifer system, and less than 0.9 pCi/L in all except two samples from the Central Basin aquifer system.

Neither the Tennessee Department of Health and Environment nor the U.S. Environmental Protection Agency has established a standard for radon in public drinking-water supplies. However, health physicists have proposed a 10,000 pCi/L limit for radon-222 (Cross and others, 1985). Concentrations of radon-222 in all samples collected for this study were substantially less than the proposed standard. Radon-222 in samples from the Highland Rim aquifer system ranged from 225 to 1,000 pCi/L, and in samples from the Central Basin aquifer system, 63 to 1,490 pCi/L.

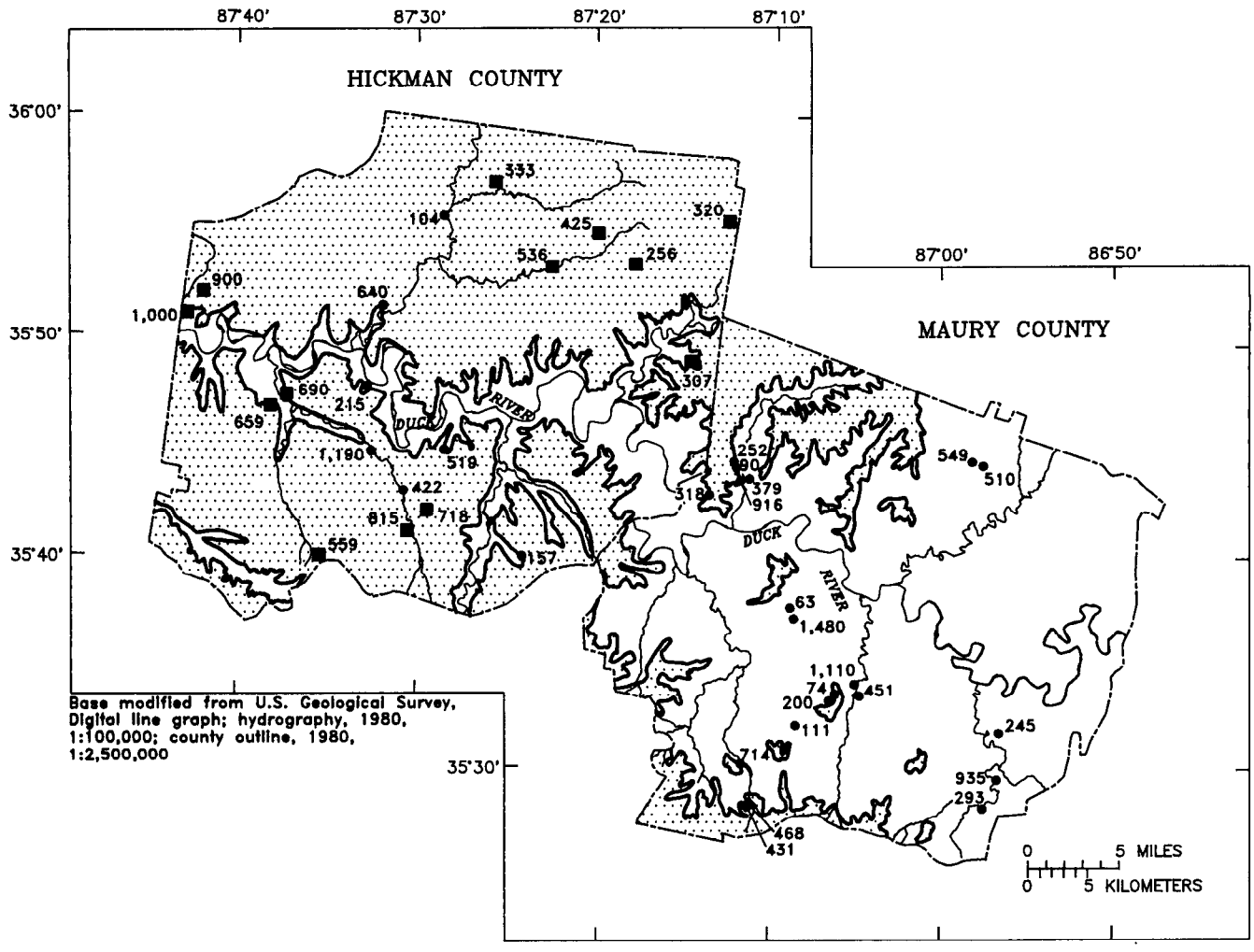
The geographic distribution of radon-222 in samples from the Highland Rim aquifer system suggests a trend. Six sites with concentrations less than the median value (559 pCi/L) are in northern Hickman County and seven sites with concentrations equal to or greater than the median value are in the southwestern two-thirds of the county (fig. 12). The data show no consistent pattern for the Central Basin aquifer system; however, maximum and minimum values are associated with wells less than one-half mile apart, and other large differences were observed in values between wells that were not far apart.

Relations Between Constituents

Correlation coefficients between radioactive constituents and key indicators of water quality were calculated to evaluate possible relations. The radioactive constituents were uranium, radium-226, radium-228, radon-222, gross alpha, and gross beta. Each of these constituents was paired individually, by aquifer system, with calcium, magnesium, sodium, barium, phosphorus, alkalinity, sulfate, dissolved oxygen, and pH. Although barium is usually considered a trace constituent, it was included because its chemical behavior is similar to that of radium. In addition, correlation coefficients were calculated by pairing each of the radionuclides, gross alpha, and gross beta with the other radionuclides of that aquifer system. For those data sets resulting in a correlation coefficient greater than 0.2, data were plotted to visually examine the correlation and evaluate the potential relation between the radionuclide and the constituent or property.

No relation was found in most of the data sets analyzed for this reconnaissance. Correlation coefficients generally were less than 0.2. A common pattern seen in plotted data (correlation coefficient greater than 0.2) was for most of the points to cluster in one part of the graph and one or two points to fall in outlying areas. The plot of uranium and calcium in water from the Central Basin aquifer system is typical (fig. 13), indicating that no relation exists between concentrations of radionuclides (dissolved uranium, radium-226, radium-228, and radon-222) in water and any of the key indicators of water chemistry in either aquifer system. One exception is radon-222 in water of the Highland Rim aquifer system, which is moderately related to pH (R^2 of 0.71) and weakly related to dissolved magnesium (R^2 of 0.51).

The relation of radon-222 concentration to pH might be indicative of hydrogeochemical environments in the lower part of the Highland Rim aquifer system. A graph of the relation between radon-222 concentrations and pH shows that they are inversely related (fig. 14). The six samples having radon-222 concentrations greater than 600 pCi/L are from water associated with the Chattanooga Shale; two of the samples are from contact springs at the top of this formation. The comparatively low pH of these samples indicates that water in this part of the aquifer has not substantially interacted with carbonate rock and, that in the area represented by these samples, the lower part of the aquifer probably contains little carbonate material. A possible explanation is that the carbonate bedrock in this area has been reduced to regolith to at least the bottom of the Fort Payne Formation, readily permitting radon-222 from the decay of uranium in the Chattanooga Shale to enter ground water in the regolith immediately above the shale. In areas characterized by lower radon levels and higher pH values, relatively unweathered bedrock overlies the Chattanooga Shale and prevents



EXPLANATION

OUTCROP AREA OF THE HIGHLAND RIM AQUIFER SYSTEM

OUTCROP AREA OF THE CENTRAL BASIN AQUIFER SYSTEM

559 WELL OPEN TO THE HIGHLAND RIM AQUIFER SYSTEM--Number indicates concentration of radon-222 in ground water, in picocuries per liter

157 WELL OPEN TO THE CENTRAL BASIN AQUIFER SYSTEM--Number indicates concentration of radon-222 in ground water, in picocuries per liter

Figure 12. Distribution of radon-222 in ground water of the study area.

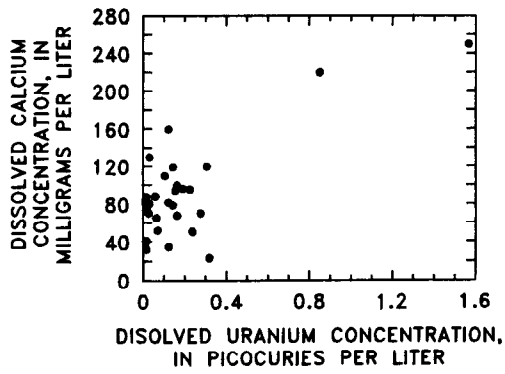


Figure 13. Relation between dissolved calcium and dissolved uranium in ground-water samples from the Central Basin aquifer system, Hickman and Maury Counties, Tennessee.

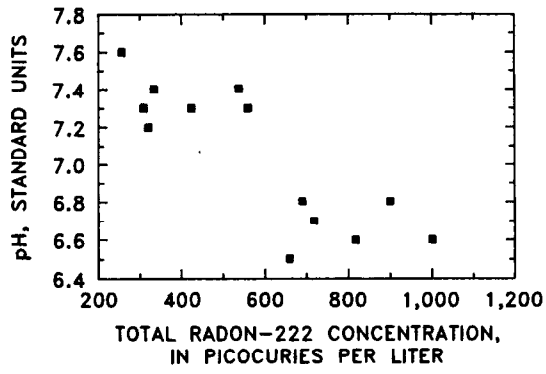


Figure 14. Relation between pH and total radon-222 in ground-water samples from the Highland Rim aquifer system, Hickman County, Tennessee.

the entry of other than minor amounts of gas into the aquifer above the shale. The presence of carbonate bedrock in the aquifer is expressed geochemically by the comparatively higher pH values of ground water that has come in contact with the rock.

The association of uranium with phosphatic rock was previously noted. However, the data did not indicate any relation between uranium and phosphorus concentrations in water from either aquifer system (figs. 15 and 16).

None of the isotopes in water from either aquifer system were related to other isotopes or to gross alpha or gross beta activities, with the exception of radium-226. Radium-226 concentrations were weakly related to gross alpha activity in water from the Highland Rim aquifer system (R^2 of 0.48) and moderately related to gross alpha activity in water from the Central Basin aquifer system (R^2 of 0.88). The latter value might be misleading, however, owing to outlying points in the data set which tend to make the correlation seem stronger than the distribution of points otherwise implies.

SUMMARY AND CONCLUSIONS

A reconnaissance of the geochemistry of and radioactivity in ground water from the Highland Rim and Central Basin aquifer systems in Hickman Maury Counties, Tennessee, was conducted in 1989 by the U.S. Geological Survey in cooperation with the Tennessee Department of Health and Environment. For the study, chemical and radiochemical analyses were made of ground-water samples from 13 sites tapping the Highland Rim aquifer system and 27 sites tapping the Central Basin aquifer system. The results of the analyses were summarized and the geochemical environments characterized by calculating saturation indexes of key minerals and discussing major geochemical processes indicated by the relations of constituents in ground-water samples. Relations between radiochemical constituents and key indicators of water chemistry and relations among radiochemical constituents also were determined.

Most of the measures of water chemistry indicate moderate differences in geochemistry between the two aquifer systems. Median values for the specific conductance of ground-water samples from the Highland Rim and Central Basin aquifer systems were 150 and 500 μS/cm, respectively; for dissolved solids, 101 and 271 mg/L; and for pH, 7.2 and 7.2 standard units. Concentrations of many ion ground-water samples typically were lower in the Highland Rim than in the Central Basin aquifer system. Comparisons of water quality between the two aquifer systems indicated

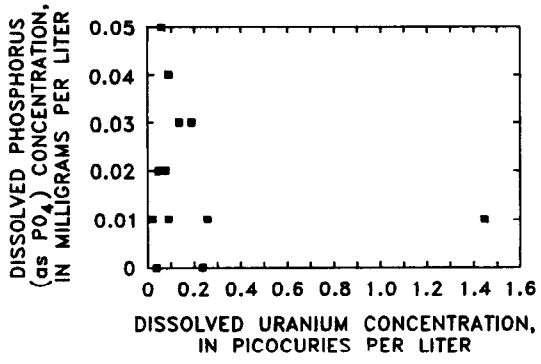


Figure 15. Relation between dissolved phosphorus and dissolved uranium in ground-water samples from the Highland Rim aquifer system, Hickman County, Tennessee.

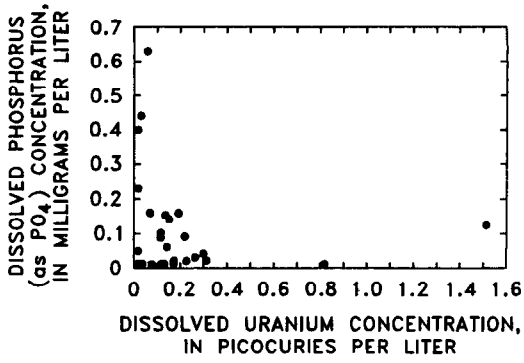


Figure 16. Relation between dissolved phosphorus and dissolved uranium in ground-water samples from the Central Basin aquifer system, Hickman and Maury Counties, Tennessee.

statistically significant differences among the major dissolved ions concentrations of calcium, magnesium, sodium, potassium, chloride, and sulfate. Despite differences between aquifer systems in major ion concentrations, the types of water in both systems are similar. Calcium and bicarbonate were the predominant ions in all 13 samples from the Highland Rim aquifer system. Calcium or calcium and magnesium were the predominant cations in 25 of the 27 Central Basin aquifer system samples and bicarbonate was the major anion in 21 of 27 samples.

Dissolution of calcite, magnesium calcite, dolomite, and gypsum are the primary geochemical processes controlling ground-water chemistry in both aquifer systems. For the Highland Rim aquifer system, saturation state calculations using chemical analyses of ground-water samples and the computer code WATEQF indicate that, with respect to calcite saturation, most of the ground-water samples ranged from significantly undersaturated to nearly saturated, except one sample that was slightly oversaturated; with respect to dolomite saturation, samples were significantly undersaturated to nearly saturated; and with respect to gypsum, all samples were significantly undersaturated. For the Central Basin aquifer system, saturation state calculations indicate that with respect to calcite, almost every ground-water sample was nearly saturated to slightly oversaturated; with respect to dolomite, all samples were undersaturated to nearly saturated except three samples that were oversaturated; and with respect to gypsum, all samples were undersaturated. Overall, ground-water samples from the Central Basin aquifer system were more saturated with respect to calcite, dolomite, and gypsum than samples from the Highland Rim aquifer system.

The relation of dissolved calcium to dissolved magnesium and the saturation state calculations indicate that slightly different geochemical environments exist in each aquifer system with respect to the dissolution of magnesium-bearing minerals. Ground-water samples from the Highland Rim aquifer system had a fairly consistent calcium to magnesium molar ratio of about 5.3 to 1, which suggests dissolution of aquifer material with a consistent calcium to magnesium ratio. The highly variable calcium to magnesium molar ratios in ground-water samples from the Central Basin aquifer system might indicate incongruent dissolution of magnesium-bearing minerals or heterogeneity in magnesium content of the aquifer.

Concentrations of radiochemical constituents were low and generally similar in ground-water samples from both aquifer systems. For the Highland Rim aquifer system, the median activity

in ground-water samples for gross alpha was 0.54 pCi/L, and for gross beta, 1.1 pCi/L. The median concentration of dissolved uranium (all isotopes) was 0.09 pCi/L; radium-226, 0.09 pCi/L; radium-228, 0.04 pCi/L; and radon-222, 559 pCi/L. For the Central Basin aquifer system, the median activity of gross alpha was 0.54 pCi/L, and of gross beta, 2.3 pCi/L. The median concentration of dissolved uranium was 0.12 pCi/L;, radium-226, 0.07 pCi/L; radium-228, 0.04 pCi/L; and radon-222, 422 pCi/L. Radium-226 plus radium-228 concentrations were less than 0.9 pCi/L in all ground-water samples from the Highland Rim aquifer system and less than 0.9 pCi/L in all except two samples from the Central Basin aquifer system. Concentrations of radium-226 plus radium-228 in all samples from both aquifer systems were less than Tennessee's maximum permissible contaminant level of 5 pCi/L for radium isotopes in community water-supply systems.

Data collected for this reconnaissance generally did not indicate any relations involving radiochemical constituents in ground-water samples. For either aquifer system, no relation exists between concentrations of the radionuclides (uranium, radium-226, radium-228, and radon-222) dissolved in water and any of the key indicators of water chemistry, with the exception of water from the Highland Rim aquifer system, for which a moderate relation (R^2 of 0.71) was indicated between radon-222 and pH, and a weak relation (R^2 of 0.51) was indicated between radon-222 and dissolved magnesium. The relation of radon-222 activity to pH might be indicative of hydrogeochemical environments of the Highland Rim aquifer system where carbonates are absent and radon-222 can easily enter ground water. None of the radionuclide concentrations in water from either aquifer system correlated to other radionuclides or to gross alpha or gross beta activities, with the exception of radium-226. Radium-226 correlated to gross alpha activity in water from both the Highland Rim and Central Basin aquifer systems (R^2 of 0.48 and 0.88, respectively).

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*USGPO 1993-750-232/80008