

# U.S. Geological Survey Research in Radioactive Waste Disposal - Fiscal Years 1986 - 1990

Compiled by N. J. Trask and P. R. Stevens

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## CONVERSION FACTORS

For use of readers who prefer to use Inch-Pound equivalent units, conversion factors for terms used in this report are listed below:

<u>Multiply metric unit</u>	<u>By</u>	<u>To Obtain inch-pound unit</u>
meter(m)	3.281	foot(ft)
square meter(m <sup>2</sup> )	10.76	square foot(ft <sup>2</sup> )
megaPascal (MPa)	10	bar
liter(l)	1.057	quart (qt)
gram(g)	0.035	ounce(oz)
becquerel (Bq)	0.027	curie(Ci)
cubic meter(m <sup>3</sup> )	35.31	cubic foot(ft <sup>3</sup> )

## SI PREFIXES

Factor by which unit is multiplied	Prefix	Symbol
10 <sup>3</sup>	kilo	k
10 <sup>2</sup>	hecto	h
10	deka	da
10 <sup>-1</sup>	deci	d
10 <sup>-2</sup>	centi	c
10 <sup>-3</sup>	milli	m
10 <sup>-6</sup>	micro	u
10 <sup>-9</sup>	nano	n
10 <sup>-12</sup>	pico	p

Temperatures in degrees Celsius (°C.) can be converted to degrees Fahrenheit (°F.) as follows:

$$^{\circ}\text{F} = 1.8^{\circ}\text{C} + 32$$

## ABBREVIATIONS

Ma, million years before present  
 m.y., million years  
 M, molality

U. S. Geological Survey Research in Radioactive  
Waste Disposal - Fiscal Years 1986-1990

Compiled by

N. J. Trask and P. R. Stevens

ABSTRACT

The report summarizes progress on geologic and hydrologic research related to the disposal of radioactive wastes. The research efforts are categorized according to whether they are related most directly to: (1) high-level wastes, (2) transuranic wastes, (3) low-level and mixed low-level and hazardous wastes, or (4) uranium mill tailings. Included is research applicable to the identification and geohydrologic characterization of waste-disposal sites, to investigations of specific sites where wastes have been stored, to development of techniques and methods for characterizing disposal sites, and to studies of geologic and hydrologic processes related to the transport and/or retention of waste radionuclides.

INTRODUCTION

The United States Geological Survey (USGS) has since the 1950's conducted research on the disposal of radioactive wastes. Projects have included work funded by other agencies including the U. S. Department of Energy (DOE) as well as research done with directly appropriated funds (USGS Surveys, Investigations, and Research). This document summarizes the research carried out during fiscal years 1986-1990.

The research of the USGS as a whole includes almost every aspect of the earth sciences. The specific research activities described here are part of the USGS program that is directly related to radioactive waste disposal. They include geologic and hydrologic research of general applicability to the identification and characterization of waste disposal sites, investigations of specific sites where wastes have been stored, and studies of processes likely to be important in evaluating the performance of specific sites. This research is discussed under the headings of:

- a) high-level radioactive waste,
- b) transuranic waste,
- c) low-level radioactive and mixed low-level radioactive and hazardous wastes, and
- d) uranium mill tailings.

Many of the results have application to more than one category of waste.

High-level radioactive wastes include fission products that initially have a high level of beta and gamma radiation and a high rate of heat generation; they also include transuranic elements with long half lives. Transuranic waste contains long-lived alpha emitters at concentrations greater than 100 nCi/g and generate little or no heat.

The term low-level waste has carried a changing and imprecise definition over the years. Currently, it generally means waste which does not fit the definition of high-level waste and in which the concentration of transuranic elements is less than 100 nCi/g. It consists in part of miscellaneous solid materials that have been irradiated and contaminated through use, as well as products of reactors and fuel reprocessing plants. Some low-level wastes are mixed with materials that are on the list of hazardous materials maintained by the Environmental Protection Agency and are thus subject to relevant environmental laws for such substances.

This is the sixth report of progress of USGS research in radioactive waste disposal; the previous review covered fiscal years 1983-1985 (Dinwiddie and Trask, 1986).

Reference:

Dinwiddie, G. A. and Trask, N. J., 1986, U. S. Geological Survey Research in Radioactive Waste Disposal - Fiscal Years 1983, 1984, and 1985: U. S. Geological Survey Water-Resources Investigations Report 87-4009.

HIGH-LEVEL RADIOACTIVE WASTE

Yucca Mountain Site, Nevada and Vicinity

The USGS has been assisting the DOE in evaluating the suitability of the Yucca Mountain area, located on and adjacent to the Nevada Test Site and about 160 km northwest of Las Vegas, as a potential repository site for the disposal of commercially generated high-level radioactive wastes and high-level radioactive wastes from DOE defense facilities. An essential part of this work is defining the geology and hydrology of the area in order to see if the site meets the guidelines for suitability outlined by the DOE in 10 CFR Part 960, Code of Federal Regulations and the criteria for licensing of a site outlined by regulations of the Nuclear Regulatory Commission (NRC) (10 CFR Part 60, Code of Federal Regulations).

In 1986, under provisions of the Nuclear Waste Policy Act of 1982 (Public Law 97-425), the Yucca Mountain site was chosen as one of 3 sites to be characterized for the nation's first high-level radioactive waste repository. In late 1987, the U. S. Congress passed the Nuclear Waste Policy Amendments Act (Title V of Public Law 100-203), which designated Yucca Mountain as the only site to be characterized. The Yucca Mountain site has not been selected for a repository; it is at this time the only site designated for study to assess its suitability to contain a repository.

The USGS contributed to the geologic and hydrologic sections of the Site Characterization Plan (SCP) (DOE, 1988a), which describes the current understanding of the characteristics and features of the site, presents a preliminary conceptual design of the repository and waste package, and describes, in general, the tests and analyses that the DOE is conducting during site characterization and the rationale used to select those tests and analyses. The studies described in the SCP are designed to provide the information necessary to determine whether the Yucca Mountain site is suitable for a geologic repository and, if so, to provide the information necessary to prepare a license application for submittal to the NRC. The SCP summarizes work by the USGS and other participants in the project at the Yucca Mountain site completed prior to 1986 and includes references through 1987.

Semiannual progress reports on the course of site characterization have been prepared for the period starting with April, 1989 (DOE, 1990a,b, 1991). These contain summaries of USGS investigations and recent references. In addition, DOE publishes a Yucca Mountain Project bibliography (DOE, 1988b; 1990c) which is updated at six-month intervals (DOE, 1991b,c) and lists significant USGS reports on the work.

### References

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- DOE (U.S. Department of Energy), Office of Scientific and Technical Information, 1988b, Nevada Nuclear Waste Storage Investigations Bibliography, 1986-1987: DOE/OSTI-3406 (Suppl.1), Oak Ridge, Tenn.
- DOE (U.S. Department of Energy), Yucca Mountain Project Office, 1990a, Yucca Mountain Project Technical Status Report, April-September 1989, NVO-334-1, Las Vegas, Nevada, 100 p.

- DOE (U.S. Department of Energy), Yucca Mountain Project Office, 1990b, Yucca Mountain Project Technical Status Report, October 1989-March 1990, NVO-334-2, Las Vegas, Nevada, 110 p.
- DOE (U.S. Department of Energy), Office of Civilian Radioactive Waste Management, 1991a, Site Characterization Progress Report: Yucca Mountain, Nevada, April 1, 1990-September 30, 1990, DOE/RW-0307P, Washington, D. C., 147 p.
- DOE (U.S. Department of Energy), Office of Scientific and Technical Information, 1990c, Yucca Mountain Project Bibliography, 1988-1989: DOE/OSTI-3406 (Suppl.2), Oak Ridge, Tenn., 177 p.
- DOE (U.S. Department of Energy), Office of Scientific and Technical Information, 1991b, Yucca Mountain Project Bibliography, January-June 1990: An update, DOE/OSTI-3406 (Suppl.2) (Add.1), Oak Ridge, Tenn., 27 p.
- DOE (U.S. Department of Energy), Office of Scientific and Technical Information, 1991c, Yucca Mountain Site Characterization Project Bibliography, July-December 1990: An update, DOE/OSTI-3406 (Suppl.2) (Add.2), Oak Ridge, Tenn., 38 p.

#### CODES AND REGULATIONS

- 10 CFR Part 60 (Code of Federal Regulations), 1987. Title 10, "Energy," Part 60, "Disposal of High-Level Radioactive Waste in Geologic Repositories," U.S. Government Printing Office, Washington, D.C., pp. 627-658.
- 10 CFR Part 960 (Code of Federal Regulations), 1987. Title 10, "Energy," Part 960, "General Guidelines for the Recommendation of Sites for Nuclear Waste Repositories," U.S. Government Printing Office, Washington, D.C., pp. 518-551.

#### Regional Studies Related to the Yucca Mountain Site

The USGS has sponsored a series of studies dealing with regional aspects of the paleoclimate, paleohydrology, and structural geology of the region surrounding the Yucca Mountain site. The results of these studies serve to describe the framework within which the Yucca Mountain site lies and also have relevance to problems of the hydrologic and geologic history of the southern Great Basin.

These studies have been conducted under the normal quality controls of the USGS and not under the more detailed quality assurance program under which the DOE program is conducted. Portions of the studies could be repeated under more detailed quality assurance practices should this be deemed necessary at a future date.

## Paleoclimatology and Paleohydrology of the Southern Great Basin

By Isaac J. Winograd, Reston, VA

An evaluation of the ability of the thick unsaturated zones of the Great Basin to isolate solidified toxic wastes from the hydrosphere for tens of millennia requires knowledge of the paleohydrology and paleoclimatology of this region. Specifically needed are estimates of regional climate and water-table altitude during the glacial and interglacial climates of the Pleistocene. Such estimates can then be used as guides for "bounding" estimates of future fluctuations of these parameters.

Objectives: a) To develop a detailed and well-dated record of oxygen-18 and carbon-13 fluctuations in ground-water recharge during the late Pliocene and Pleistocene Epochs; b) to compare this isotopic record -- a proxy of temperature change -- with marine and Antarctic oxygen-18 records and sea-level records in order to test the Milankovitch Theory of climate change; c) to determine fluctuations of regional water table with climate, an item of major importance to use of thick unsaturated zones for disposal of solidified toxic wastes; and d) to determine the time of isolation of the pupfish *Cyprinodon diabolis* in Devils Hole, Ash Meadows, Nevada. Knowledge of this isolation time is of major interest to zoologists studying rates of evolution.

Approach: The isotopic record is being developed through uranium-series disequilibrium dating of dense calcitic veins that mark the routes of paleo-ground water flow along fissures. Dated veins are then analyzed millimeter by millimeter for variations of oxygen-18 and carbon-13 in the calcite. Paleo-water levels are being determined by the mapping and dating of a variety of geologic features including: tufa, calcitic veins, and speleothem.

Progress: A 500,000 year record of climate change in the Great Basin was developed from variations in the oxygen-18 and carbon-13 of uranium-series dated vein calcite collected by scuba drilling in Devils Hole, Nevada. The oxygen-18 variations reflect variations in cool-season temperature, while the carbon-13 variations reflect changes in vegetative density in the recharge areas. The configuration of the

oxygen-18 curve very closely resembles the marine and Antarctic oxygen-18 curves, reinforcing the belief (Winograd and others, 1988) that the Devils Hole vein calcite records a global climate signal. Four complete glacial-interglacial cycles are recorded. A comparison of the timing of these cycles with retrodictions of solar insolation calls into question the validity of the Milankovitch theory of climate change in the time domain, and in turn the marine record which is "tuned" to the insolation cycles. The Devils Hole climatic record -- now dated with the precise mass-spectrometric method -- is likely to serve as a standard against which other paleoclimatic records will be recorded. Our next step will be to extend this record back to one million years.

Because the calcite veins mark the sites of discharge from paleo-springs, dating of such veins in the central Amargosa Desert has also enabled us to reconstruct the approximate altitude of the regional water table in the past one million years. A preliminary study (Winograd and Szabo, 1988) indicated that the regional water table in the Amargosa Desert has been declining during the Quaternary Period due to a combination of increasing aridity (caused by uplift of the Sierra Nevada and Transverse Ranges) and by tectonic lowering of ground-water base level in Death Valley.

The cited work, and work in progress, indicate that this decline was at least 70 m. Superimposed on this general decline were small (<10 m) rises in water table in response to wetter, full-glacial climates.

### Reports

Winograd, I. J., Szabo, B. J., Coplen, T. B., and Riggs, A. C., 1988, A 250,000-year climatic record from Great Basin vein calcite: Implications for Milankovitch theory: *Science*, v. 242, p. 1275-1280 (Discussion in v. 246, p. 262-263).

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of America, Abstracts with Programs, v. 22, no. 7, p. 209.

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Ludwig, K. R., Simmons, K. R., Szabo, B. J., Riggs, A. C., and Winograd, I. J., 1990, Mass-spectrometric  $^{230}\text{Th}$ - $^{234}\text{U}$ - $^{238}\text{U}$  dating of the Devils Hole calcite vein: A precise record of continuous growth from ~566 Ka to 60 Ka: Geological Society of America, Abstracts with Programs, v. 22, no. 7, p. 310.

## Paleohydrology of the Great Basin

by Larry Benson and Steve Hostetler, Boulder, Colorado

The possible siting of waste disposal facilities in the Great Basin creates a need to understand past climatic fluctuations and their effect on the hydrology of the region. Past weather patterns associated with extremes in glacial and solar insolation boundary conditions may have been substantially different from present patterns.

Objectives: To document changes in the hydrologic cycle that have occurred in the Great Basin over the last few hundred thousand years. A secondary objective is to link changes in the hydrologic balance to changes in climate through the use of physically based models of the hydrologic balance.

Approach: Map the geology of lake deposits laid down over the last few hundred years. Use stable isotope-studies of dated sediments and waters to infer past conditions.

Progress: Since 1987 we have demonstrated the following:

- (1) Both Lake Bonneville (Great Salt Lake) and Lake Lahontan (Pyramid Lake) achieved their last highstands ~13,500 yr B.P. At this time, their surface areas were 8 to 10 times larger than today.

(2) Between 13,500 and 12,000 yr B.P., lakes in both basins fell. Between 12,000 and ~10,500 yr B.P., lake levels stabilized or rose slightly. Surface areas in the latter time period were ~2 to 3 times larger than today.

(3) Between 18,500 and 9,000 yr B.P. a substantial amount of ground-water recharge occurred in southern Nevada. Stable-isotope values for these waters indicate an increase in condensation air temperature of ~5° C. during this time period and that westerly storm systems were responsible for ground-water recharge.

Ongoing research on Lakes Estancia and San Agustin by Roger Anderson (University of New Mexico) and Fred Phillips (New Mexico Institute of Mining and Technology) indicate that highstands in these lakes occurred between ~20,000 and 18,000 yr B.P. with subsequent rises in lake level between ~14,000 and 12,000 yr B.P. We have used the foregoing data together with atmospheric global-climate modelling experiments of John Kurzbaach (University of Wisconsin) to suggest that the size and shape of the continental ice sheet governs the position of the polar jetstream. When the ice sheet is at its maximum extent, the jetstream splits and the southern branch remains over the southwestern United States all year bringing cooler air temperatures and more cloudiness and precipitation. As the ice sheet shrinks, the mean position of the southern branch migrates northward over the Great Basin.

The jetstream/ice-sheet linkage does not necessarily explain the stabilization in lake levels that occurred between 12,000 and 10,500 yr B.P. During this time, change in the circulation of the North Atlantic was responsible for two European climatic events, the Allerod and the Younger Dryas. We have hypothesized that these periods of climatic change were at least hemispheric in scope and were responsible for climatic change in the western United States. Importing of a jetstream climatology (7 to 10° C. reduction in air temperature and 270 days of cloudiness) in a lake thermal/evaporation model indicates that Lake Lahontan could have achieved its last highstand with only 2.6 times modern discharge. Preliminary experiments with the MM4 mesoscale model of the National Center for Atmospheric Research by Filippo Giorgi indicate that the large surface areas of Lakes Bonneville and Lahontan induced lake-effect storms during at least the winter season. These storms may have been responsible for most of the additional discharge entering the lakes during the last highstand.

There are three consequences of this work:

(1) The mesoscale experiments explain the observed differences in lake-surface areas between the Bonneville and Lahontan lake systems and the Russell and Owens Valley

lake systems; i.e., the two latter systems were never large enough to induce lake-effect storms.

(2) While it was wet and cool over and downstream of the large lake systems, it may have been cloudy and cool elsewhere in the Great Basin.

(3) If the hypothesized linkage of ice-sheet size and ground-water recharge is correct, then substantial amounts of ground-water recharge occur infrequently in the Great Basin and may not occur again for tens of thousands of years.

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# Studies of Processes Related to the Yucca Mountain Site

## Geochemical Kinetics

by Hans C. Claassen, Lakewood, CO

The interaction of the solid and liquid phases in natural ground- and surface-water systems results in a water quality which is a function of the chemical characteristics of rock and water, the temperature, the effective surface area of rock in contact with the water, and the interaction time. Differences in lithology and climatic conditions result in a range of water qualities that is surprisingly narrow, when the broad range of variables is considered. This would suggest that chemical controls are operant, but it is not presently known whether they are thermodynamic (equilibrium) or kinetic (rate process).

Objective: Presently, the project objectives focus on three detailed studies relating specific aspects of water quality to hydroclimatic variables:

(1) Chloride-ion mass balance. It has long been accepted that chloride-ion concentration in ground- and surface-water is an indication of effective or available moisture in the watershed. However, extension of this relation to a quantitative water balance requires detailed functional relations of inputs and transformations affecting chloride-ion concentrations in the watershed. The precise model describing these processes depends on the climate extant when the water was recharged. In watersheds where recharge is frequent and substantial, a different model will be appropriate than in more arid watersheds where recharge is infrequent or absent. Detailed studies are continuing in two watersheds in the Colorado mountains to define the correct model for the former situation. A project at Organ Pipe Cactus National Monument, Arizona is underway to develop a model for more arid conditions.

(2) Water stable-isotope mass balance. The geochemical literature reflects significant effort to use water-isotope evidence in describing hydrologic and climatic regimes. In many instances, no mass balance was considered, leading to questionable models. In other instances, some of the known processes affecting water-isotope composition were either not considered or were not quantified, resulting in an inaccurate or incomplete mass balance. Studies taking place in the watersheds indicated in (1) above are intended to eliminate or minimize these shortcomings.

(3) Major-ion solute mass balance. This study will include determining the influence of environmental factors, such as

precipitation quantity, quality and time distribution, vegetative cover, and temperature, as well as the role of the soil zone on water quality. The influence on water chemistry of changes resulting from chemical precipitation of authigenic phases will be quantitatively addressed.

The above specified detailed studies are intended to provide models that can be used to discern information about paleoclimates contained in paleowater chemistry. This paleoclimate data may, in turn, be used to estimate the future behavior of waste repositories.

Approach: As indicated, a mass-balance approach to inputs, outputs, and intermediate processes will be used. The methods and techniques will depend on the hydrologic regime, but, in general, atmospheric inputs (wetfall, dryfall) will be quantitatively measured or estimated using micrometeorological techniques. Changes occurring during interception by vegetation or storage at ground surface will be determined. Losses and chemical modification of soil moisture will be measured, as will changes resulting from dissolution/chemical precipitation reactions during ground-water recharge.

Progress: A continuing, detailed mass balance of aqueous chloride ion for the dacite-andesite tuff watershed in the Colorado mountains reinforces the utility of that ion in partitioning precipitation input among its various hydrologic components: evaporation, recharge and runoff. The ability of the model to provide accurate partitioning indicates that the correct choice of controlling variables has been made and directs that the model be applied to other watersheds in similar environments. To achieve this, a computer code has been written and verified for the model watershed. A literature search has located other watersheds in the American West having an independent data base that resolves the hydrologic balance into its components. The chloride-ion model is being applied to these watersheds and the results compared to results reported in the literature.

Water-isotope measurements in the aforementioned watershed continue and reveal a small isotopic shift in the composition of precipitation when it is intercepted by vegetation. The degree of isotopic modification depends on the length of time the precipitation is in the intercepted state; thus, only snow is significantly modified and only under conditions when atmospheric temperatures remain well below freezing. A computer code is being written to quantitatively describe this process.

Although summer precipitation is generally less than potential evaporation, time distribution results in small amounts of ground-water recharge occurring in some years.

The importance of summer precipitation lies primarily in its effect on the isotopic composition of snowmelt recharge, as significant amounts of isotopically-modified soil moisture are incorporated during the spring recharge process. This process is being modeled and its impact on the climatic interpretation of ground-water isotope signals evaluated.

Studies at Organ Pipe Cactus National Monument, an arid site, are just beginning, but consultation with cooperating experts at the University of New Mexico, New Mexico Institute of Mining and Technology, and Utah State University indicate that the site was well chosen for the purpose discussed above. Instrumentation of the site is proceeding.

#### Reports:

Claassen, H. C., Reddy, M. M., and Halm, D. R., 1986, Use of the chloride-ion in determining hydrologic-basin water budgets - A 3-year case study in the San Juan Mountains, Colorado, U.S.A., Journal of Hydrology, v.85, no. 1-2, p.49-72.

Claassen, H. C., Sublimation of evergreen-intercepted snow and its effect on hydrogen and oxygen isotopes in a Rocky Mountain, Colorado watershed, (to be published in Journal of Hydrology)

Claassen, H. C., Modification of hydrogen- and oxygen-isotope composition during groundwater recharge at a watershed in the San Juan Mountains, Colorado, (to be published in Journal of Hydrology)

#### Studies of Processes and Techniques Related to the Location of High-Level Radioactive Waste Repositories in Crystalline Rocks

During the period 1982-1986, the DOE carried out a screening of three regions in the northeast, southeast and north-central United States to determine if suitable areas existed in which a second high-level, radioactive waste repository might be located. In addition, the DOE and the USGS carried out generic studies of 1) techniques for characterizing sites in crystalline rocks and 2) the processes occurring in crystalline rocks that might be relevant for the assessment of potential repository sites. Some of these generic studies were done in cooperation with the radioactive waste programs of Sweden and Canada.

In 1986, the DOE terminated the search for potential repository sites in the three regions but continued to sponsor generic studies of crystalline rocks as a host medium, as did the USGS. In 1987, passage of the Nuclear

Waste Policy Amendments Act mandated an end to all study of specific bodies of crystalline rocks as repository hosts; however, cooperative work of a generic nature with the Swedish and Canadian programs continued. Except for this cooperative work, the research described here was completed prior to 1987; some of the results were published in later years.

Isotopic Studies of the Eye-Dashua Lakes Pluton, Atikokan,  
Ontario

By Z. E. Peterman, Lakewood, Colorado

The Eye-Dashua Lakes pluton, located near Atikokan, Ontario, is one of the granitic plutons selected by Atomic Energy of Canada Limited (AECL) in 1979 for studies to assess the concept of nuclear waste disposal in crystalline rock. Investigations designed to characterize the pluton and to evaluate its suitability as a representative type of host rock include various aspects of geology, geochemistry, geophysics, and hydrogeology. In 1981, a cooperative project was formulated between the USGS and AECL to study the radiogenic-isotope systematics of the Eye-Dashua Lakes pluton.

Objective: To apply isotope geochemical techniques to evaluation of the Eye-Dashua Lake pluton for the long-term containment of radioactive wastes. To determine the timing of geologic events on regional and local scales for assessing long- and short-term geologic stability. To determine the effects of repeated tectonism on the integrity of crystalline rocks. To determine the sources and residence times of fluids and gaseous species in crystalline rocks, the migration of fluids and gaseous species, and their communication with the biosphere.

Approach: The establishment of the geologic framework of areas involves virtually all phases of isotope geochronology. Credible, long-range predictions concerning the performance of a repository rock mass as an effective barrier for the containment of radionuclides under a variety of hydrogeologic scenarios must include a thorough understanding of the past alteration history of the rock mass. The naturally occurring radiogenic-isotope systems (e.g. Rb-Sr, K-Ar-Ca, Sm-Nd, U-Th-Pb) sequestered in major and trace minerals are variably sensitive to alteration and recrystallization in low-temperature, hydrous environments. Ages of emplacement, as well as ages of subsequent alteration events, can be determined, and the degree, extent, and the scale of element mobility in the bulk rock and alteration zones can be determined. The past behavior of these isotope systems can be used to assess the future response of a rock mass to such

events and to predict its performance under analogous conditions. Additionally, some of the isotopes in the natural systems are geochemically similar or identical to those in spent fuel rods, and their behaviors may be analogous to the behavior of radionuclides that may be dissolved from the waste.

Progress: A series of reports on isotopic and dating studies of the Eye-Dashwa Lakes pluton near Atikokan, Ontario, conducted in collaboration with the AECL, was completed and published (Peterman and Kamineni, 1990).

Isotopic data obtained in the studies of the Eye-Dashwa Lakes pluton related broadly to two topics: tectonic stability of the pluton and the extent of rock-water interaction. Isotopic ages (determined on fresh samples) that are commonly interpreted to date crystallization or early magmatic cooling are in close agreement. A nearly concordant U-Pb age of 2665 Ma (Zartman and Kwak, 1990) agrees well with whole-rock Rb-Sr (Peterman and others, 1990) and whole-rock U-Pb (Doe and Peterman, 1990) ages of 2637 Ma and 2684 Ma, respectively. A whole-rock Sm-Nd age (Futa, 1990) of 2680 Ma (depleted mantle model) indicates a short residence time for the magma protolith.

Identical internal Rb-Sr isochron ages for drill-core samples, separated vertically by nearly one km, indicate rapid cooling through uplift some 100 m.y. after crystallization. The pluton was probably emplaced in an environment where the ambient temperature exceeded 300 degrees Celsius, and intermineral exchange of Sr continued until 2541 Ma, when it was uplifted rapidly and cooled. Fission-track ages of apatite (Naeser and Crowley, 1990) progressively decrease with depth from 515 Ma (4 m) to 376 Ma (993 m), suggesting extreme stability during the Phanerozoic Eon with a maximum uplift rate of 0.02 mm/y. The time-integrated uplift rate over the history of the pluton is much lower, about 0.008 mm/y, further suggesting long intervals of unusual crustal stability.

Local disturbances of the isotopic systems occurred in fracture and fault zones formed in the Early Proterozoic as indicated by a Rb-Sr isochron age of 2282 and perturbed U-Th-Pb systems. This Early Proterozoic fracturing event opened pathways for the intrusion of water which facilitated alteration of primary minerals, deposition of secondary minerals, and movement of Sr and Pb, possibly U, Th, and Rb, on a scale of centimeters or decimeters. However, Rb-Sr and U-Th-Pb systematics in large domains of fresh rock occurring between major fracture zones were not perturbed, indicating that matrix flow of water was extremely limited during the history of the pluton.

These studies of the Eye-Dashwa Lakes pluton illustrate the utility of the naturally occurring, radiogenic isotope systems for deciphering the alteration history of igneous rocks. Characterization of any rock mass for the possible containment of nuclear waste will be incomplete without a thorough understanding of the behavior of its radiogenic isotope systems.

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Effects of a Brief Thermal Pulse on the Mineralogy and Chemistry of Granite Gneiss in the Eastern Beartooth Mountains, Wyoming

By Brian D. Marshall, Lakewood, Colorado

The study of contact metamorphism as an analog to the heating that will occur adjacent to a mined geologic repository for high-level radioactive waste provides data on element

mobility not easily obtained in other ways. Although only certain aspects of contact metamorphism can be considered analogous to the conditions in a waste repository, the complete petrographic, chemical and isotopic characterization of an igneous contact zone can provide data pertinent to the problem of waste isolation.

Objective: To determine the effects of a limited thermal pulse on a mineral assemblage that is partly analogous to a high-level radioactive waste repository.

Approach: The site chosen was the Christmas Lake dike, a steeply dipping, Late Proterozoic diabase dike that intrudes Archean granite gneiss in the Beartooth Mountains of Wyoming. This geologic setting provides an excellent opportunity to study element mobility in response to a sudden thermal pulse. Thermal modeling of the 42.6m-thick dike indicates that it crystallized in  $\approx 9$  years and that the dike and wall rocks cooled to below 200 °C in less than 1000 years.

Samples of granite gneiss were collected along traverses nearly perpendicular to the contact out to a distance of  $\approx 25$ m. All of the samples contain potash feldspar, quartz and plagioclase as major minerals and show little evidence of gneissic texture.

Progress: Samples within  $\approx 3$ m of the contact exhibit some reaction textures with more anhedral and even embayed grains. In contrast to the major minerals, the minor phases, biotite, chlorite, and muscovite, all show trends in abundance with distance from the contact. Chlorite peaks at  $\approx 2$ m from the contact, and muscovite is completely absent within  $\approx 5$ m of the dike indicating a reaction relationship between the two minerals. Optical and x-ray diffraction analyses of the potash feldspars show that those within  $\approx 10$  m of the dike have been transformed to orthoclase; farther away from the contact the potash feldspars are microcline. The thermal model gives a maximum temperature of  $\approx 500$  °C at 10m from the contact.

Chemical analyses by X-ray fluorescence and neutron activation show that a wide variety of elements were mobilized in response to this brief thermal metamorphism. Of the major elements, only aluminum shows a definite trend with a maximum in normative corundum occurring at  $\approx 2$ m. Of the alkali and alkaline earth elements, only cesium shows evidence of mobility, again with a peak at  $\approx 2$ m. The high field strength elements niobium and tantalum show similar trends, and uranium is generally enriched toward the contact. Most striking is the disparity in behavior between the light and heavy rare-earth elements. Although the light rare earths show no evidence of mobility, the heavy rare earths show a definite peak at  $\approx 2$ m from the contact.

These data indicate that certain elements were mobile during the brief thermal pulse that affected the granite gneiss. The concentration of certain trace elements at  $\approx 2\text{m}$  from the contact indicates the presence of a fluid phase near the contact. The behavior of particular trace elements may be controlled by the mineralogy.

Report:

Marshall, B. D., and Futa, Kiyoto, 1991, Lanthanide element mobility and Sm-Nd systematics in a contact metamorphic zone: *Radiochemica Acta*, v. 52/53, p. 405-411.

Geochemistry of Trace Elements in Natural Waters

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The movement of radioisotopes, major elements, and trace elements through the hydrogeologic environment depends upon both biological and chemical as well as hydrodynamic processes. The chemical form of an element in an aqueous environment often determines which chemical or biological process is operating, e.g., adsorption on solids, precipitation, dissolution, uptake by biota, ion exchange, degassing, or oxidation-reduction. Chemical forms can be predicted from water analyses by simultaneous equilibrium calculations; however, there are several major uncertainties with this approach to chemical modeling.

Current limitations on chemical modeling include: (1) lack of adequate testing of speciation calculations against experimental and field data; (2) absence of a quantitative assessment of the redox status of natural waters; (3) absence of quantitative assessments of temperature, ionic strength, and compositional limits for use in specific models; (4) lack of adequate testing of adsorption model subroutines; (5) lack of an evaluation of available thermodynamic data; and (6) inadequate testing of equilibrium calculations against analytical determinations for specific forms of dissolved constituents.

Another aspect of hydrogeochemical processes of significance to the characterization of radioactive-waste repository environments is the evolution of ground-water chemistry in granitic terranes at great depth. Data on trace-element concentrations and trace-element geochemistry in deep ground waters from granitic rocks are almost nonexistent. Such data would be valuable in assessing the hydrogeologic characteristics of granitic rock masses in relation to their suitability for repository purposes.

Objective: The objective was to (1) define the limitations of current chemical models, (2) improve the reliability of current models to predict precipitation, dissolution, complexation, and bioavailability of elements in both contaminated and uncontaminated natural waters, and (3) improve our knowledge of trace-element behavior in deeply buried granitic rock masses.

Approach: A variety of laboratory, field, and theoretical techniques are being developed or adapted to overcome deficiencies in models and to test the reliability of models. These techniques include development of analytical methods for direct determination of free ion or bound species, and redox species, the compilation and evaluation of thermodynamic data used in speciation calculations, the performance of sensitivity analyses, and the investigation of field sites where water-mineral reactions can be observed directly and analytical and theoretical calculations can be tested. Water samples from the Stripa mine in Sweden, an experimental site for studying the suitability of crystalline rock for developing a repository, will be used to develop concepts and methodology for studying trace-element behavior in granitic rock masses and their application to the origin and evolution of ground waters from granitic terranes.

Progress: Research has continued on the relationship between fluid inclusions and the deep ground-water chemistry at Stripa, Sweden. Major ion chemistry is dominated by carbonate geochemistry, and a general model for hydrochemical processes in the Stripa ground waters has been formulated. Fluid inclusion leakage is now considered an important potential source of salts in ground-waters infiltrating crystalline rocks. Research is also underway on the Pocos de Caldas natural analogue study to investigate transport of radionuclides, colloids and microbes, as well as major and trace elements, at two sites having high concentrations of uranium and thorium. Thermodynamic data for chemical modeling of major ions and selected trace elements have been revised and published (Nordstrom and others, 1990; Nordstrom and May, 1989). A new approach to modeling chemical changes in seawater after it has infiltrated granitic bedrock has been investigated (Nordstrom, 1986, 1989).

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### Application of Borehole Geophysics to High-Level Radioactive Waste Disposal Studies

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Borehole geophysics, widely known as well logging, is one of the primary methods for characterizing the physical and hydrological properties of rocks encountered by boreholes. For this reason, well-logging is an important part of site characterization and hydrologic investigations related to the siting and monitoring of radioactive waste repositories. A detailed review of the potential applications of well logging in radioactive waste disposal and repository siting studies conducted in cooperation with Atomic Energy of Canada Limited (AECL) is given by Paillet (1990). In these studies, geophysical measurements in boreholes are shown to contribute to the larger scale investigation in several specific ways. These include the ability of well logs to provide a vertical profile of the in-situ properties of rocks adjacent to the borehole, where they are not affected by the release of stress, drilling damage, or alteration on exposure to the atmosphere. Careful manipulation of the measurement process allows rock properties to be evaluated at several different scales, increasing the ability to understand the effects of scale on rock bulk properties, and improving the ability to correlate logs with surface and tomographic soundings. Logs not only provide precise depth measurements with which to calibrate the vertical scale on such soundings, but also provide multiple, independent measurements. These multiple measurements can improve the ability to interpret much larger scale soundings and tomographs where anomalies are found using a single measurement, such as radar propagation or seismic velocities.

Objective: To improve the equipment and analysis techniques in borehole geophysics that have potential applications for high-level radioactive waste disposal studies.

Approach: Research is focused on development of techniques to distinguish lithologies and mechanical properties of rocks considered potentially suitable for radioactive waste repositories, techniques to identify and characterize fractures as potential flow conduits in otherwise nearly impermeable rocks, and techniques to monitor the hydraulics of flow in and around boreholes under naturally-occurring flow regimes or during hydraulic tests (Paillet, 1989).

Progress: Significant progress has been made in: 1) the application of acoustic full waveform logging methods to the interpretation of the geomechanical properties of rocks (Paillet and Cheng, 1990), and the estimation of fracture permeability in situ (Paillet, 1991); and 2) the development and application of a high-resolution flowmeter for rapid identification of water production from individual fractures during hydraulic tests (Hess and Paillet, 1990). These and other applications are reviewed below.

#### Major Research Thrusts

Low-permeability repository rocks - crystalline basement and shales - One of the primary problems in radioactive waste repository studies is the characterization of homogeneous, low-permeability rocks such as those of batholiths, continental shield areas, and thick shales or other plastic rocks in sedimentary basins. These rocks might be effective barriers in the migration of contaminants, and might not be as susceptible to fracturing and dilatation as more heterogeneous materials under the stresses imposed by the construction and thermal loading of a repository. Acoustic waveform logs appear especially useful in yielding estimates of the bulk moduli and microcrack distributions of homogeneous rocks by means of the interpretation of seismic velocities and attenuation inferred from them. Considerable progress has been made in recent years by this research project and other organizations in defining field procedures and data analysis techniques for the use of waveform logs in the characterization of the geomechanical properties of these rocks. These methods are reviewed in detail in Paillet and Cheng (1990).

Identifying and characterizing fractures - One of the most important measurements in characterizing potential repository rocks is the distribution of fractures that could serve as conduits for the migration of contaminants in otherwise impermeable rocks. Earlier work by this research project and others indicated that fractures can be reliably identified, using either a combination of conventional geophysical logs, or one of several different acoustic, electric, or optical borehole wall imaging systems (Paillet and others, 1990). Recent progress has demonstrated that a combination of image

logs designed to identify the location of fractures with other geophysical logs can be used to separate, qualitatively or semiquantitatively, the effects of borehole enlargement, alteration, and permeability in the composite geophysical response.

The scale problem in fractured or heterogeneous rocks - Even when the latest geophysical logging techniques furnish reliable estimates of fracture location, orientation, degree of associated alteration, and effective hydraulic aperture, individual fractures have proven difficult to relate to the large-scale mechanical and hydraulic properties of rock masses. For example, several recent studies have shown that large scale flow paths determined from tracer studies and cross-hole pumping tests bypass many fractures that appear permeable in individual boreholes, and that the orientation of the larger permeability zone over which flow occurs is different from the orientation of the individual fracture segments observed to be conducting the flow. The scale problem has been approached in recent studies by constructing logging systems to encompass different radii of investigation, and comparing the results of these studies with those of larger scale tomographic or vertical seismic profiling studies (Paillet and others, 1987; Paillet, 1989). These kinds of studies are expected to be an important part of fracture hydrology investigations at several research sites in the future.

#### New Equipment and Data Analysis Methods

Adaptation of existing equipment - Ongoing efforts in the characterization of low-permeability rocks, including their lithology and fractures, has focused on the modification of conventional geophysical logging techniques to improve resolution in such environments. This has involved the use of longer source-to-receiver spacings to increase the scale of investigation, the development of low-frequency acoustic transducers for generation of both deeper penetration into the formation and excitation of a limited number of acoustic modes, and expansion of data arrays (dual-detector neutron logging, acoustic waveform, and multi-channel gamma spectral logging). All of these techniques are designed to increase the amount of information and reduce the ambiguity associated with the interpretation of fractures, fracture alteration and subtle variations in lithology.

Fracture identification and characterization - Fracture characterization efforts are based on enhancement of acoustic televiewer borehole imaging, comparative studies relating image, acoustic, and other logs to detailed core descriptions of fractures, and development of specific criteria for assigning relative degrees of alteration to fractures.

Complete descriptions of these techniques are given by Paillet and Hess (1987) and Paillet (1988, 1989).

Acoustic characterization in-situ - The in-situ estimation of fracture permeability using acoustic waveform data (tube-wave) attenuation appears to be the single best borehole method for measuring fracture permeability. Recent case studies continue to demonstrate the close correlation between fracture permeability and tube wave attenuation, while recently published theory (Paillet and others, 1989) demonstrates that the observed attenuation can be directly related to the hydraulic properties of fractures. The latest results indicate that natural fractures are most effectively modeled as thin layers of porous materials, rather than plane-parallel openings of uniform aperture (Paillet, 1991).

Multiple scales of investigation - Because of the scale problem associated with interpretation of the hydraulic and mechanical properties of fractured rock, research has continued to focus on comparison of results obtained at multiple scales of investigation. An example of the comparison of fracture properties determined at several different scales of investigation using borehole techniques, compared to fracture properties obtained using surface soundings and cross-borehole measurements, is given by Paillet and others (1987).

High-resolution flowmetering in tight rocks - A recently-developed, high-resolution flowmeter (Hess, 1990; Hess and Paillet, 1990) has been improved and adapted for use in identifying depths where water enters or leaves boreholes in "tight" formations during pumping tests. For example, the flowmeter can be used to identify specific fractures where water enters and leaves observation boreholes during regional pumping tests. The latest applications of this technique are based on measuring the evolution of transient flows in such observation boreholes (Paillet, 1990).

Permeability profiling in boreholes - One application for the high-resolution heat-pulse flowmeter in fractured formations, is the measurement of vertical flow distributions during production or injection tests in order to generate vertical permeability profiles. This approach is useful for the direct measurement of permeability, and for identifying fracture intervals that need to be isolated for long term monitoring of hydraulic head distribution (Paillet and others, 1989). This application is important because packer installation and monitoring is a long term process, and changes during the middle of an extended regional pumping test, such as that generated by shaft dewatering at the AECL's Underground Research Laboratory (URL), can greatly complicate the interpretation of hydraulic data.

## Results from Research Sites

The various equipment modifications and analysis techniques have been applied to the research topics described above at several research sites in studies related to radioactive waste disposal, or at sites typical of those that might be considered for a high-level radioactive waste repository. The most important of these are described in the following paragraphs.

AECL research sites on the Canadian shield - The Underground Research Laboratory (URL) and several other sites in the Canadian Shield of Manitoba and Ontario, Canada, have been the primary locations for testing logging techniques related to repository siting. Numerous boreholes, including several more than 1000 m deep, have been logged at the URL from 1987 to 1989; the results of these investigations are described by Paillet (1988, 1989, 1990). These results include extensive studies of the application of acoustic tube-wave logging for the estimation of permeability in situ, advanced televiewer log interpretation, and the generation of permeability profiles using the heat-pulse flowmeter. The most recent study at the URL is based on two boreholes, 130 m apart, penetrating a typical subhorizontal permeability zone. The zone was shown to connect the two boreholes as a single, planar zone of low radar velocity about 5 m thick in a tomography survey obtained by AECL. The objective of our study was to identify how individual fractures within and adjacent to the permeability zone conducted flow during pumping of one of the boreholes. This is an important aspect of the scale problem in relating individual fractures identified in boreholes to larger-scale ground-water flow paths. The results indicated a complex pattern of permeability values making up larger scale hydraulic features that serve as regional flow conduits (Paillet, 1989).

NAGRA site at Siblingen, Switzerland - The Swiss National Cooperative for Radioactive Waste Storage (NAGRA) sponsored a test of our permeability profiling techniques in a 1550 m deep borehole near Siblingen, Switzerland in 1989 (Paillet and others, 1990). The results were useful in identifying numerous zones of fracture permeability. They demonstrate the finite transmissivity of several fracture zones below 1400 m in depth, and give relative transmissivity values consistent with data from other NAGRA studies.

Argonne, Illinois site - Various conventional geophysical logs, the acoustic borehole televiewer, and the heat-pulse flowmeter were used to investigate the fracture flow characteristics of a carbonate rock body located at a site near Argonne, Illinois. The results of the study showed how the quality of the water sampled in boreholes could be related to the flow distribution produced by naturally

occurring hydraulic head differences between individual fractures. The flowmeter was also used to monitor transient flows along boreholes between fractures during pumping tests in adjacent boreholes. These results greatly improved the efficiency of efforts to define zones within boreholes for packer isolation and testing.

New England fractured rock study sites - Geophysical logs were obtained at a number of crystalline rock study sites in New England, including a nearly 2000-m deep borehole in gabbroic basement rocks near Gloucester, Massachusetts. Acoustic televiewer logs obtained in this borehole can be used to identify the vertical distribution of stress in situ associated with intrusive contacts and fractures. Analysis of these televiewer logs indicates rotation of the regionally east-west compression in association with contacts and fractures. These results and similar data from the URL are being analyzed to determine the relationship between lithology, stress, and fracture permeability.

Front Range crystalline rock studies, Colorado - Numerous studies in basement crystalline rocks appear to indicate that permeability along fracture flow paths is controlled by the "bottlenecks" where individual segments of permeable fractures intersect. One way to characterize these interconnections is to quantify the changes in fracture permeability associated with hydraulic stimulation and sand injection. A recent study (Paillet and others, 1989) indicates that such changes can be identified. In this example, the stimulation appeared to affect several relatively minor fractures, while no water was produced from fractures with the largest apparent aperture on the televiewer log, either before or after stimulation. These preliminary results demonstrate that such stimulation studies can provide important insights into the hydraulics of flow paths in fractured crystalline rocks.

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## TRANSURANIC WASTE

### Waste Isolation Pilot Plant, Southeastern New Mexico

#### Regional Ground-Water Flow System

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Understanding of the hydrodynamics of ground waters in saline strata at the Waste Isolation Pilot Plant (WIPP) in the Delaware Basin of southeastern New Mexico is important for evaluating the containment capability of this proposed facility. The water-bearing zones in the mixed evaporitic strata of the Rustler Formation above the repository horizon in the Salado Formation salt have been of principal concern with respect to the possible transport of radionuclides to the biosphere by ground water. The flow paths, ground-water velocities, hydrochemistry, and other properties differ with each zone in the Rustler, whereas previous analytical and numerical modeling considered the formation as a single unit. The complex flow system in the Rustler Formation in some areas is associated with dissolution and collapse of evaporative rocks, whereas in other areas water movement in individual zones is under confined conditions, with vertical communication inhibited by sequences of nearly impermeable anhydrite and halite strata. Interpretation of the flow system using conventional methods is hindered by very low permeabilities and the dominance of fracture flow where significant movement does occur.

Objective: To model the regional flow systems in the individual water-bearing zones of the Rustler Formation, with special emphasis on the Culebra Dolomite Member, which was identified as the most likely to support significant regional ground-water movement. The study compliments more detailed site-specific well-testing and modeling efforts by Sandia Laboratories and associated contractors in the WIPP area.

Approach: The USGS and others have conducted hydrogeological investigations continuously since 1975 in support of geotechnical analysis of the site. A comprehensive data base was developed and evaluated for input to the models. Multi-dimensional simulation codes were used to develop a regional-scale, variable-density model of ground-water flow. Baseline, approximately steady-state flow simulation for the Culebra Dolomite Member of the Rustler Formation, was calibrated to the distribution of equivalent fresh water

heads. Investigation included analysis of the relative magnitude of pressure-related and density-related flow-driving forces, and sensitivity simulations to analyze boundary effects and vertical flux.

Progress: A regional-scale, variable-density flow model was developed that could serve as a base not only for hydrodynamic analysis and transport simulations, but for geochemical consideration as well. The flow field from baseline simulation, in conjunction with long-term brine transport patterns, indicates that flow velocities are relatively fast west of the site and extremely slow north and east, with high variability in between. Force analysis indicates that density effects on flow are not significant at the WIPP site, but are particularly important in the area to the south because of potential transport pathways from the site.

Sensitivity simulations from the model suggest that the central and western parts of the region, including the WIPP site, are insensitive to boundary conditions north and east of the site. However, simulation of a head increase along the Pecos River boundary suggests that about half of any change in river elevation would eventually reach the WIPP site, though there is considerable uncertainty about how long this would take. Other simulations indicate that as much as 25 percent of total inflow to the Culebra Dolomite Member could be entering as vertical flux, mostly in the transition zone adjacent to Nash Draw. In addition, a simple cross-sectional model was developed to examine response of the flow system to recharge during a past glacial pluvial. This modeling suggests that the system as a whole drains very slowly and could sustain flow from purely transient drainage following Pleistocene recharge.

#### Reports:

Davies, P. B., 1989, Variable-density ground-water flow and paleohydrology in the Waste Isolation Pilot Plant (WIPP) region, southeastern New Mexico. U.S. Geological Survey Open-File Report 88-490, 139 pp.

Richey, S. F., 1989, Geologic and hydrologic data for the Rustler Formation near the Waste Isolation Pilot Plant, southeastern New Mexico. U.S. Geological Survey Open-File Report, 89-32.

## Normative Analysis of Ground Waters from the Rustler Formation,

By Marc W. Bodine, Jr., Lakewood, Colo., and Blair F. Jones, Reston, Va.

A diagnostic chemical and/or mineralogic characterization of natural water can aid in the interpretation of solute origin and the identification of important water-rock interaction in subsurface environments. It can also contribute to determining the evolution of water chemistry within a flow system. The "salt norm" (SNORM code of Bodine & Jones, 1986) was originally developed to provide such a characterization for the wide range of compositions, solute origins, and flow regimes present in the saline subsurface waters associated with the WIPP site.

Objective: To use the techniques of normative analysis not only to provide a characterization of the hydrochemistry of the ground waters of the Rustler Formation at the WIPP area, but to relate the wide chemical variation in these fluids to the range of detail in the geologic and hydrodynamic framework of the region, and to indicate the major processes involved in their geochemical evolution.

Approach: After review, testing, and compilation of existing data on area well-water chemistry, including additional determinations, especially chloride/bromide ratios, salt norms and simple salt assemblages were calculated, using the USGS SNORM code, for 124 analyses from the water-bearing units of the Rustler Formation (plus 4 analyses from overlying strata). Geochemical interpretations were worked out and related to the hydrodynamics of the flow system in the Rustler.

Progress: Normative analysis indicates that Rustler Formation waters range from hypersaline, primitive-diagenetic fluids, probably syngenic, to meteoric recharge waters. The former exist in isolated areas of the Culebra and Magenta Dolomite aquifers in the Rustler because of very low formational transmissivity. The extensive infiltration of the latter in areas where the water-bearing units of the Rustler Formation are not overlain by halite is correlative with the normative sulfate content. Much halite dissolution effected by Rustler-Salado contact-zone waters takes place in the uppermost Salado Formation, whereas mixtures of primitive-diagenetic solutes with alkali sulfate-bearing recharge can produce NaCl-rich fluids without halite dissolution. Ground-water flux from overlying continental (siliciclastic) sediment can be detected in minute levels of normative alkali sulfate. Interpretation of the norms is

consistent with the north-south flow direction and velocities in the Davies (1989) model, and is compatible with paleo-recharge from Nash Draw to the north and west of the WIPP site.

Report:

Bodine, M. W., Jr., and Jones, B. F., 1990, Normative analysis of ground waters from the Rustler Formation associated with the Waste Isolation Pilot Plant (WIPP), southeastern New Mexico: in Fluid-Mineral Interactions: A Tribute to H.P. Eugster; The Geochemical Society, Special Publication No. 2, p. 213-269.

Normative Analysis of Brines from Pre-Rustler Strata

By Scott Anderholm, Albuquerque, New Mexico and Blair F. Jones, Reston Va.,

The comprehensive model for regional ground-water flow in the Rustler Formation, described in the previous section, enabled study of the deeper brines of the WIPP site area and more detailed investigation of possible important geochemical reactions at all levels.

Objective: To extend the flow system information and geochemical considerations to more detailed examination of solute characterization and origins in Salado Formation brines and deeper brines, and to study the mixing effects of other fluid solute matrices from above or below the repository horizon, searching for extraneous inputs and their influences.

Approach: The techniques and criteria of normative analysis (SNORM) used in the analysis of ground-waters from the Rustler Formation are being applied to the investigation of Salado brines. Studies include fluid inclusions, the underlying Castile and deeper fluids, and fluids from the Capitan reef aquifer. Mass transfer modeling is being performed on well-to-well hydrochemical variations along indicated or projected flow paths, particularly in the Culebra Dolomite aquifer, and on potential fluid mixes in the overall system. These efforts are employing special modifications of the USGS BALANCE and PHRQPITZ mass transfer codes to include mineral dissolution-precipitation effects, the nature of variations in Rustler-Salado brines, and the consequences of other natural fluid inputs.

Progress: Normative comparison of Salado brines with natural and artificially evaporated seawater suggests that Salado brines may have such an origin, but there is a clear

depletion in sulfate and a more subtle variation in potassium. Both these differences are compatible with small amounts of syndepositional mixing of evaporative brine with continental or normal marine waters. With very few exceptions, brines appearing in the workings of the repository horizon are higher in normative potassium chloride than any of the inclusions, even those from the McNutt potash zone, suggesting preferential potash leaching by fluid of even very limited mobility. Castile Formation "pocket brines" are unique and hydrochemically may reflect a complex redox reaction sequence initiated by migration of methane. Deeper brines are typical, diagenetic, sedimentary basin "oilfield" fluids whose normative signature precludes direct connection with overlying brines from the repository horizon.

Mass transfer calculations are being conducted to consider combinations of mineral-, water-, and gas-reactions, plus mixing of different ground waters, that are consistent with the mass transfers between pairs of wells along postulated modern flow paths in the Culebra Dolomite Member. A large number of possible reaction sets correspond with the observed hydrochemical and mineralogical data, but the mass transfers cannot be modeled by mixing of Culebra or Rustler-Salado contact waters. In general, two kinds of models fit the observed changes in water chemistry. One set of models requires ion exchange; the second set needs dissolution of large amounts of sparsely occurring evaporite. The former type seems more reasonable.

## Transuranium Elements in Ground Water

by J. M. Cleveland, Lakewood, CO

In view of the long half lives of many of the transuranium elements (plutonium, neptunium, and americium) and uncertainties regarding their geochemistry, it is difficult to predict their transport and distribution characteristics in ground waters. In particular, it is fruitless to attempt to make distribution measurements of the transuranium elements or to model their ground-water transport without first determining their chemical forms and solubilities in the ground water of interest. This speciation is relatively straightforward for elements with only one common oxidation state, such as americium, but is somewhat more complex for the multivalent elements, plutonium and neptunium.

Objective: To define the chemical speciation and the chemical and hydrologic behavior of transuranium elements in ground waters.

Approach: Research has been conducted, primarily on americium solubility as a function of ionic strength and on

the speciation of plutonium and americium in ground waters from strata at the Idaho National Engineering Laboratory (INEL).

Progress: Variation of americium solubility with ionic strength: Several studies have been conducted on the effect of ground-water composition on plutonium and americium speciation. The influence of water composition and redox potential on solubility were obvious, but there was no clear-cut indication of the effect of ionic strength. To remedy this lack, a study was undertaken to determine the solubility of americium in solutions containing various concentrations of sodium perchlorate, a noncomplexing solute, dissolved in deionized water. The results indicated a small inverse effect of ionic strength on americium solubility. At pH 7.3, americium solubility after 30 days decreased from 16 per cent at an ionic strength of 0.001 M to about 5 per cent at an ionic strength of 0.1 M; that is, a hundredfold increase in ionic strength decreased solubility by only about a factor of three.

Speciation of plutonium and americium from ground water at INEL: Previous studies have firmly established the influence of ground-water composition and redox properties on plutonium speciation and americium solubility. Because the Radioactive Waste Management Complex (RWMC) at INEL contains sizable quantities of both of these elements with minimal containment, a study of plutonium speciation in ground waters from the RWMC area was undertaken. The study is still in progress, but results from four of the anticipated eight to ten waters that will be studied have already been obtained.

The study involves the addition of plutonium or americium to samples of the waters, storing them in tightly-capped Teflon containers in darkness for various time periods up to 30 days, and sampling them for plutonium (or americium) concentration before and after filtration through a 0.1-micrometer filter, and also for plutonium oxidation-state-distribution in the filtrate. (Since americium would occur only in trivalent state, its oxidation state was not determined.) In the plutonium runs, waters were spiked with either high-oxidation-state [(PuV) and (VI)] or low-oxidation-state [Pu(III) and (IV)] plutonium. After they were spiked, the waters were adjusted to their original pH values, which ranged from about 8.0 to 8.5. The results indicated that RWMC waters tended to be oxidizing towards plutonium. In the high-oxidation-state runs after 30 days more than 95 per cent of the plutonium remained in solution, predominantly in the oxidized form. In the low-oxidation-state runs, the percentages of added plutonium remaining in solution in each of the four waters were as follows: well 88, 18; well 177, 34; well 120, 60; production well, 72. The influence of redox properties is evident from the fact that

the plutonium remaining in the production-well water, which was added in the reduced form, was predominantly in the oxidized form after 30 days. The enhanced solubility of plutonium in the production-well water suggests that existing conditions at the RWMC may enhance the possible mobilization and transport of this element.

Americium, being present in the trivalent state, tends to behave like plutonium (III). After 30 days, the percentages of added americium remaining in solution in each of the four waters were as follows: well 88, 9; well 117, 24; well 120, 31; production well, 29. Note that there is a general parallel between the solubilities of the two elements in the four waters.

LOW-LEVEL RADIOACTIVE WASTE AND MIXED LOW-LEVEL RADIOACTIVE  
AND HAZARDOUS WASTES

Investigations at Commercial Low-Level Radioactive Waste  
Burial Grounds

From the late 1970s into the mid-1980s, the USGS conducted hydrologic studies at five commercial low-level radioactive waste sites: Beatty, Nevada; Sheffield, Illinois; West Valley, New York; Maxey Flats, Kentucky; and Barnwell, South Carolina. The sites included a range of distinctly different geohydrologic settings and climates that range from arid to humid. Reviews of the hydrologic conditions at these sites provide a background of useful information that can be considered in establishing criteria for future site selection and design of low-level radioactive waste repositories. Information derived from the studies and a summary of lessons learned from past practises have been presented by Bedinger (1990).

Studies at one of the commercial sites, Beatty, are being continued as a research project to understand fundamental processes operating in arid zones where the water table is deep. Final reports at the other 4 sites have been completed.

Report:

Bedinger, M. S., 1989, Geohydrologic aspects for siting and design of low-level radioactive waste disposal:  
U. S. Geological Survey Circular 1034, 36 p..

Beatty, Nevada

By B. J. Andraski, Carson City, Nevada, Jeffrey M. Fischer, West Trenton, New Jersey, and David E. Prudic, Carson City, Nevada

A low-level radioactive-waste disposal facility in the Amargosa Desert of Nevada, about 17 km southeast of Beatty and 169 km northwest of Las Vegas, has been operating since 1962. This was the first commercially operated radioactive waste disposal facility in the United States. Wastes at the facility are emplaced in 2 to 15-m deep trenches and covered by backfilling with previously excavated materials. Annual precipitation in the area averages about 112 mm. Vegetation is sparse with creosote bush (*Larrea tridentata*) being the dominant species. Soils in the area are skeletal and are underlain by more than 170 m of unconsolidated alluvial-fan,

fluvial, and ephemeral-lake deposits. Depth to water is about 85 m.

Initial field investigations (1976-1980) included monitoring of soil-water content and water potential in an unvegetated soil profile, and collection of meteorological data at the disposal facility. Design of additional hydrogeologic investigations and long-term studies of soil-water movement in a vegetated soil profile began in 1982 and field data collection has been ongoing since 1984. Studies to evaluate the modifying effects of trench construction on the natural site environment and to determine changes in trench structural stability began in 1987. Design of studies to measure gas and vapor movement in the trenches at the facility began in 1989.

Objectives: To define the hydrology of the extremely dry, upper-unsaturated zone at the site under natural conditions, with emphasis on estimating the downward flux (recharge rate) of water through the vegetated soil profile under current climatic conditions; to describe the meteorological conditions at the site; to better define the geology and ground-water system in the immediate area; and to determine if radionuclides from buried waste have migrated to ground water.

To determine the effects of trench construction on the natural site environment; to determine rates and directions of soil-water movement within and adjacent to trenches, allowing estimation of potential transport rates and the extent of radionuclide migration within and beneath the burial trenches; to define the mechanisms and soil properties that control soil-water movement within and adjacent to the trenches under unvegetated-current climatic conditions; and to determine trench subsidence and erosion rates.

Evaluate and develop methods for reliable measurement of physical properties and water movement in the dry alluvial soils and subsoils at this site and with transfer capability for use in similar environments.

Approach: A vertical shaft provides access for installation and retrieval of instrumentation in the upper 13 m of the soil profile at the vegetated site (Morgan and Fischer, 1984). Soil-water potential and temperature are measured using thermocouple psychrometers. Soil-water content is measured to a depth of about 30 m with a neutron-moisture probe. A limited number of soil samples were collected for laboratory determination of chemical and hydrologic properties.

The meteorological environment at the site is being described through measurements of precipitation, air temperature,

relative humidity, incident solar radiation, wind speed, and wind direction.

Two experimental burial trenches were constructed in September 1987 by means of burial practices that simulated those used at the disposal facility (Andraski, 1990a). The trenches and the undisturbed area between the trenches are kept free of vegetation. During construction, samples of the undisturbed soil profile and trench backfill were collected at several depths. The samples are being used to characterize physical properties of the trench backfill, relative to those of the natural soil profile, and to evaluate the vertical variability of these properties through analysis of variance techniques. Rates and directions of soil-water movement within and adjacent to the experimental trenches are being determined by measurements of soil-water content and water potential using a neutron-moisture probe and thermocouple psychrometers, respectively. Devices used to monitor trench subsidence and erosion were installed during and(or) immediately following trench construction.

Progress: From 1981 through 1989, an annual average of 112 mm of precipitation was recorded at the waste disposal facility (R.L. Marchand, US Ecology, Inc., written commun., 1990). Precipitation falls predominantly in the winter, typically in the form of rain, and ranged from as little as 17 mm in 1989 to as much as 225 mm in 1983. Air temperatures typically range from a minimum of about 20<sup>0</sup> C. to a maximum of over 40<sup>0</sup> C. during the summer and from below 0<sup>0</sup> C. to a maximum of about 15<sup>0</sup> C. during the winter. Maximum incident solar radiation typically exceeds 1000 W/m<sup>2</sup> during the summer.

Soil-water monitoring at the instrument-shaft site continues. Soil-water contents measured between 1984 and 1988 indicate that, under vegetated conditions, redistribution of infiltrated water did not occur below a depth of about 1 m in the soil profile. Soil-water contents measured below a depth of 10 cm typically ranged from about 0.02 to 0.08 kg/kg and varied little over time. Soil-water potentials measured from depths of 2 to 13 m, ranged from about -3.0 to -5.5 MPa. Soil-water potential gradients and soil temperature gradients varied seasonally above a depth of 9 m. From 9 to 13 m, temperature and soil-water potential gradients indicate relatively constant upward movement of soil water. Calculated liquid-water fluxes were typically less than 1x10<sup>-7</sup> g/cm<sup>2</sup>/d. On the basis of chloride concentrations determined for sediment samples, the long-term maximum percolation depth of liquid water appears to be eight meters. Water movement through the undisturbed sediments appears to be dominated by vapor flow with calculated fluxes generally being near 1x10<sup>-5</sup> g/cm<sup>2</sup>/day. Because water vapor does not carry dissolved constituents, the major threat of contaminant migration under

current conditions at this study site would be from radionuclides, such as tritium and carbon-14, that can move as vapor.

Visual examination of excavations and lithologic and geophysical logs from boreholes indicate the sediments consist of fluvial, debris-flow and ephemeral lake deposits with a great range in thickness and areal extent. Since 1988, several test holes and monitoring wells have been drilled under the auspices of the site operator, US Ecology, Inc., near the low-level radioactive waste facility. Data from these holes have been made available to the USGS. The depth to ground water east of the facility is typically 85 m below land surface and the gradient is about 0.03 m/m to the south-southeast. Water levels drop considerably across the low-level radioactive waste facility with depth to ground water near 110 m below land surface near the southwest corner. The gradient across the facility is about 0.06 m/m in a southwest direction. The change in gradient and direction of flow implies that some type of barrier inhibits westward flow across the facility. Additional test holes and monitoring wells are being drilled by the site operator, and a seismic survey is planned adjacent to the waste facility to test this hypothesis.

Water samples from six wells were collected in August 1989 for determination of dissolved major and minor constituents, radionuclides, and stable isotopes. Water from all wells sampled has nearly the same chemical constituents. The dominant cation is sodium, and the dominant anions are bicarbonate and sulfate. These constituents are typical of ground water in the area. Dissolved silica concentrations range from 67 to 73 mg/L and fluoride concentrations range from 2.5 to 4.0 mg/L. Gross-alpha and gross-beta concentrations are similar for all wells with gross alpha ranging from 5.8 to 14.6 pCi/L as uranium and gross beta ranging from 13.6 to 17.5 pCi/L as strontium-90. A gamma scan detected concentrations of potassium-40, radium-224, and thorium-234, all naturally occurring radionuclides in the sediments. Tritium concentrations are at or near background (<2.5 pCi/L) except for one water sample which has a concentration of about 6 pCi/L. The slightly higher concentration in this sample may have been caused by remnant drilling fluid in the well. Radon concentrations range from 318 to 457 pCi/L and are typical of ground water in the area.

Laboratory analyses of soil samples collected during construction of the experimental trenches continues. Initial results showed soil-water content in the undisturbed profile ranged from 0.013 to 0.095 m<sup>3</sup>/m<sup>3</sup>, compared with 0.027 to 0.041 m<sup>3</sup>/m<sup>3</sup> for the backfill. Profile bulk density ranged from 1.36 to 1.91 Mg/m<sup>3</sup> versus 1.54 to 1.74 Mg/m<sup>3</sup> for the backfill. Rock-fragment content in the profile ranged from

0.11 to 0.63 kg/kg, compared with 0.34 to 0.49 kg/kg for the backfill. Relative to the natural soil profile, water content, bulk density, and rock-fragment content values for the backfill were generally greater near the near surface and less at depth. Although several significant differences were observed at depth within the soil profile, none were typically observed within the backfill. Textural classification (<2-mm fraction) for the soils ranges from sandy loam (76% sand, 18% clay) to sand (95% sand, 3% clay). Texture of the backfill is loamy sand throughout the vertical profile. Saturated-hydraulic conductivity values range from  $3.5 \times 10^{-4}$  to  $4.0 \times 10^{-2}$  cm/s for the soil profile and from  $1.2 \times 10^{-4}$  to  $9.3 \times 10^{-4}$  cm/s for the backfill. Determination of soil-water retention is currently in progress. Preliminary laboratory results indicate that construction of shallow land burial trenches can significantly modify the natural site environment. Whereas the vertical variability of the soil profile could greatly impede percolation, the lack of such variability in the backfill could result in increased infiltration and redistribution. In addition, water movement into the trenches could occur as a result of lateral flow from the soil profile. Both of these factors could increase the potential for migration of radionuclides.

Soil-water monitoring at the experimental trench facility continues. To date, soil-water content data indicate that infiltrated waters have not penetrated more than about 1 m below the surface of the trench covers. Soil-water content of the near-surface backfill ranged from an initial and present low of about  $0.030 \text{ m}^3/\text{m}^3$  to a maximum of  $0.11 \text{ m}^3/\text{m}^3$ , following 25 mm of precipitation in January 1988. Soil-water content of the deeper backfill and the undisturbed soil beneath the trenches remained relatively constant at about  $0.048 \text{ m}^3/\text{m}^3$  and  $0.062 \text{ m}^3/\text{m}^3$ , respectively. As much as 84 percent of the total precipitation that infiltrated the trench covers has been depleted by evaporation. To date, soil-water potentials in the backfill have ranged from greater than -1.0 MPa near the surface to less than -7.8 MPa below 1.25 m. Soil-water potentials in the undisturbed soil adjacent to and beneath the trenches typically range from greater than -1.0 MPa in the upper meter to less than -5.7 MPa at depths of 1.85 to 5.5 m. Soil-water potential gradients indicate that water is moving toward the simulated waste from all three directions, but the quantity of water and rates of movement are small. Water that infiltrated the trench covers and water from the undisturbed soils adjacent to and beneath the trenches has not come in contact with the simulated waste. Under current climatic conditions at this arid site, low precipitation amounts and high evaporation rates limit the amount of infiltrated water that may come in contact with the waste.

Structural monitoring at the experimental trench facility continues. Within the first year, 80 to 95 percent of the subsidence within the trench body occurred during construction, with maximum values of 23 and 46 mm being observed within the stacked-drum and random-drum trenches, respectively. Total subsidence for both trench covers was about 6 mm. More than 80 percent of the total subsidence was observed within the first six months following construction. Most of the observed changes in trench-cover elevation were attributed to natural settling of the uncompacted backfill. Drum placement appeared to have no effect on surface subsidence of the trench covers. Soil loss from the surface of one trench totaled about 7 mm, whereas no measurable soil loss was observed from the second trench. Most of the observed soil loss was attributed to deflation. Lack of erosion for the second trench was attributed to a higher percentage of rock fragments armoring the surface.

The lack of percolating water could help preserve the structural stability of the trenches. Field monitoring will continue to evaluate whether or not adequate containment of low-level radioactive waste can be provided by the shallow land burial techniques used at this type of arid-site facility.

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Andraski, B. J., 1990a, Water movement and trench stability at a simulated arid burial site for low-level radioactive waste: LaGrange, IL, Am. Nuclear Soc., Proc. of the Topical Conference on Nuclear Waste Isolation in the Unsaturated Zone, Las Vegas, Nevada, September 17-21, 1989, p. 166-173.

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Andraski, B. J., 1990c, Rubber-balloon and drive-core sampling for determining bulk density of an alluvial desert soil [abs.]: Agronomy Abstracts, v. 82, p. 208.

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### Sheffield, Illinois

By Patrick C. Mills, Urbana, IL

The Sheffield low-level radioactive-waste disposal site, located in northwestern Illinois, was operated from 1967 to 1978. Wastes were buried at the 8-ha site in 21 disposal trenches constructed in unconsolidated sediments of glacial and post-glacial origin. The drainage divides of three small drainage basins form natural hydrologic boundaries for the site. Ground-water divides correspond with the topographic drainage divides. Precipitation is the source of water for the hydrologic system, as there is no underflow from adjacent areas except for flow from a divide just west of the site. Average annual precipitation is about 846 mm, of which 635 mm evapotranspires, 140 mm runs off, and 77 mm recharges the underlying outwash-sand aquifer.

Hydrogeologic studies of the site were undertaken from 1976 to 1987. As part of the early work, a study of water and tritium movement through the unsaturated zone was conducted; results from 1981 to 1985 are described in Mills and Healy (1987). Described herein are the final (1986-87) results of

that study, as well as results of a study of gas transport in the unsaturated zone.

Objectives: The objective of the 1981 to 1987 study of water movement in the unsaturated zone was to define the mechanisms that control the movement of water and radionuclides through the trench covers and beneath the trenches. Objectives of the later studies were: 1) to determine longer-term (greater than 2 years) temporal trends in the water budget and in water movement and water chemistry in the unsaturated zone and saturated zone; 2) to characterize water movement (with emphasis on preferential flow) and water chemistry in an unsaturated sand deposit that directly underlies several trenches; and 3) to determine the effect of a research tunnel (constructed to provide access to the below-trench environment) on water movement in the adjacent geologic deposits.

The objectives of the study of gas transport in the unsaturated zone were: 1) to identify, quantify, and determine the distribution of gases in the unsaturated zone adjacent to buried low-level radioactive wastes; 2) to estimate the fluxes of waste-generated gases from a disposal trench to the surrounding unsaturated zone; and 3) to determine physical and chemical properties that retard the transport of carbon-14 dioxide in unsaturated deposits.

Approach: A gamma-attenuation soil-moisture gage was used to monitor soil-water movement into trenches and to provide data for a water-budget analysis. A 2-m-diameter by 120-m-long horizontal tunnel provided access for monitoring glacial till and sand deposits below four trenches. Soil-water movement and water chemistry in the unsaturated zone were monitored using soil-moisture tensiometers, gravity (free-drainage) lysimeters, vacuum lysimeters, and sediment cores. Depth to the water table was measured in wells; samples from wells were used to describe ground-water chemistry. A cross-sectional, numerical, ground-water-flow model aided interpretation of the effect of the tunnel on water movement.

Three boreholes were augered to bedrock and five gas piezometers were installed in each borehole for collection of gas samples. Time-averaged partial pressures of methane and carbon-14 were numerically simulated using a two-dimensional finite-difference solution for Fickian diffusion.

Progress: Annual (July through June) precipitation decreased from 929 mm (1982-83) and 968 mm (1983-84) to 774 mm (1984-85), 864 mm (1985-86), and 695 mm (1986-87). A corresponding decrease occurred in estimates of annual drainage to the trenches and(or) saturated zone below the trenches from 107 mm (1982-83 and 1983-84) to 49 mm (1984-85), 74 mm (1985-86), and 48 mm (1986-87).

The seasonal pattern of early spring recharge to the unsaturated zone below the trenches, observed from 1981 to 1985, was obscured in 1986-87 by the large reduction in pressure-head fluctuation and an overall decline in pressure heads; also, an additional recharge period was observed in the fall, 1986. Peak soil-water fluxes at two gravity-lysimeter locations decreased from about 15 and 11 mm/d, respectively in 1985 to about 0 and 6 mm/d, respectively, in 1986-87. Water-table altitudes decreased from historical highs in March 1985 to near-historical lows in 1986-87.

Average tritium concentrations in soil water increased from 70,100 pCi/L from July 1982 through June 1984 to 153,000 pCi/L from January 1986 through June 1987 at below-trench vacuum-lysimeter locations. Tritium concentrations as high as  $15 \times 10^6$  pCi/L were detected in soil water from the below-trench till deposit. Volatile organic compounds were detected in a synoptic sample from the below-trench sand deposit. Concentrations of inorganic ions in soil water and ground water changed little from 1982 to 1987.

Flow is unevenly distributed through the unsaturated sand deposit. Slow, continuous water movement appears to occur through most areas of the deposit; localized, preferential flow along near-saturated to saturated flow paths occurs as well. Saturated, preferential flow paths in unsaturated sand have been identified in other studies, but rarely in field settings. The drainage data indicate that the flow paths could be less than  $1 \text{ mm}^2$  in cross-sectional area. Because of their localized occurrence and small size, the flow paths were not readily identified by soil-moisture tensiometers. Average annual tritium flux through the sand deposit was estimated to be  $0.59 \text{ uC/yr/cm}^2$ .

The location of preferential flow paths in the sand deposit appears to be related to the location of flow paths in the overlying trenches. The timing and rates of water movement are related to precipitation patterns, seasonal climatic cycles, and factors such as trench-interior characteristics.

Tritium concentrations in the water moving preferentially through the sand deposit fluctuate seasonally. Concentrations increase as tritiated water is flushed from the trenches during spring recharge; in some cases concentrations decrease, apparently as the result of dilution by recent recharge water. Changes in tritium concentrations also appear to occur as waste containers deteriorate and flow paths in the unsaturated trenches change location. Pressure-head data from tensiometers, soil-moisture content, and tritium-concentration data from sediment cores and numerical simulations indicate that the tunnel has neither a consistent nor pronounced effect on the natural movement of

water. The tunnel appears to disturb natural water movement most directly above and below the tunnel.

Gases identified in the unsaturated zone were of natural and waste origin. The gases included nitrogen, oxygen, argon, carbon dioxide, methane, propane, butane, tritiated water vapor, carbon-14 dioxide, and radon-222. Partial pressure gradients of methane and carbon-14 dioxide originated at the wastes; carbon-14 dioxide partial pressures exceeded natural background partial pressures by factors greater than  $10^6$  at some locations. Variations in partial pressures and carbon dioxide were seasonal because of increased root and soil-microbe respiration during summer. Variations in methane and carbon-14 dioxide partial pressures were apparently related to discrete releases from waste sources at unpredictable intervals. No greater-than-background partial pressures for tritiated water vapor or radon-222 were measured.

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Maxey Flats, Kentucky

By Mark A., Lyverse, Karen S. Wilson, and Bridgette E. Lyons,  
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The closed Maxey Flats Low-Level Radioactive Waste Disposal Site near Morehead in northeastern Kentucky encompasses approximately 1.13 square kilometers near the edge of a flat-topped ridge. The ridge is underlain by fractured shale and sandstone beds of the Nancy Member and the Farmers Member of the Borden Formation of Mississippian age. The only appreciable ground-water flow in the strata beneath the site occurs through fractures, and flow patterns are difficult to delineate. The configuration of the potentiometric surface also is difficult to determine because several saturated and unsaturated zones are present in the rocks.

Objective: To describe the subsurface hydrology of the site in terms of volumes, rates, and pathways of ground-water flow from burial trenches and to relate the hydrology to the migration of leached radionuclides so that generalizations can be made regarding the fate of buried waste in this type of geohydrologic environment.

Approach: The shallow sedimentary ground-water system has been described based on the collection of water-level and water-quality data from approximately 70 wells. Water-level data were also collected from about 20 burial trenches.

Ground-water levels and occurrence of tritium in ground water at the site were monitored by the USGS, in cooperation with the Kentucky Natural Resources and Environmental Protection Cabinet from June 1984 to April 1989.

Progress: Generally, ground-water levels in wells intercepting permeable fractures fluctuated seasonally and were lowest from December through June and highest from July through November. Water levels in the disposal trenches fluctuated less than those in wells and for most trenches the fluctuations were less than one-half foot.

From June 1984 to April 1989, tritium concentrations in ground water ranged from 0 to 2,402,200 picocuries per milliliter. The greatest and most variable tritium concentrations were in wells in the northwest section of the site. The major conduit of ground-water flow from the trenches in the northwestern part of the site was a non-homogeneously fractured sandstone bed, which formed the base of most trenches. Elsewhere along the site perimeter,

elevated levels of tritium were not detected in wells, and mean tritium concentrations indicated little change between 1986 and 1988.

Generally, fluctuations in tritium concentrations and specific conductance values were not related. Also, a general relation could not be determined between fluctuations of tritium concentrations and water levels.

The lower sandstone bed ranges in depth from about 5 to 7m below the land surface and formed the bottom for most trenches during burial operations. This bed appears to have more open joints and fractures than the overlying and underlying shale. These joints and fractures are conduits for water moving from the topographically higher burial trenches to downslope observation wells. Where the lower sandstone bed is absent, the surrounding shale tends to impede movement of tritiated water. Some of the migrating tritiated water may move to lower rock units; some water moves to the edge of the plateau and into the colluvium. The water then flows downslope through the shallow subsurface regime and becomes partially transpired (seasonally) through trees and vegetation on surrounding hillslopes. Wet-weather seeps have been observed on surrounding hillsides, and water sampled from seeps below the outcrop of the lower sandstone bed contained elevated tritium concentrations. The seeps generally flow after snowmelt and rainfall and generally cease flowing as plant transpiration increases in the late spring and summer.

Runoff volumes and velocity substantially increased in three main drainage channels after installation of PVC cover over the trenches. This increased runoff volume and velocity has caused accelerated bank enlargement in surrounding channels that drain water away from the site.

About 50 percent of the water drained from the PVC-covered area leaves the site through the main drainage channels. Channel-profile changes for several main drainage channel cross sections were compared with channel-profile changes from a drainage channel unaffected by the PVC cover. Comparisons indicate that the channel dimensions of the main drainage channels continued to adjust in order to reach equilibrium conditions several years after the PVC cover was installed.

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#### Barnwell, South Carolina

The final report on hydrologic investigations at this operating commercial low-level waste site in the coastal plain of South Carolina was summarized in the last review of U. S. Geological Survey research in radioactive waste disposal (Dinwiddie and Trask, 1986). The final published reports on the project are listed below. The site is scheduled to close in 1992.

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Investigations at Low-Level Radioactive Waste Sites on  
U. S. Department of Energy Reservations

During the period 1986-1990, the USGS conducted hydrologic studies and/or research at 7 locations of the U. S. Department of Energy (DOE) connected with weapons fabrication or testing: Idaho National Engineering Laboratory; Hanford Reservation; Nevada Test Site; Oak Ridge Reservation; Savannah River Site; Weldon Spring site; and Palos Forest Preserve, Argonne National Laboratory. The DOE has committed to a 30-year program of environmental restoration, corrective activities, improved waste management operations, and technology development at its nuclear weapons installations across the country (DOE, 1990). The USGS work ranged from monitoring of surface- and ground-water conditions to fundamental studies of water movement and solute transport in the subsurface.

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Idaho National Engineering Laboratory  
By Larry J. Mann, Idaho Falls, Idaho

The Idaho National Engineering Laboratory (INEL) has disposed of waste water containing low-level radioactive and chemical wastes to the environment since 1952. Prior to 1984, waste water was directly injected into the Snake River Plain aquifer through deep disposal wells or was placed in infiltration ponds and percolated down to the aquifer. Since February 1984, infiltration ponds have been used to dispose of most of the waste water. The aquifer consists primarily of a sequence of basaltic lava flows, intercalated with sediment and cinders. The INEL obtains its entire water supply from the aquifer which also is the primary source of water for irrigation, municipal, domestic, commercial fisheries and recreation in other areas. In addition, low-level radioactive, transuranic, and chemical wastes have been disposed to the subsurface at the Radioactive Waste Management Complex (RWMC). The waste is buried in pits and trenches excavated into a veneer of surficial sediment less than 30 feet in thickness. Because of concern about water pollution and the long-term effects of waste-water disposal and migration of leachates from waste buried at the RWMC, detailed knowledge is needed about the geological, hydrological, and geochemical influences of solutes on the unsaturated zone and the aquifer.

Objective: Describe, evaluate, and assess the effects of radioactive- and chemical-waste disposal at the INEL on the ground-water resources. Document distribution patterns of waste constituents in the ground water and evaluate geochemical controls on the migration of wastewater and leachates. Document and evaluate the geologic framework and hydraulic properties of the aquifer and their controls on the movement of ground water and waste constituents contained therein.

Approach: The study of the subsurface effects of waste disposal on the Snake River Plain aquifer compares current conditions with data on previous conditions. Distribution and dispersion patterns, rates of movement, dilution factors, and mass balances of various components are evaluated and compared to those from several time periods. Radiochemical, geochemical, and hydraulic processes affecting changes are also evaluated. Hydraulic dispersion, radioactive decay, and sorption phenomena are being studied by means of digital models. The unsaturated zone underlying the RWMC has been instrumented and studies are being conducted to determine the possibility of solute-transport toward the regional aquifer.

Progress: Studies of distribution and migration of radioactive- and chemical-waste constituents in the Snake River Plain aquifer were continued and consultation was provided to the DOE for a variety of hydrogeologic topics. The geologic framework of the unsaturated zone and Snake River Plain aquifer is being described using geophysical logs from about 300 boreholes and paleomagnetic inclinations and potassium-argon ages of drill core. Aquifer tests have been performed and analyzed for transmissivity at 109 wells in the aquifer and 22 wells in perched ground-water zones that overlie the aquifer. Mineralogy and grain size of surficial sediment along major streams and selected sedimentary interbeds in the unsaturated zone have been described for input to geochemical studies. Analyses of water samples by multiple laboratories were comparatively evaluated to determine precision and accuracy of analytical results. Hydrological, meteorological and geohydrological data for the unsaturated zone near the RWMC were collected and compiled. A long-term ground-water quality monitoring network was established that consists of 70 offsite wells and springs, hydraulically downgradient from the INEL. Background concentrations of radioactive, trace elements, and organic compounds historically used and disposed of at the INEL have been documented.

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## Recharge of Meteoric Water at the Hanford, Washington Site

By Edmund A. Prych, Tacoma, Wash.

As a result of the past storage and disposal of waste products from plutonium production at the U.S. Department of Energy Hanford Site in south-central Washington, a wide range of radioactive, organic, and inorganic wastes have entered and are presently found in the unsaturated sediments at Hanford. A fundamental concern influencing the remediation of these waste sites is that meteoric water draining through the unsaturated sediments may carry contaminants to the underlying unconfined aquifer that discharges to the Columbia River. The quantity of recharge through the unsaturated zone is uncertain because of difficulties in applying conventional, physical, unsaturated-flow characterization techniques to the arid, heterogeneous, Hanford soils.

Characterizing recharge through these contaminated sediments is critical for assessing the environmental risk posed by contaminants and for selecting the appropriate isolation or treatment strategy for remediation.

Objectives: To test the applicability of using a chloride mass-balance method and a chlorine-36 isotopic-tracer method for estimating infiltration of precipitation and natural recharge to the water table of the unconfined aquifer at Hanford. If tests are successful, to apply one or both methods for estimating long-term average recharge at locations on the Hanford Site with different soil, vegetation, and topographic characteristics.

Approach: Collect soil samples from drill-cores and excavated trenches at areas on the Hanford Site with different soil types and vegetative covers. Measure the moisture content and chloride concentration of soil samples from discrete intervals through the soil column. Calculate the mass balance of water and chloride within the soil column and estimate infiltration of meteoric water and recharge below the root zone.

Once the chloride content of the soils is determined, collect a second set of soil samples of an appropriate size to extract chloride and analyze the extract for chlorine-36. Plot the chlorine-36 profile with soil depth, identify the chlorine-36 peak, and calculate the long-term average recharge. Compare recharge rates calculated by each method with recharge estimates derived from Hanford-Site lysimeter facilities. If either method for estimating recharge is successful, apply the method on a waste-site specific basis.

Progress: The project began in the spring of 1990. Soil samples have been collected from eight bore holes and four trenches at three locations.

#### Recharge From Ephemeral Streams at the Hanford, Washington Site

By Richard S. Dinicola, Tacoma, Wash.

For the past 45 years, infiltration of waters from several types of chemical processing has composed the majority of recharge to the unconfined aquifer underlying the U.S. Department of Energy Hanford Site in south-central Washington. This "artificial" recharge has had a major influence on controlling the rate and direction of ground-water flow within the aquifer. As plutonium production is phased out, infiltration of process water will cease and the ground-water flow system will revert to "pre-Hanford" conditions. The infiltration of water from Cold and Dry

Creeks, ephemeral streams draining onto the Hanford Reservation, then will constitute the majority of natural recharge and will exert a substantial influence on the ground-water flow system. Information on recharge from these streams is needed for modeling ground-water flow and contaminant transport through the unconfined aquifer underlying the Hanford Site. Presently, the quantity of discharge from Cold and Dry Creeks is largely unknown; therefore, the quantity of water available for recharge is also unknown.

Objective: To estimate the long-term discharge of the Cold and Dry Creek watersheds and to determine the portion of the discharge that recharges the unconfined aquifer underlying the Hanford Site.

Approach: Select a rainfall-runoff model to simulate streamflow hydrographs of Cold and Dry Creeks. Collect three to four years of field data, including precipitation, streamflow, soil moisture, air temperature, relative humidity, and solar radiation; measure infiltration of streamflow through the stream channels and flood plain; estimate evapotranspiration of infiltrated water; and calculate the remaining volume of water available for recharge. Calibrate the rainfall-runoff model by means of field-measured data, and simulate the long-term discharges of Cold and Dry Creeks and the recharge to the unconfined aquifer by means of long-term Hanford Site climate data as model input.

Progress: The study began in September of 1989. The U.S. Geological Survey's Precipitation Runoff Modelling System (Leavensley and others, 1983) was selected for use in the study. The Cold and Dry Creek study watersheds have been fully instrumented and field data collection activities are underway.

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Nevada Test Site Weapons Testing Program

by William B. Scott, Las Vegas, NV

The Nevada Test Site (NTS), operated by the U.S. Department of Energy (DOE) Nevada Operations Office, functions primarily as a continental site for testing nuclear weapons. Nuclear detonation experiments conducted at the NTS since 1951 have

been both above and below ground. Since 1960, the majority of nuclear weapons tested have been detonated underground. Underground nuclear detonation experiments have and will continue to introduce radionuclides into the subsurface environment. The potential for transport of these source radionuclides and their associated daughter isotopes must be determined to ensure that the public's health and safety are protected and the NTS operations are conducted in full compliance with the Safe Drinking Water; Resource Conservation and Recovery; and Comprehensive Environmental Response Compensation, and Liability acts. A thorough understanding of the movement of water into, through, and out of the ground-water flow system and of the processes which control these movements is essential in assessing the potential for local and offsite migration of radionuclides.

The USGS regional hydrology program for the NTS includes several aspects of the water regime, with a primary focus on determining the potential for movement of radionuclides away from underground event sites. The collection of hydrologic and geohydrologic data at and in the vicinity of the NTS is necessary to aid in developing an improved conceptual understanding of the ground-water flow system. This improved understanding is needed to accurately define the directions and rates of ground-water flow, which aid in evaluating radionuclide migration.

Objective: Characterize the regional ground-water flow system underlying the NTS and vicinity; determine the potential for and extent of radionuclide migration related to nuclear weapons testing; and provide hydrologic support to DOE and its National Laboratories.

Approach: A variety of specially designed studies will be developed and documented to attain the above objectives. A network of selected test holes and wells will be established to collect hydrogeologic data. Data collected will be stored in a computerized data base.

Progress: The USGS, as a participating organization in the DOE Hydrology/Radionuclide Migration Program (HRMP), continued to provide hydrologic expertise and technical support to the Weapons Testing Program at the NTS. Hydrologic data collected from numerous test and emplacement drill-holes were analyzed and documented in several USGS reports. Straddle-packer testing was performed in selected exploratory holes in Yucca Flat and data analyses confirmed the presence of elevated fluid pressures in formations beneath the water table. Further drilling and testing at the NTS will aid in characterizing the extent of the anomalously elevated fluid pressures and in developing methodologies for predicting these occurrences. Hydrogeologic data continued to be compiled, and field data collected and entered into the

USGS-GWSI data base system. An automated Geographic Information System (GIS) for data analysis, management, and presentation was developed with the current focus on well siting and data verification. Future focus of the system will be on hydrogeologic framework development and model grid and input generation.

Hydrologic support continued to be provided to the DOE-Containment Evaluation Panel (CEP). This support included providing hydrologic expertise in the siting of proposed underground nuclear tests, reviewing containment document packages, participating in CEP meetings, evaluating geohydrologic data, and conducting site-specific hydrologic investigations and research at the NTS related to containment of underground nuclear explosions. Radionuclide migration research and monitoring continued at the NTS with technical oversight from the DOE designated Steering Committee. Key categories of focus for the HRMP were defined as source-term identification, transport mechanisms, regional hydrology, and local hydrology.

#### Reports:

Scott, W. B., and Morgan, C. O., 1987, Hydrologic activities of the U.S. Geological Survey in support of the radionuclide migration program, Nevada Test Site, Nye County, Nevada, fiscal year 1986: Special Report (Dept. of Energy-1986 Summary Report for the Radionuclide Migration Project), 10 p.

Hawkins, W. L., Trudeau, D. A., and Mihevc, T. M., 1989, Hydrologic testing in exploratory drill hole UE 4t, Yucca Flat, Nevada Test Site: Proceedings of the 5th Symposium on the Containment of Underground Nuclear Explosions, September 19-21, 1989, Santa Barbara, Calif.

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Savard, C. S., and Morgan, C. O., 1986, Radionuclide migration studies associated with underground nuclear weapon testing, Nevada Test Site, Nevada [abs.]: Nevada Water Conference, Las Vegas, Nevada, February 22, 1986.

Scott, W. B., and Morgan, C. O., 1986, Hydrologic activities of the U.S. Geological Survey in support of the radionuclide migration program, Nevada Test Site, Nye County, Nevada, fiscal year 1986: U.S. Department of Energy Summary Report for 1986.

## Savannah River Plant, South Carolina

By Kevin F. Dennehy,, Columbia, SC, David C. Prowell,  
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The Savannah River Plant (SRP), a DOE facility that produces nuclear materials for national defense, is located along the Savannah River approximately 100 km southwest of Columbia, South Carolina and about 150 km inland from the Atlantic Ocean. It occupies 700 km<sup>2</sup> in parts of Aiken, Barnwell, and Allendale Counties in southwestern South Carolina near the Georgia border.

The Defense Waste Processing Facility (DWPF), located near the geographic center of the SRP, processes for permanent storage high-level radioactive waste that has accumulated over 30 years from the production of defense nuclear materials. The DWPF was constructed adjacent to a chemical separation facility. Waste generated by the separation facility currently is being stored on-site as a concentrated liquid in large carbon-steel tanks. At the DWPF, the waste is further separated into two forms: a high-level radioactive sludge and a concentrated salt solution containing low-level radioactivity.

The sludge component is transported through a pipeline to the DWPF where it is incorporated into borosilicate glass, encapsulated, and stored until a national repository for high-level radioactive waste is available. The concentrated salt solution is transported by separate pipeline to a disposal facility to the north, where it is mixed with pulverized furnace slag and concrete, poured into near-surface vaults, and maintained in permanent storage.

Objectives: To: 1) define the hydrogeologic conditions in the vicinity of the DWPF and, 2) evaluate the potential for movement of a concentrated salt-solution waste if released at or near the DWPF.

Approach: The objectives were accomplished by: 1) assembling and evaluating existing hydrogeologic data, 2) collecting additional geologic, hydrologic, and water-quality data, 3) developing a local geologic framework, 4) developing a conceptual model of the local ground-water flow system, and 5) performing laboratory experiments to determine the mobility of salt-solution waste in surface and near-surface sediments.

Progress: A geologic framework was developed for sedimentary rocks of Tertiary and younger age, including in ascending order the Ellenton, Congaree, and McBean Formations, the Barnwell Formation, the upland unit, and colluvium/alluvium. Results include the identification of a calcareous zone near

the base of the McBean Formation, east of the DWPF. In addition, a green clay within the McBean Formation seems to be continuous over the study area, whereas a tan clay within the Barnwell Formation is absent northeast of the DWPF. The green and tan clays range in thickness from 0.5 to 2 meters and from 0 to 3 meters respectively.

The hydrogeologic framework of the study area consists of two to three separate water-bearing units. In the northern half of the study area, the Barnwell and McBean aquifers are considered to be one aquifer, because of the absence of the tan clay. In contrast, they are separated into two aquifers in the southern half by the tan clay. Underlying these aquifers, and separated from them by the green clay, is the Congaree aquifer. The hydraulic conductivities of the aquifers range from  $10^{-8}$  to  $10^{-4}$  ft/s. Depth to water ranges from 20 m at the center of the study area to 8 m in the southwestern part. Directions of ground-water flow in the Barnwell and McBean aquifers are generally to the north, with a component of flow directed downward across the green clay and into the Congaree aquifer. Part of the flow in the Barnwell and McBean aquifers is intercepted by local streams that downcut into these aquifers. On the basis of limited data, the direction of flow in the Congaree aquifer seems to be to the northwest. At least part of the flow in the Congaree aquifer discharges into Upper Three Runs stream northwest of the study area.

Water samples collected from the Barnwell-McBean and Congaree aquifers indicate that water in these aquifers can be classified as : 1) acidic mixed-cation type (upper Barnwell-McBean); 2) alkaline calcium-bicarbonate type (lower Barnwell-McBean and Congaree); and 3) a water intermediate between 1) and 2) (upper Barnwell-McBean). Chemical changes in water reflect the influence of reactions between sediments and water as water moves from the upper part of the Barnwell-McBean aquifer, to the lower Barnwell-McBean aquifer, and to the Congaree aquifer.

Laboratory column experiments conducted to examine the potential for movement of a simulated salt-solution in sediments from the Barnwell Formation indicate that chemical reactions between the solution and sediment caused sediment permeabilities to decrease 90 to 95 percent. These results suggest that salt solution released to sediments of the Barnwell would move more slowly than would be expected from measurements of hydraulic conductivity of the unit in its natural state.

Report:

Dennehy, K. F., Prowell, D. C., and McMahon, P. B., 1989,  
Reconnaissance hydrogeologic investigation of the

Flow and Transport in Fractured Rock, Palos Forest Preserve,  
Argonne National Laboratory

By Allen M. Shapiro, Reston, VA

Low level radioactive waste was buried during the period 1943 to 1949 on the former grounds of the Argonne National Laboratory in the Palos Forest Preserve about 22 km southwest of Chicago, Illinois. The burial site was decommissioned and capped with concrete in 1956.

Tritiated water has migrated out of the trenches into the underlying 40 m of glacial drift. Concentrations in the drift range from 0.2 to more than 100,000 nCi/L. Tritium is also present in the fractured dolomite beneath the drift in concentrations ranging from 0.2 to 30 nCi/L. A hydrogeologic study of the site with emphasis on locations, concentrations, and migration directions of tritium in the drift and dolomite was completed in 1984. A second study, described here, focused on ground-water flow and tritium transport in the fractured dolomite.

Objective: To investigate fluid movement and chemical transport in discrete fractures in the vicinity of a closed low-level radioactive-waste disposal site originally used by the Argonne National Laboratory. The investigation was conducted approximately 350 m north of the disposal site; it employed seven 0.15 m-diameter boreholes that were drilled into the Silurian dolomite and cased through the overlying glacial drift. The investigation considers fractures in the upper 50 m of the Silurian dolomite. The dolomite is horizontally bedded, and fractures have formed along bedding planes. The bedding-plane fractures are areally continuous over the distances considered in this investigation, approximately 100 m. Aquifer tests indicated that the bedding-plane fractures are highly permeable. Over the scale of the investigation, the dolomite can be conceptualized as a series of horizontally permeable layers separated by a less permeable matrix.

Approach: Aquifer tests were conducted in the dolomite by isolating individual bedding-plane fractures in all seven boreholes with inflatable packers. Pumping was carried out in selected wells and the effects observed in others. In the observation wells, the water level in the packed-off interval was allowed to rise in a standpipe open to the atmosphere.

Radially converging tracer tests were conducted in the bedding-plane fractures by isolating an individual fracture with inflatable packers and pumping from the packed-off interval in one well. After approximate steady-state hydraulic conditions were observed, a tracer solution was injected into the packed-off interval of a second well. The injection was conducted over a short time interval to represent the introduction of a slug of the tracer into the fracture at a given time. The concentration of the tracer was then measured as function of time at the pumped well. Tracer tests were conducted between several pairs of wells at different orientations.

Progress: In the testing of one of the bedding plane fractures, oscillatory water levels were recorded in the observation wells at the onset of pumping. A new well-hydraulics solution was developed to interpret the early-time oscillatory water levels in order to identify the transmissivity and storativity of the fracture (Shapiro, 1989). Analyzing the early-time oscillatory water levels showed that the fracture responded analogously to a dual-porosity medium, indicating that the rock matrix adjacent to fractures yields water or that the bedding-plane fracture cannot be interpreted as an individual, planar fracture but rather as an irregularly weathered section of the dolomite.

In all tracer tests, the breakthrough of the tracer at the pumped well exhibited a rapid rise to a peak value and then a slow decline. The results of the tracer test were first analyzed using an advection-dispersion model. The parameters of the advection-dispersion model were estimated to reproduce the mean and variance of the arrival time of the tracer at the pumped well. However, the breakthrough curves predicted by the advection-dispersion model could not reproduce the rapid rise or the elongated tail of the measured breakthrough curves, suggesting that the mechanisms controlling solute movement in the radially converging flow regime over the distance of these tests (approximately 20 meters) are incongruent with the assumptions implicit in the advection-dispersion model (Shapiro, 1988).

An alternative conceptual model of chemical transport was also used to analyze the tracer tests. In this analysis, an assumption about the spatial heterogeneity of the bedding-plane fracture was imposed. The bedding-plane fracture was assumed to be represented by a distribution of constant aperture channels. It was further assumed that the distribution of apertures is directly responsible for the shape of the breakthrough curve at the pumped well. Two methods of identifying the parameters of the distribution were used. The first method used the measured transmissivity and the measured mean arrival time of the tracer at the

pumped well. The second method used the measured mean and variance of the arrival time of the tracer at the pumped well. If the conceptual model of heterogeneity in the fracture is correct, the two methods should yield the same estimates of the parameters of the distribution of apertures. A lognormal distribution of apertures was employed; however, it could not reproduce the abrupt rising limb of the breakthrough curves. A truncated gamma distribution better reproduced the character of the breakthrough curves, however, the properties of the distribution obtained by the two methods were not the same, indicating that the conceptual model of heterogeneity is significantly more complex (Shapiro and Nicholas, 1989).

#### Reports:

- Shapiro, A. M., 1988, Interpretation of tracer tests conducted in an areally extensive fracture in a northeastern Illinois dolomite, Proceedings of the International Conference on Fluid Flow in Fractured Rock, Georgia State University, Atlanta, GA, May 16-18, 1988.
- Shapiro, A. M., and Nicholas, J. R., 1989, Assessing the validity of the channel model of fracture aperture under field conditions: Water Resources Research, v. 25, no. 5, p. 817-828.
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Oak Ridge National Laboratory, Tennessee

By Patrick Tucci, San Juan, Puerto Rico, D. A. Webster, Nashville, Tenn., and H. H. Zehner, Knoxville, Tenn.

Low-level radioactive waste has been placed in shallow land burial facilities at the U. S. Department of Energy's (DOE) Oak Ridge National Laboratory (ORNL) since 1943. Most of the waste is in three burial grounds in Melton Valley; lesser amounts are in three smaller burial grounds in adjacent Bethel Valley. Both valleys are underlain by a folded, faulted, and jointed complex sequence of mudstone, siltstone, and limestone units of the Conasauga Group of Cambrian age. A weathered zone consisting of regolith up to 12 m thick overlies the bedrock and is the principal unit of ground-water flow.

Drainage of the area is by White Oak Creek and its tributary, Melton Branch. These streams flow into White Oak Lake, a

settling basin that discharges into the Clinch River. Dissolved radionuclides have been found in ground water in the burial grounds and in White Oak Creek and Melton Branch. Principal hydrologic interest regarding radioactive waste disposal are the lower 4.8 km of Melton Valley and the lower 4.0 km of Bethel Valley.

Water quality is monitored by the ORNL in several wells and at White Oak Creek and Melton Branch. In the past, construction of most wells has not been suitable for water-quality sampling; and the collection of stream-discharge data has been sporadic. Knowledge of the ground-water flow system has been insufficient to adequately describe water volume, direction of flow, and flow rate, particularly for the unweathered bedrock.

Objective: To describe the hydrology of the ORNL site in sufficient detail to provide recommendations to the DOE for establishing ground-water and surface-water monitoring systems. The purpose of the monitoring systems is to evaluate water quality in Melton and Bethel Valleys.

Approach: Continuous discharge data are now being collected at two USGS stations installed in 1985 and at three stations installed by ORNL. Head data were collected from well and well clusters constructed by both USGS and ORNL. Hydraulic characteristics of the rocks were determined and computer models describing ground-water flow were developed for Melton and Bethel Valleys. Water-quality samples were collected to provide additional information on ground-water flow and ground-water surface-water relationships.

Progress: The burial grounds for low-level radioactive waste at Oak Ridge National Laboratory, Tennessee, are described by Webster (1990). A conceptual analysis of the ground-water system, particularly of the shallow flow in the burial grounds has been made by Webster (1988). He concluded that: 1) White Oak Creek and its tributaries received all surface run-off and ground-water discharge from each site (including both regolith and bedrock); 2) most ground-water flow is through the regolith and contamination decreases substantially with increasing depth in bedrock; and 3) the burial grounds are a principal source of  $^3\text{H}$  and a major contributor of  $^{90}\text{Sr}$  in White Oak Creek.

Collection of additional data, from coring, well construction, geophysical investigations, and flow measurements resulted in improved definition of head distribution, aquifer characteristics, recharge, discharge, base flow, and water quality in the regolith, bedrock, and streams. Numerical models of ground-water flow (both cross-sectional and areal) provided an improved understanding of the ground-water flow system. The models indicate that 90 to

94 percent of the recharge to the water table flows within the upper 15 m of the ground-water system and from 96 to 98 percent of the flow is within the upper 30 m. Less than one percent of the total ground-water flow circulates to depths greater than 76 m. Radiochemical data also indicate that most of the ground-water flow is in the regolith, although radionuclides have reached depths of at least 58 m within one burial ground. Water quality data indicate that ground water at depths less than 30 m is distinctly of the calcium-magnesium type, and that at depths of greater than 30 m is distinctly of the sodium-bicarbonate type.

#### Reports:

Tucci, Patrick, 1987, Surface geophysical investigations in Melton Valley, Oak Ridge Reservation, Tennessee: U.S. Geological Survey Water Resources Investigations Report 87-4184, 29 p.

Tucci, Patrick, and Hanchar, D. A., 1989, Lithologic, geophysical, and well construction data for observation wells in the Melton Valley area, Oak Ridge Reservation, Tennessee: U.S. Geological Survey, Water Resources Investigations Report 88-4197, 22 p.

Webster, D. A. and Bradley, M. W., 1988, Hydrology of the Melton Valley Radioactive Waste burial grounds at Oak Ridge National Laboratory, Tennessee: U.S. Geological Survey Open-File Report 87-686, 115 p.

Webster, D. A., 1990, The burial grounds at Oak Ridge National Laboratory, Tennessee in Bedinger, M. S. and Stevens P. R., Eds., Safe disposal of radionuclides in low-level radioactive-waste repository sites: Low-level radioactive-waste disposal workshop, U.S. Geological Survey, July 10-16, 1987, Big Bear Lake, California, Proceedings: U.S. Geological Survey Circular 1036, p. 16.

Zehner, H. H., and Tucci, Patrick, (in press), Hydrogeology of Melton Valley near Oak Ridge National Laboratory, Tennessee: U. S. Geological Survey Water Resources Investigations Report.

Bear Creek Valley, Oak Ridge Reservation, Tennessee

by Zelda C. Bailey, San Juan Puerto Rico, and Anne Hoos and Joseph F. Connell, Nashville, Tenn.

Several hazardous waste disposal sites where contaminants are leaking into ground and surface water are present on the upper part of Bear Creek Valley which contains the Y-12

plant, a nuclear production facility. The largest disposal areas are the S-3 ponds, New Hope Pond, Bear Creek burial grounds, and the oil landfarm. Disposed wastes include oils, uranium, solvents, heavy metals, and nitric-acid waste. Bear Creek Valley is underlain primarily by calcareous shale containing limestone units. Pine Ridge, on the north side of the Valley, is underlain by interbedded sandstone, siltstone, and shale of the Rome Formation. Chestnut Ridge, on the South Side, is underlain by massive, siliceous dolomite of the Knox Group and contains karst features. A weathered zone consisting of regolith up to 24 m thick generally overlies the bedrock. The rocks strike north, 56 degrees east. The dip of the rocks varies from 30 to 70 degrees to the southeast; the average dip is about 45 degrees. Many site specific studies of waste disposal areas, contaminated areas, or areas under investigation for future use have been conducted in Bear Creek Valley. The information resulting from these studies, which includes the results from more than 400 wells and test borings, needs to be integrated into an area-wide understanding of the ground-water flow system. The distribution of hydrologic data is not adequate to describe ground-water flow within most of the valley. Also, the general lack of wells greater than 30 m deep precludes adequate understanding of ground-water flow in the bedrock and its connection to the regolith.

Objective: To describe the ground-water flow system in Bear Creek Valley and the potential paths of contaminant transport from hazardous-waste disposal areas.

Approach: Available geohydrologic data were compiled and analyzed to develop a conceptual model of the system and to determine where additional data were needed. Logs from existing boreholes and small-area maps were used to map the geology, thickness of the regolith, and the top of unweathered bedrock. Clusters of wells distributed along the valley boundaries were drilled, tested, and sampled to provide geohydrologic information not previously available. Discharge measurements along the major streams were used to determine the seepage characteristics of the rock units and the interrelations of ground and surface water. Additional information on the interrelations of ground and surface waters and on transport of contaminants from the disposal areas was provided by water-quality analyses of ground water, surface water, and springs. Cross-sectional and areal flow models were used to integrate existing data, to test the conceptual system, to identify additional data needs, to evaluate the flow pattern, and to estimate a water budget.

Progress: Ground-water flow in Bear Creek Valley is primarily from the topographically high ridges toward the main streams on the valley floor (Bailey and Lee, 1991). Water from recharge, primarily on Pine and Chestnut Ridges,

discharges to the Maynardville Limestone and ultimately to streams flowing on the Maynardville. Contaminants reaching the Maynardville could be transported by ground water along the strike of this formation, i.e. to the northeast or out of the valley by streams.

Ground-water flow in the valley is primarily normal to the strike; however, short flow paths along strike (down the Valley) are controlled by closely spaced ephemeral streams that are normal to strike. Ground-water flow along strike is also facilitated by localized zones of intense fracturing and solution cavities. The flow system is generally continuous across geologic formations and at depth. Two zones of distinct water chemistry, from 0 to 30 m and from 30 to 152 m below land surface, were distinguished. Although water in both zones is dominantly of the calcium bicarbonate type, the deeper zone is distinguished by evolution to sodium and bicarbonate dominance along ground-water flow paths. Areas of elevated concentrations of dissolved solids (as much as 15,000 mg/L in the deep zone and 20,000 mg/L in the shallow zone) indicate contamination from waste-disposal sites.

Areal recharge provided all the incoming water to the modeled system and all of the discharge from the system is to the main streams. The most likely area of potential transport of contaminants outside the Oak Ridge Reservation through the ground-water system is from the eastern end of the Y-12 Plant, where ground water in the East Fork Poplar Creek basin may flow into the Scarboro Creek Basin.

#### Reports:

- Bailey, Z. C., 1988, Preliminary evaluation of ground water flow in Bear Creek Valley, the Oak Ridge Reservation, Tennessee: U.S. Geological Survey Water Resources Investigations Report 88-4010, 12 p.
- Bailey, Z. C., and Withington, D. B., 1988, Well construction, lithology, and geophysical logs for boreholes in Bear Creek Valley, near Oak Ridge, Tennessee: U.S. Geological Survey Water-Resources Investigations Report 88-4068, 21 p.
- Bailey, Z. C., and Lee, R. W., 1991, Hydrogeology and geochemistry in Bear Creek and Union Valleys, near Oak Ridge, Tennessee: U.S. Geological Survey Water-Resources Investigations Report 90-2088
- Connell, J. F., and Bailey, Z. C., 1989, Statistical and simulation analysis of hydraulic-conductivity data for Bear Creek and Melton Valleys, Oak Ridge Reservation, Tennessee: U.S. Geological Survey Water-Resources Investigations Report 89-4062, 49 p.

Evaldi, R. D., 1986, Stream flow and specific conductance data for Bear Creak, August 13, 1985, the Oak Ridge Reservation, Tennessee: U.S. Geological Survey Open-File Report 85-682, 8 p.

Hoos, A. B., and Bailey, Z. C., 1986, Reconnaissance of surficial geology, regolith thickness, and configuration of the bedrock surface in Bear Creek and Union Valleys, near Oak Ridge, Tennessee: U.S. Geological Survey Water-Resources Investigations Report 86-4165, 9 p.

### Weldon Spring, Missouri

By M. J. Kleeschulte and J. G. Schumacher, Rolla, Missouri

Two low-level radioactive waste-disposal sites are located in the Weldon Spring area of Missouri. One site consists of the abandoned Weldon Spring Feed Materials Plant (chemical plant) and four disposal pits located on slightly rolling terrain just north of the Mississippi-Missouri River drainage divide. The other site is an abandoned rock quarry in the bluff adjacent to the Missouri River alluvial plain and about four kilometers southwest of the feed materials plant.

The abandoned Weldon Spring Feed Materials Plant is located in St. Charles County, Missouri, about 24 kilometers southwest of the city of St. Charles and about 48 kilometers west of the city of St. Louis. The plant was in operation from 1957 to 1966. During that time, the plant converted impure uranium concentrates to pure uranium salts and metal. Residues from the refining operation were pumped as a slurry to four pits totaling about 25 acres in surface area. Water overlying these residues have much larger concentrations of calcium, sodium, sulfate, fluoride, nitrate, lithium, molybdenum, strontium, vanadium, radium, and uranium than native surface and ground water. Ground water from five monitoring wells adjacent to the pits had nitrate concentrations ranging from 53 to 990 mg/L as nitrogen. Most of the water samples from these wells also had increased concentrations of the following inorganic constituents (the maximum concentrations, in mg/L, are shown in parenthesis): calcium (900), magnesium (320), sodium (340), and sulfate (320). Concentrations of several trace elements also were increased (the maximum concentrations, in micrograms per liter, are shown in parenthesis): lithium (1,700), strontium (1,900), and uranium (86). The presence of these constituents in increased concentrations indicates water from the pits has seeped into the underlying clays that overlie permeable limestone.

Lossing streams and springs are common in the vicinity of the feed materials plant. Uranium concentrations as large as 250

ug/L have been detected in water from Burgermeister spring, which is located about two kilometers north of the pits and in a different drainage basin. Concentrations of nitrate, as nitrogen, as large as 54 mg/L and lithium concentrations as large as 77 mg/L also have been detected in the water from Burgermeister spring. Dye tracing has shown the losing streams north of the feed materials plant are hydrologically connected to Burgermeister spring. Because no contamination plume has been detected between the pits and Burgermeister spring, it appears that the contaminants are migrating through highly localized paths in the ground-water system, such as fractures and solution openings.

The Weldon Spring quarry, about 4 km southwest of the chemical plant, became a low-level radioactive waste-disposal site in 1959. Water sampled from 13 wells near the quarry and north of Femme Osage slough in 1986 had uranium concentrations ranging from 8.9 to 14,000 ug/L. Water from six observation wells sampled south of the slough had uranium concentrations of less than 5 ug/L. Water samples collected from the St. Charles County well field, 1.3 kilometers southeast of the quarry, indicate the water in the well field has background concentrations of uranium (less than 2.1 ug/L).

Objective: In 1987, the U.S. Geological Survey began a new phase of the work in cooperation with the DOE at the feed materials plant. The new work consists of two separate projects: one project describes the hydrogeology of the area, the other addresses the geochemistry of the area. The primary purpose of the first project is to better define the extent and magnitude of contamination of the water resources in the vicinity of the feed materials plant. Other objectives include determination of affects of climate on surface- and ground-water systems and preparation of a ground-water flow model that includes most of St. Charles County. The primary objective of the second project is to describe the geochemical controls on contaminant migration from the pits at the feed materials plant.

Approach: The extent and magnitude of contamination of the water resources in the vicinity of the feed materials plant will be determined by sampling streams, springs, and wells that surround the feed materials plant. The investigations will be aided by the use of potentiometric maps drawn from water levels measured in the monitoring wells at the site and on nearby property.

The effects of climatic conditions on surface- and ground-water systems will be determined from reviewing both quarterly potentiometric maps drawn for the area and the record from six continuous water-level recorders installed in monitoring wells on site. Emphasis for the data obtained

from the potentiometric maps will be on the response of water levels during different climatic conditions throughout the year. The continuous water-level records will be reviewed to determine what effect precipitation events have on water levels.

A ground-water flow model is being developed that includes most of St. Charles County and parts of adjacent counties. The three-dimensional finite-difference ground-water flow model will be used to determine the flow characteristics of the geohydrologic units of the area. The quantity of water that infiltrates into the deep aquifer from the shallow aquifer is of special interest.

The geochemical controls on contaminant migration from the pits will be determined using the results of laboratory sorption experiments between a simulated leachate and various overburden units. Mineralogical analyses will be performed on selected overburden samples to demonstrate the effects of overburden mineralogy on laboratory results and contaminant distributions in the shallow ground water. Interpretation of ground-water geochemistry and contaminant migration will be aided by the use of geochemical speciation-equilibrium and reaction-path models.

Progress: The needed data for both projects have been collected and are reported in the publications listed below.

Reports:

- Kleeschulte, M. J., Emmett, L. F., and Barks, J. H., 1986, Hydrologic data for the Weldon Spring radioactive waste-disposal sites, St. Charles County, Missouri--1984-1986: U.S. Geological Survey Open-File Report 86-488, 61 p.
- Kleeschulte, M. J. and Emmett, L. F., 1987, Hydrology and water quality at the Weldon Spring radioactive waste-disposal sites, St. Charles County, Missouri: U.S. Geological Survey Water-Resources Investigations Report 87-4169, 65 p.
- Kleeschulte, M. J., and Cross, P. W., 1990, Hydrologic data for the Weldon Spring chemical plant site and vicinity property, St. Charles County, Missouri, 1986--1989: U.S. Geological Survey Open-File Report 90-552
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Studies of Processes Related to the Disposal of Low-Level  
Radioactive Waste

Processes Controlling Transfer of Carbon-14 at Radioactive  
Waste Disposal Sites

By Robert G. Striegl, Denver, CO

Carbon-14 is a persistent radionuclide (half-life 5,730 years) that is included in a variety of waste forms of low-level and high-level radioactive waste. Disposal of  $^{14}\text{C}$  is of environmental concern because it readily exchanges among solid, aqueous, and gaseous phases; it is highly mobile in aqueous and gaseous phases; and it is readily assimilated by living organisms.

Objectives: a) To estimate from current theory, the chemical form and the concentration distribution of  $^{14}\text{C}$  in the unsaturated zone near buried, low-level radioactive waste; b) to characterize that distribution by in-situ measurement at existing waste-disposal facilities; and c) to compare theoretical and measured distributions in order to increase the understanding of the transfer of carbon isotopes among components of the unsaturated zone, the overlying atmosphere, and the biosphere.

Approach: The distribution of  $^{14}\text{C}$  is measured among aqueous and gaseous species in the unsaturated zone and in plants growing at low-level waste sites. These distributions give insight to mechanisms that can potentially retard or enhance  $^{14}\text{C}$  transport. The influence of those mechanisms on  $^{14}\text{C}$  transport are further isolated by laboratory investigation and numerical modeling.

Progress: The concentration distribution of  $^{14}\text{CO}_2$  and  $\text{CH}_4$  was determined at the low-level radioactive waste-disposal site near Sheffield, Illinois. Subsequent laboratory investigation and numerical modeling demonstrated that movement of  $^{14}\text{CO}_2$  in the unsaturated zone was controlled by coupled  $\text{CO}_2$  adsorption and isotopic exchange, and that movement of  $\text{CH}_4$  was controlled by methylotrophy in the root zone. Other field investigations are in progress at the West Valley, New York and the Nevada Test sites. Those investigations focus on processes that control fluxes of  $^{14}\text{CO}_2$  and  $\text{CH}_4$  through trench caps, to the atmosphere, and on the uptake of  $^{14}\text{C}$  by local vegetation.

Reports:

Striegl, R. G., 1988, Distribution of gases in the unsaturated zone at a low-level, radioactive-waste

disposal site near Sheffield, Illinois: U.S. Geological Survey Water-Resources Investigations Report 88-4025, 69 p.

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## Microbial Reduction of Radioactive Waste Metals

by Derek R. Lovley, Reston, Va

Previous studies have identified microorganisms which can effectively reduce Fe(III) to Fe(II) and Mn(IV) to Mn(II). Many radioactive waste metals, such as uranium, technetium-99, chromium-51, manganese-54, ruthenium-103, and -106, and selenium-79, have multiple oxidation states. It is likely that microorganisms may also be able to reduce these radioactive contaminants. The reduction of these radioactive contaminants will greatly affect their solubility and adsorption, and hence, mobility. For example in its oxidized state uranium [U(VI)] is soluble; but in its reduced state [U(IV)], it is only slightly soluble.

Objective: To determine the ability of microorganisms to reduce various radioactive metals and, for those metals that can be reduced, to determine the mechanisms for reduction and the factors controlling the rate and extent of metal reduction.

Approach: The ability of various types of microorganisms to reduce radioactive metals under laboratory conditions will be evaluated. The enzymatic process responsible for metal reduction will be determined. Once the process is understood, microbial metal reduction will be studied in natural environments.

Progress: The Fe(III)-reducing microorganism, GS-15, was found to be able to reduce U(VI) to U(IV) (Lovley and others, 1991). GS-15 obtains energy to support growth by coupling

the oxidation of organic compounds to the reduction of U(VI). U(VI) reduction is an enzymatic reaction that may be similar to Fe(III) reduction. Enzymatic reduction of U(VI) was found to be much more rapid and extensive than commonly cited abiological mechanisms for U(VI) reduction, such as reduction by sulfide, hydrogen, or organic compounds. Not only do these findings expand the known potential terminal electron acceptors for microbial energy transduction, they offer a likely explanation for the deposition of uranium in aquatic sediments and aquifers. Furthermore, the results suggest that it may be possible to immobilize uranium in contaminated waters by stimulating microbial U(VI) reduction. Other radioactive metals could potentially be immobilized in a similar manner.

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### Water Flux in the Unsaturated Zone of Deserts

By John R. Nimmo, David A. Stonestrom, and Jacob Rubin, Menlo Park, CA

The unsaturated zone in deserts is one of the barriers to the movement of radioactive substances which could be used to design and site repositories for radioactive wastes. A major element in predicting the effectiveness of such a barrier is the estimation, for a specific location, of prevailing, long-term water fluxes at considerable depths. Field data on fluxes of this type are extremely scarce. Understanding of these fluxes is incomplete. To improve such understanding and predictive capability, the applicability of unsaturated flow theory to the relevant conditions must be established and the influence of additional factors, including gas fluxes, gas trapping, and temperature fluctuations, must be understood.

Soil-water conditions in the uppermost (root zone) layers of unsaturated zones are known to fluctuate widely with time. Theoretical considerations show that the effects of these fluctuations are transmitted downward, but are rapidly attenuated with depth. As a result, at sufficient depths, nearly steady water fluxes may exist in unsaturated zones. Such fluxes reflect long-term trends. By studying profiles of selected water properties at appropriate depths in the unsaturated zone, it should be possible to find indications of whether an approximate steady state exists and to measure the extant water fluxes. Knowledge of such fluxes should improve understanding of the factors that control them, and

may allow identification of surrogate indicators of these fluxes. In addition, it may be possible to relate the observed fluxes to other long-term trends, such as climatological changes.

Objective: To develop methods for measuring unsaturated-zone water and gas fluxes and to assess the validity of theoretical formulations relating to these fluxes.

Approach: Water conditions in the unsaturated zone will be studied. To measure water fluxes, particular attention is paid to sampling depths at which fluxes are apt to reflect long-term trends. Using pressure-head and hydraulic-conductivity data, existing water fluxes will be computed from which estimates of long-term flux trends will be extracted. These estimates will be compared with analyses based on isotopic data and with simulation-based predictions.

The fundamental nature of water flow and the effects on it of gas flow, gas trapping, and temperature fluctuations will be investigated by developing and implementing specially designed laboratory apparatus capable of controlling and measuring conditions, as required. The experimental data will be used in critical testing of current theories as well as in developing new predictive theories.

Progress: To evaluate long-term average water fluxes in areally extensive unsaturated zones of arid regions, a new, rapid, steady-flow method for determining hydraulic conductivities of unsaturated sediment cores at low water content has been developed. This method uses centrifugation at forces up to 2000 times normal gravity to quickly establish steady-state flow, permitting measurements with up to +/-8 percent accuracy. The method has been extensively tested with packed samples of a sandy soil, showing good agreement with available data (at high water contents) from alternate, more time-consuming methods. New developments in the means of regulating flow during centrifugation allow for fine tuning of the hydraulic conductivity (K value) measured and have reduced the value of K that can be attained by the steady-state method to unprecedented levels: down to  $1.1 \times 10^{-11}$  m/s at 21 percent saturation in a sandy soil. Additional developments demonstrated the validity of analogous steady-state and falling-head centrifuge methods for saturated media; the results agreed with measurements by the standard, gravity-driven, falling-head method.

Because centrifuge methods necessarily induce some degree of artificial soil compaction, experiments have been done to assess the effect of this compaction on unsaturated K measurements. For packed samples of a sandy soil at saturations between 24 and 36 percent, the results show a negligible influence of degree of centrifuge compaction on

both K and water retention, although as much as a five-fold variation resulted from differences in the pre-centrifugation, soil-packing procedure. The measured differences in K agreed with capillary-theory based predictions based on water-retention measurements. Similar experiments are being conducted to assess the effects of compaction on the moisture properties of fine-textured soils.

Measurements of unsaturated K by the steady-state centrifuge method on core samples obtained from a sandy sediment at 8 m depth, at a site in the San Joaquin Valley, showed the method to be well suited for these field cores.

Extensive tests of the field techniques were conducted at two topographically distinct, but otherwise similar, settings in a thick, unsaturated, loessal deposit in southwest Washington State. Water-content profiles were obtained using traditional sampling techniques near the end of the rainy season and near the end of the dry season. Supplemental water-content determinations were made using a newly developed, down-bore technique based on time-domain reflectometry. Matric-pressure measurements were made using a specially designed bore-bottom tensiometer. In addition, tritium concentrations were determined on water extracted from unsaturated-zone samples. Results indicated that seasonal fluctuations are attenuated below depths of 4 to 5 m in both topographic settings, and that nearly steady-state conditions exist below these depths. Tritium-concentration profiles gave preliminary flux estimates of 10 cm/y and 16 cm/y at the two settings. These distinct flux estimates corresponded to large contrasts in soil-profile development and water-content profiles. Such a result is encouraging, suggesting that these properties may serve as surrogate indicators of deep, unsaturated fluxes.

A study of the temperature dependence of hysteretic soil-water retention showed the temperature sensitivity to be as much as eight times greater than predictions based on the temperature dependence of the surface tension of pure water. The magnitude of the discrepancy increased with soil dryness. Drying and wetting curves showed equal temperature sensitivity.

Tests of steady-state, unsaturated flow theory strongly supported Darcy's law for a sandy soil at 27 percent saturation over a range of forces from 216 to 1650 times normal gravity. Tests of transient-flow theory showed agreement within a factor of four or better between measurements and predictions based on Richards' equation for a sandy soil between 25 and 36 percent of saturation. These results suggest that the generally accepted theory of unsaturated flow is applicable under much drier conditions than those for which it has previously been tested. This support for existing theory is encouraging for attempts to

model radioactive-waste transport under dry conditions. The results also contribute evidence challenging the validity of the concept of residual water content (below which K is zero), as used in certain popular models of soil-moisture characteristics. This issue is critical to the quantitative prediction of water movement in arid-region repositories.

To improve understanding of unsaturated-zone gas transport relevant to nuclear-waste disposal, a two-phase permeameter was developed. This device permitted measurements of gas and water permeabilities, gas slip, gas trapping, and critical gas saturations (saturations above which gas permeabilities are negligible) under conditions of matric-pressure controlled wetting and drying. In addition, a small-step, implicit-inversion method for determining the saturation dependence of water permeability was developed and successfully used for a range of intermediate water contents. This method will be used to check some of the centrifuge-method measurements on field cores. Gas-permeability results demonstrated an intimate association of the critical gas saturation level with the appearance (on wetting) and disappearance (on drying) of detectable quantities of trapped gas. Results also showed that the development of hysteresis in the relation between gas permeability and fluid saturation does not require measurable quantities of trapped gas. Measurements of gas slip in various unsaturated soils indicated that corrections for gas slip in current use substantially underestimate its magnitude (by up to 300 percent). These results have important implications for predicting the movement of any unsaturated-zone gas, including radioisotope-containing gases associated with disposal of spent fuel rods.

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### Solute Transport in the Unsaturated Zone

by Jacob Rubin, R. V. James, J. M. Bahr and C. Willis,  
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If the unsaturated zone is to be considered a host medium for the emplacement of radioactive wastes, the medium must be an effective barrier to radionuclide transport. The design of a suitable repository depends on our capability to predict the effectiveness of this medium as a barrier. Predictive models have been proposed, and others are being developed for solute transport in unsaturated, porous media. The usefulness of these models is limited, however, by the difficulty of analyzing and predicting solute transport influenced by large reaction-networks. Also, there is lack of information relating solute-transport mixing parameters and reaction parameters to the water content of the medium.

Objective: To develop methods for analyzing and predicting solute transport in porous media when large, transport-influencing reaction-networks are present; and to acquire, for unsaturated porous media, the necessary data on (and understanding of) the dependence of physical and chemical, transport-relevant properties on the water content of the medium.

Approach: The first step will be to choose the most appropriate formulation of the basic transport equations and transform these equations so they are computationally more efficient. To understand the influence of water-content on the transport properties of unsaturated porous media, the project will conduct laboratory, soil column experiments under uniform water-contents and develop theoretical explanations of the results obtained. These results will be formulated in a computationally useful form.

Progress: Water-flow in unsaturated zones, especially in deep unsaturated zones, is typically very slow. When water flow is slow, modelling of reaction-influenced solute transport may often (but not always) be simplified by using the local-equilibrium assumption (LEA). Procedures for determining when LEA can be used were published (Bahr, 1986; Bahr and Rubin, 1987).

A new method for mathematically transforming the ordinarily used, LEA-based transport equations into especially efficient operational equations has been developed and later called the Feed-Forward (FF) method. Each one of the three main reaction-classes was shown to require a different kind of transformation. Nevertheless, these transformations can be used when networks consist of reactions which do not belong to a single class. Ordinarily, the LEA-based transport equations are a simultaneous set, which consists of two parts - a partial-differential-equations (PDEs) part and an algebraic equations (AEs) part. The transformations remove the simultaneity of these two parts, while avoiding or at least minimizing iteration between them. The transformations also remove the simultaneity of all or at least part of the PDEs. Finally, they make transport modelling more algebraic by replacing a significant part (or, occasionally, all) of the PDEs with AEs. These changes are responsible for the considerable efficiency of the FF-method-based algorithms. These results are presented in a recent paper, treating a large variety of large networks, which, however, consist only of simple reactions (Rubin, 1990).

When LEA is used, a mathematically difficult, moving boundary problem may arise. Three different ways of solving this problem were found for cases in which the moving boundary is created by dissolution of solid-phase reactants. Another

computational study demonstrated the applicability of the FF-method to a two-dimensional transport problem which involves a relatively small network of reactions that belong to two different classes.

Transport of chloride ion and of tritium was studied by means of steady-water-flux columns of a rather sandy soil. Each column was at a different, unchanging water-content (41 to 100 percent saturation). It was shown that transport of chloride in this soil (3 percent clay, 0.3 percent organic carbon) could not be understood without taking into account anion exclusion. Once the anion exclusion was accounted for by previously developed theory, the agreement between transport theory and experimental results was very good. There was no need for assuming existence of immobile water. For the soil studied, the dispersivity of saturated soil was three times higher than that of the unsaturated soils (at saturations not exceeding 55 percent).

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## Fluid Flow in Fractured Rocks

By P. A. Hsieh, Menlo Park, CA

Attempts to predict the movement of fluid in a low-permeability, fractured rock mass will depend on the ability to 1) mathematically describe the physical laws that govern the movement of fluids in such an environment, and 2) quantitatively characterize the porosity and permeability of the rock mass and the flow system by meaningful and measurable quantities.

Objective: To develop field methods and modeling techniques for characterizing the flow and transport properties of fractured rocks, and to assess the usefulness of these techniques by applying them to various field sites.

Approach: A number of theoretical approaches have been proposed to model the movement of fluids and solute in fractured rocks. The discrete approach attempts to characterize fluid flow in individual fractures. The continuum approach characterizes the rock mass as an equivalent porous medium. A third approach, known as the stochastic-continuum approach, attempts to represent the spatial variations in rock properties by random functions, based on geostatistical concepts. The usefulness of these approaches, however, remains unclear. There is a need to test these approaches at various field sites in order to assess their strengths and weaknesses. A major effort of this work is devoted to this purpose. A research field site is being established at the Mirror Lake watershed, near West Thornton, New Hampshire. Theoretical approaches mentioned above will be used to model the movement of tracers introduced into the crystalline bedrock. Comparisons between the theoretical predictions and the experimental results should provide insight into the relative merits of the various approaches and the extent of their data requirements.

Hydraulic and tracer test methods will be developed for determining flow and transport properties of fractured rocks. Traditional methods, developed in ground-water hydrology, are often not applicable due to the low-permeability and highly heterogeneous nature of fractured rocks. Methods for

analyzing hydraulic tests will account for non-radial flow, and presence of zones of contrasting permeabilities (e.g., resulting from the presence of a highly fractured zone in an otherwise tight bedrock). Other methods, based on analysis of water-level fluctuations due to natural forces, such as earth tides and barometric pressure fluctuations, are also being investigated.

Progress: A method was developed for determining aquifer properties from water-level fluctuations due to earth tides (Hsieh and others, 1987, 1988). This method is particularly suitable for low permeability aquifers, such as fractured rock aquifers, for which pumping tests are often not feasible. More recent developments have extended this work to include the stochastic forcing effects of barometric pressure fluctuations (Ritzi and others, in press).

A new hydraulic test was developed to determine the hydraulic properties of a bedrock aquifer and its interconnection with an overlying layer of regolith. This method was applied to investigate the bedrock aquifer underlying Lee Valley, San Diego County, California. Test results show that the bedrock and regolith are hydraulically connected, forming an interactive ground-water flow system. Although the bedrock may be highly permeable in places, most of the ground water is stored within the regolith. Therefore, protection of shallow ground water is critical to prevent contamination of water resources in bedrock terranes. Details of the study are given in Kaehler and Hsieh (in press).

Field investigations have commenced at the research site at the Mirror Lake watershed. Work at this site is conducted in cooperation with other scientists within and outside the Water Resources Division. Activities to date have been directed toward obtaining background information, and include well drilling, geophysical logging, hydraulic testing, tracer testing, geological mapping, geochemical sampling, and installing long-term monitoring equipment. Results from these investigations form the basis for designing future experiments on solute transport in fractured rocks.

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### Radionuclides in Ground Water

by Thomas F. Kraemer, Reston, VA

Naturally occurring radionuclides in the uranium and thorium decay series occur virtually everywhere--in sediments, rocks, minerals, water, and even gas. The research in this project is aimed at developing a clear understanding of the environmental chemistry of the more long-lived members of these decay chains (U-238, U-234, Th-232, Th-230, Th-228, Ra-228, Ra-226, Ra-224, Rn-222), how the distribution of these nuclides has changed within the environment as a result of human activity, and whether these nuclides can be used as tracers of hydrologic processes.

Objective: The objective of this project is to examine the behavior of naturally occurring uranium and thorium series nuclides in the laboratory and in the field, and to identify processes which lead to their redistribution within the environment, as well as to develop techniques using these radioisotopes that will allow their use as tracers of hydrologic processes.

Approach: Basic data, such as solubility and adsorption behavior of the nuclides, is acquired in the laboratory under controlled conditions so that a clear knowledge of the chemistry of these nuclides is achieved. Along with these studies, field investigations into the environmental behavior of the nuclides are carried out to gain understanding of the reactions of the nuclides involved in various natural processes. Data from these two types of investigations are useful in such studies as detecting cross-formational flow in aquifers, revealing water masses of differing ages in lakes, and using radioisotopes to identify and distinguish between different water sources in surface and ground-water systems.

Progress: Analyses of uranium concentrations in brines from deeply buried, geopressured aquifers in the United States

Gulf Coast region showed that in virtually every case the water is at or close to saturation with respect to the uranium minerals coffinite or uraninite, and that the uranium activity ratio (U-234/U-238) of the water is apparently controlled by a small amount of these minerals in the aquifer's solid phase. For this reason, uranium activity ratios are not commonly found in such environments.

The discharge of an industrial waste rich in radium into the Mississippi River was investigated and the behavior of the radium upon introduction to the river was examined. It was found that the discharge of the waste doubled the Ra-226 content of the river and that the sediment and water in the river reequilibrated chemically and isotopically with the waste very quickly (within 3 hours). Also developed in this study was a method for accurately estimating the total flux of radium from a river to the ocean.

An investigation of radon in natural gas from wells located in the United States Gulf Coast region revealed that radon is higher in natural gas extracted from brine solution (unconventional natural gas) than in natural gas extracted from gas-cap reservoirs (conventional natural gas). It was noted that although the radon content of the unconventional gas was high, it was well within the range of values found for conventional gases produced throughout the United States, and that no serious health threats were likely to occur as a result of its use.

Uranium concentration and isotopic data have been collected on the Mississippi River at St. Francisville, Louisiana for several years (1988-present). The results show a seasonal variation in dissolved uranium in the Mississippi River from about 0.5 ug/L at the lowest value to more than 2.0 ug/L at the highest value. This variability seems to correlate quite well with the relative flows of the Ohio and Missouri rivers, which are relatively low (~0.5 ug/L) and relatively high (~4 ug/L) in uranium, respectively. Uranium and radium isotopic data of rivers within the Mississippi-Ohio river drainage basin show variations that can be explained on the basis of regional differences in the geology of the drained areas.

A study was conducted to explain the source of radium-rich water in the Charlotte Harbor, Florida, estuary. Results of the study indicated that radium-rich ground water underlying the area is migrating upward into the estuary and surrounding land areas, causing elevated radium concentrations, which are too low, however, to cause environmental or health problems.

Laboratory studies of the solubility of radon in water and organic liquids is underway. Results so far have confirmed earlier work on radon solubility in distilled water and sodium chloride solutions and have corroborated measured

radon solubilities in petroleum and various organic liquids at various temperatures. It is planned that eventually a comprehensive data base will be established to enable modelers to predict the movements of radon in gas-liquid-solid systems more precisely.

Studies of radium isotopes in lakes have established that there are temporal and areal variations of these isotopes within lakes which can be used to study circulation patterns and seasonal overturns. This is important in that in many instances no other natural substances are known which can be used to study lake dynamics. It may also be possible to use these isotopes to determine the residence time of the water in the lake basin through the use of the decay of Ra-228.

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#### Application of Surface-Geophysical Methods at Existing and Future Low-Level Radioactive Waste Sites

By F. P. Haeni, Hartford, Conn.

Geophysical methods consist of many individual techniques that measure differences in physical properties of subsurface

units and the fluids in them. Surface-geophysical methods can potentially be used to map geohydrologic boundaries, characterize the composition of geologic material, locate individual fractures or fracture zones in consolidated rock, and detect contaminated ground water. This information is useful in investigating contamination problems at existing low-level radioactive-waste sites or defining the hydrologic setting at future sites.

Objective: (a) to assess the capabilities and limitations of existing surface-geophysical methods in determining the hydrologic characteristics and physical properties of subsurface units; and (b) to modify existing data acquisition and interpretation procedures in order to improve the hydrologic data that can be obtained from surface-geophysical methods.

Approach: Identify surface-geophysical methods that have the potential for identifying important hydrologic characteristics of subsurface units. Conduct literature searches, modeling studies, and field experiments in order to assess the potential of these methods. Develop improved data acquisition techniques and/or interpretation procedures for these methods. Research will be technique oriented for specific methods, or problem oriented where several methods are used to conjunctively describe the subsurface.

Progress: Forward-modeling programs for the interpretation of Frequency-Domain and Very-Low-Frequency (VLF) Electromagnetic methods were developed for use in personal computers. These programs were then used to quantitatively interpret water-quality data in a contamination plume coming from a landfill and to determine the hydrologic boundaries in an aquifer system.

An annotated bibliography on the use of surface-geophysical methods to detect fractures in bedrock was compiled. Field experiments using the most promising methods were conducted at two field sites with crystalline bedrock. Multiple data sets consisting of azimuthal-DC-resistivity, ground-probing radar, seismic-compressional-wave velocity, and frequency domain electromagnetic methods were tested in the field, and conjunctively interpreted. These geophysical interpretations were compared with results from geologic mapping, hydrologic testing, and borehole geophysical studies. A controlled research experiment has begun at Mirror Lake, New Hampshire, another crystalline bedrock site, with the collection of azimuthal-DC-resistivity, very-low-frequency electromagnetic data from fixed VLF and portable VLF stations, and frequency-domain electromagnetic data.

Ground-probing radar methods have been used to map the hydrologic boundaries of aquifer units and a modified seismic

facies/reflection character interpretation method has been developed to interpret these records. Detailed experiments were conducted wherein radar records were compared with logs from continuously cored, unconsolidated, stratified-drift deposits. Experiments are presently being conducted at a Cape Cod research site where a three-dimensional, radar data set was collected. Attempts will be made to display the data set and to extract physical property data from it.

Continuous-seismic-reflection-profiling methods have been used to map unconsolidated aquifer systems underlying streams, lakes, and ponds. A field data collection method was developed for obtaining high quality records at several sites in New England. A field experiment was conducted on the Clinch River, bordering the Oak Ridge National Laboratory in Tennessee, in order to define bedding planes, fractures or fracture sets, and other structural features in the consolidated bedrock units underlying the river.

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## URANIUM MILL TAILINGS

### Radium Geochemistry

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Radium appears to be the most important nongaseous radioactive contaminant associated with uranium mill tailings. Therefore, research is needed to focus on its chemical state in the tailings, and its potential mobilization and retention in natural aqueous systems. In order to investigate the behavior of radium in comparison with other radioactive and toxic elements, and in association with a range of earth materials, the studies underway are addressing (1) uranium, thorium, and selected stable elements, as well as radium associated with tailings, and (2) nontailings materials including uranium ore, sediments receiving radium-rich brines, and residues from the radium extraction industries of the early 20th century, as well as uranium mill tailings.

Objective: To study the chemical form in which radium, its parent radionuclides, and selected stable elements are retained in surficial earth materials, particularly uranium mill tailings, and to identify processes operating in natural aqueous systems that may influence the transport of radium and other elements from these earth materials.

Approach: Selective extraction techniques and measurement of sorption/desorption properties of radionuclides in uranium mill tailings will be carried out. Microorganisms, which have the potential to solubilize radionuclides from tailings, or otherwise influence the environmental mobility of radionuclides in the hydrosphere, will be studied.

Progress: 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. 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 currently estimated to be about 200 million tons. 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.

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 relation 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 less 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 the 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.

The solubilization and subsequent resorption of radionuclides by ore components or by reaction products during the milling of uranium ores may have both economic and environmental consequences. Particle-size redistribution of radium during milling has been demonstrated by previous investigators; however, the identification of sorbing components in the tailings has received little experimental attention. A study was done in which uranium-bearing sandstone ore was milled, on a laboratory scale, with sulfuric acid. At regular intervals, filtrate from this suspension was placed in contact with mixtures of quartz sand and various potential sorbents that occur as gangue in uranium ores; the potential sorbents included clay minerals, iron and aluminum oxides, feldspar, fluorspar, barite, jarosite, coal, and volcanic glass. After equilibration, the quartz sand-sorbent mixtures were separated from the filtrate and radioassayed by gamma-spectrometry to determine the quantities of uranium-238, thorium-230, radium-226, lead-210 sorbed, and the radon emanation coefficients. Sorption of uranium-238 was low in all cases, with maximal sorptions of 1-2 percent by the bentonite- and coal-bearing samples. Thorium-230 sorption also was generally less than 1 percent; maximal sorption here was observed in the fluorspar-bearing sample and appears to be associated with the formation of gypsum during milling. Radium-226 and lead-210 generally showed higher sorption than the other nuclides; more than 60 percent of the radium-226 solubilized from the ore was sorbed on the barite-bearing sample. The mechanism(s) for this sorption by a wide variety of substrates is not yet understood. Radon emanation coefficients of the samples ranged from about 5 to 30

percent, with the coal-bearing samples clearly demonstrating an emanating power greater than any of the other materials.

The emanating radium contents of background and contaminated soils were measured by means of commercial alpha-track detectors, sealed with samples in glass Mason jars for a 179-day period. The observed track densities were linearly correlated with independently measured emanating radium contents derived from gamma assays for total radium and radon emanation measurements. This simple jar test exhibits high sensitivity and requires minimal equipment and user's training. It has applications in indoor radon and building material studies, as well as in geochemical exploration.

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