MASTER

RESEARCH SUMMARY: CHARACTERIZATION OF
RADIONUCLIDE AND MOISTURE MOVEMENT
THROUGH ARID REGION SEDIMENTS

G. W. Gee
T. L. Jones
D. Rai

September 1981

Presented at the
Annual Meeting of the
IAEA Coordinated Research
Group on Migration of
Radionuclides in the
Terrestrial Environment

Work Supported by
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

now

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

RESEARCH SUMMARY FOR

International Atomic Energy Coordinated Research Program: Migration and Dispersion of Radioactive Wastes in Terrestrial Environments

Subject: Research Agreement 2444/CF: Characterization of Radionuclide and Moisture Movement Through Arid Region Sediments.

Authors: G. W. Gee, T. L. Jones, and D. Rai - Battelle Pacific Northwest Laboratory, USA.

Introduction

This project has the task of understanding the movement of moisture and radionuclides under arid region conditions. This understanding will be used to maximize the isolation of low level waste from the environment. Specific objectives include:

1. Field monitoring of moisture and radionuclide transport at an arid region site.
2. Assessment of the interaction of radionuclides with unsaturated soils in arid regions.
3. Evaluation of radionuclide transport in unsaturated soils by appropriate mathematical models.
4. Assessment of the importance of upward migration of radionuclides by evaporation and diffusion processes.

Field Test Facility

The Burial Waste Test Facility (BWTF) located near Richland, Washington on the Department of Energy (DOE) Hanford Site has been monitored for water content and radionuclide transport for the past two years. Tritium movement has been observed to depths of 7.6 m in both irrigated and nonirrigated lysimeters. Figure 1 shows a plan view of the test facility. Figure 2 shows the tritium profiles observed after initial application in October 1979. Rainfall patterns differ greatly from year to year as indicated by Figures 3. Observed water flow indicates that seasonal rains can infiltrate to significant depths in the sandy soil even under low annual precipitation. When moderate

FIGURE 1. Burial Waste Test Facility at Hanford, Washington

FIGURE 2. Tritium Concentrations as a Function of Time in Field Lysimeters at Hanford, Washington

FIGURE 3. Water Balance and Climatic Data at Hanford Washington Lysimeter Site.

amounts of irrigation are applied, leaching is rapid and deep in the coarse textured Hanford soils at the field test site. Work is continuing in analyzing weather data and soil moisture profiles in order to determine water balance and simulate the tritium and cobalt-60 movement and to estimate evaporation and upward flow of radionuclides.

Laboratory Analysis

Seepage Tests. Laboratory tests were conducted to determine how leachate from uranium mill tailings interacts with geologic materials. Acid leach tailings and tailings solution and geologic materials typical of mill site tailings pits, were physically and chemically characterized. Investigation was made of the sorption characteristics of heavy metals and radionuclides on the geologic materials under low and neutral pH conditions. Tests of clay liner stability were designed to evaluate the mechanisms by which a clay liner is altered under acid attack, and to assess any long-term changes in the permeability of the clay liner materials. Under low pH conditions, most of the radionuclides and heavy metals concentrated in the tailings solution were found to be soluble, hence, potential ground-water contaminants. When tailings solution was neutralized either by addition of NaOH or by contact with clay liner or overburden material, the concentration of most of the nonradioactive and all of the radioactive constituents dropped to or below their maximum permissible concentrations (MPC). The mineralogy of the clay liner material changed only slightly after extended contact with the acid tailings solution. Also, the permeability of the clay liner material decreased with time. Measured permeabilities ranged from 2.3×10^{-9} cm/s to 2.8×10^{-11} cm/s.

Solubility Tests. In a companion project at the Pacific Northwest Laboratory (PNL), thermodynamic considerations predicted that for the Eh-pH range of natural aqueous environments, the dominant species of Pu is likely to be Pu(V) in relatively oxidizing environments and Pu(III) in reducing environments. Because of the lack of methods of determining Pu(V) in environmental samples containing trace concentrations of Pu, Pu(V) has not been previously identified in these samples. Plutonium (VI) has generally been assumed to be the dominant

species in relatively oxidizing environments. However, results at PNL show that solutions in equilibrium with $^{239}\text{Pu(IV)}$ hydroxide contain Pu(V) . The presence of Pu(V) in solutions contacting $^{239}\text{PuO}_2(\text{S})$ is also inferred from the data. These data for Pu and others for Am, Np, and U indicate that actinide migration is controlled largely by the solubility of the actinide species and that in most cases a waste repository can be designed to provide an environment which will control these actinides at concentration levels below environmental concern.

Modeling

Solute Flow. Radionuclide transport through unsaturated media was investigated by using two solute transport models to describe the transport of tritium and strontium-85 in laboratory columns. A two parameter convective-dispersive model was compared with a four parameter mobile-immobile water model. Both models adequately described the movement of tritium and strontium through small (5 cm x 27.5 cm) columns and the movement of tritium through a large (0.5 m x 1.7 m) column. The dispersion coefficient was found to be sensitive to changes in both velocity and column length. The mobile-immobile water equations were not as sensitive to changes in experimental scales as the convective-dispersive equation. Both models were relatively successful in describing the rapid flush of strontium-85 from a column initially leached with a low salt solution followed by a high salt solution, a phenomena called the snow plow effect. The four parameter mobile-immobile water model predicted the initial release of the strontium more accurately than the two parameter convective-dispersive model. Both models confirm enhanced mobility of strontium-85 with leaching solutions of increased salt concentration.

Radon Gas and Water Flow. A new approach was used to analyze radon emissions from uranium mill tailings. The approach has been developed based upon the analysis of partially saturated water flow effects on diffusion of radon through the cover and tailings materials. Such an analysis provides the moisture distribution with depth and time in the profile, hence, the gas filled pore space available for radon diffusion is known as a function of both depth and time. Through using the diffusion coefficient for radon gas in air, appropriately reduced to account for the porous material at the particular gas filled pore

space, the bulk diffusion coefficient, which in the past could only be obtained experimentally, is provided directly. Solution for the radon concentration in the engineered tailings-cover profile is then possible so the radon emission rate at the ground surface can be calculated directly.

The modeling of radon diffusion through cover materials has allowed for detailed analysis of the requirements needed in design of multilayer earth (soil) covers to control radon to prescribed limits. Figure 4 shows the multilayer cover system designed and tested by PNL. This design shows promise in effectively reducing radon emissions from uranium tailings to levels acceptable by proposed US government standards ($<2 \text{ pCi m}^{-2} \text{ sec}^{-1}$). Simulation of water flow and retention in soil covers was carried out using UNSATV, an unsaturated water flow model with the capability of simulating the long-term (multilayer) water balance of a soil cover system. The simulations indicate that a multilayer cover system using as its radon control layer, a compacted, wet clay/gravel layer can be constructed in such a way that it maintains its wetness for extended time periods.

FIGURE 4. Multilayer Cover for Radon Control. Air Filled Pore Space Curves are for Control Layer, H.

International Atomic Energy Agency Coordinated Research Program:
Migration and Dispersion of Radionuclides from Storage of Radioactive
Wastes in Terrestrial Environments

Research Agreement 2444/CF: Characterization of Radionuclide and Moisture
Movement Through Sediments in an Arid Region.

Battelle Pacific Northwest Laboratory, USA.

PUBLICATION LIST

- Gee, G. W., and C. S. Simmons. 1979. Characterization of the Hanford 300 Area Burial Grounds: Task III - Fluid Transport and Modeling. PNL-2921, p. 111.
- Gee, G. W., A. C. Campbell, B. E. Opitz, and D. R. Sherwood. 1980. Interaction of Uranium Mill Tailings Leachate with Morton Ranch Clay Liner Soil Material. Proceedings of the Third Symposium on Uranium Mill Tailings Management. Civil Engr. Dept. Colo. State Univ. Ft. Collins, CO. p. 333-352.
- Gee, G. W., A. C. Campbell, D. R. Sherwood, R. G. Strickert, and S. J. Phillips. 1980. Interaction of Uranium Mill Tailings Leachate with Soils and Clay Liners: Laboratory analysis/progress report. NUREG/CR 1494. USNRC Washington, DC.
- Gee, G. W., A. C. Campbell, P. J. Wierenga and T. L. Jones. 1981. Unsaturated Moisture and Radionuclide Transport: Laboratory analysis and modeling. PNL-3616. Pacific Northwest Laboratory, Richland, WA.
- Jones, T. L., W. A. Jordan and G. W. Gee. 1979. A Test of the Bresler-Russo-Miller method for Unsaturated Conductivities. RHO-SA-125. Rockwell Hanford Operations, Richland, WA.
- Nelson, R. W., G. W. Gee, and C. A. Oster. 1980. Radon Control by Multilayer Earth Barriers. 1. Modeling of Moisture and Density Effects on Radon Diffusion from Uranium Mill Tailings. Proceedings of the Third Symposium on Uranium Mill Tailings Management. Civil Engr. Dept. Colorado State University, Ft. Collins, CO. p. 79-88.
- Nelson, R. W., A. E. Reisenauer, G. W. Gee. 1980. Model Assessment of Alternatives for Reducing Seepage of Contaminants from Buried Uranium Mill Tailings at the Morton Ranch Site in Central Wyoming. NUREG/CR 1495. USNRC Washington, DC.
- Nelson, R. W., A. E. Reisenauer, and G. W. Gee. 1980. Model Assessment of Contaminant Seepage from Buried Uranium Mill Tailings. Proceedings of the First International Conference on Uranium Mine Waste Disposal. Society of Mining Engr. New York. PNL-SA-8457. p. 245-270.

Phillips, S. J., L. L. Ames, R. E. Fitzner, G. W. Gee, G. A. Sandness and C.S. Simmons. 1980. Characterization of the Hanford 300 Area Burial Grounds. Final Report. Decontamination and Decommissioning. PNL-2557. Pacific Northwest Laboratory, Richland, WA.

Phillips, S. J., A. C. Campbell, M. D. Campbell, G. W. Gee, H. H. Hooper, and K.O. Schwarzmiller. 1979. A Field Test Facility for Monitoring Water/Radionuclide Transport through Partially Saturated Geologic Media: Design Construction and Preliminary Description. PNL-3226. Pacific Northwest Laboratory, Richland, WA.

Rai, D., and R. J. Serne. 1978. Solid Phases and Solution Species of Different Elements in Geologic Environments. PNL-2651, Pacific Northwest Laboratory, Richland, WA.

Rai, D., R. J. Serne, and D. A. Moore. 1979. Solubility of Plutonium Compounds and Their Behavior in Soils. PNL-SA-7892. Pacific Northwest Laboratory, Richland, WA.

Relyea, J. F., D. Rai, and R. J. Serne. 1979. Interaction of Waste Radionuclides with Geomedia: Program, Approach, and Progress. Scientific Basis for Nuclear Waste Management, Vol. 1, pp. 379-394. PNL-SA-7289.

Serne, R. J. and D. J. Bradley. 1979. Methods Used to Estimate Geochemical Aspects of Radionuclide Migration from Waste Repositories. Transactions, 32:161-162, 1979 Annual Meeting, American Nuclear Society, Atlanta, GA. PNL-SA-7519.

Serne, R. J., D. Rai, M. J. Mason, and M. A. Molecke. 1977. Batch Kd Measurements of Nuclides to Estimate Migration Potential at the Proposed Waste Isolation Pilot Plant in New Mexico. PNL-2448, Pacific Northwest Laboratory, Richland, WA.

INTERNATIONAL
ATOMIC
ENERGY AGENCY

CLAIM
FOR REIMBURSEMENT
OF TRAVEL EXPENSES

To be approved by relevant DDG if travel
dates, itinerary or mode of travel are other
than those shown on Travel-Authorization.

Name of Claimant

Dr. G.W. GEE

Personnel No.

Travel-Authorization No.

TR-81-

Accompanied by dependents:

NAME

RELATIONSHIP

DATE OF BIRTH

Department / Division

TONF-WM

Title and Grade

Partic. CR Progr.

Date

Signature

If leave was taken during travel period,
indicate dates and type:

Currency in which payment requested: U.S. Dollars

Payment to be made by ☒ Check / ☐ Bank Transfer. (If bank transfer requested
give name and address of bank and name and number of current account):

A. Tilly-Frey, 2675

Travel Report will be submitted on:

n/a

Date

ITINERARY	ARRIVAL			DEPARTURE			MODE OF TRAVEL	is TERMINAL ALLOWANCE claimed (yes or no)		Approved Transit and Subsistence			Subtotals
	Day	Month	Year	Day	Month	Year		Arrival	Depart	Days	Daily rate	Amount	
Richland, USA				27	06	81	Air/		Yes				
London	28	06	81		07	81	"	Yes	"				
Richland	03	07	81				"	"					

PLEASE NOTE:

Approximately 80% of expenses are covered by the
attached cheque.

Please complete the attached claim form:

- 1). Signature of claimant
- 2). Details of any additional expenses (train, bus)
- 3). Date of departure and arrival
- 4). Please return completed claim form to the

Scientific Secretary before you leave the meeting.

1	29/6/81	Pl. reimburse train	
		London-Harwell-London	(\$17 USA)
		* travel by rental car	
		claim for equivalent	
		train fare includes	
		parking fee while unloading.	
2	03/7/81	Richland-Seattle-Richland	
		(round trip from Seattle)	
		Car parking + personal car expense	\$ 116.30
		\$28.30 + 140 mi @ 0.20/mi	133.30

I certify that the entries represent my actual itinerary and my actual authorized
expenses while on official travel, and for which payment (other than advances
listed on reverse of Travel-Authorization) has not been received.

1981-06-29

Date

Signature of Claimant

(For Duty Travel only)

1981-07-

Date

E. Kecskemeti

Signature of Approving Official

TOTAL

LESS ADVANCES

BALANCE DUE IAEA IF ANY

NET PAYMENT IN US \$

RATE OF EXCHANGE

NET PAYMENT IN
REQUESTED CURRENCY