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Water infiltration rates in the unsaturated zone at the Idaho National Engineering Laboratory estimated from chlorine-36 and tritium profiles, and neutron logging

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ABSTRACT: Environmental tracers (chlorine-36 and tritium) were used at the Radioactive Waste Management Complex (RWMC) to estimate natural water infiltration rates in the unsaturated zone near buried nuclear waste. Chlorine-36 and tritium were measured in the soil column to determine the depth of the maximum concentration of these radionuclides produced by atmospheric testing of nuclear devices in the late 1950's and early 1960's. Historically, the nuclear fuel reprocessing operations at the Idaho Chemical Processing Plant also have contributed to the atmospheric deposition of these radionuclides at the Idaho National Engineering Laboratory.

Soil cores for analysis of chlorine-36 and tritium were taken from undisturbed soil near subsurface instrumentation (thermocouple psychrometers, tensiometers, and a neutron moisture gage) that has been used since 1985 to establish the vertical hydraulic head distribution in the unsaturated zone. The neutron-probe access hole used is in an undisturbed soil profile adjacent to the RWMC. Rates calculated from the profiles of the environmental tracers and from the neutron-logging indicate that site-specific net infiltration to the unsaturated zone ranges from 0.36 to 1.1 cm/year. This represents 2 to 5 percent of average annual precipitation.

1 INTRODUCTION

The Idaho National Engineering Laboratory (INEL) comprises about 2300 km² of the eastern Snake River Plain in southeastern Idaho. The INEL was established in 1949 and is used by the U.S. Department of Energy (DOE) to construct and test nuclear reactors and to participate in various defense programs. Radiochemical and chemical wastes generated at the INEL and other DOE facilities have been buried in trenches and pits at the Radioactive Waste Management Complex (RWMC) since 1952. Since 1970, low-level radioactive wastes have been buried and transuranic wastes have been stored on above-ground asphalt pads in retrievable containers. From 1952 to 1986, approximately 180,000 m³ of low-level radiochemical and transuranic wastes that contained about 9.5 million curies of radioactivity were buried (Pittman, 1989, p. 2).

Because of the potential for radionuclides to migrate from wastes buried in the unsaturated zone to the aquifer about 177 m below land surface, the U.S. Geological Survey constructed two instrumented test trenches in the surficial sediment adjacent to the northern boundary of the RWMC (Figure 1) to collect data for determining net infiltration rates of water. The test trench area is a sparsely vegetated, predominately silt loam site. The depth to the sediment-basalt contact varies from about 4 to 6 m. Vegetation is mixed Big Sagebrush (Artemisia tridentata ssp. wyomingenis) and Crested Wheatgrass (Agropyron cristatum). Data collection began in 1985 and is continuing. Subsurface data are collected using thermocouple psychrometers, tensiometers, and a neutron moisture gage. The data and calculations presented herein are preliminary and this is the first in a series of reports on the unsaturated zone at the INEL.

In 1990 and 1991, chlorine-36 (^{36}Cl) and tritium (^{3}H) concentrations were measured in soil samples collected from boreholes near the test trenches. This report presents net water infiltration rates calculated from the depth of the maximum concentration of these radionuclides and compares them to rates calculated from neutron-probe data. The relative movement of these environmental tracers has been studied in similar semi-arid areas by Phillips and others (1988) in New Mexico and Hunt and others (1990) in Nevada.

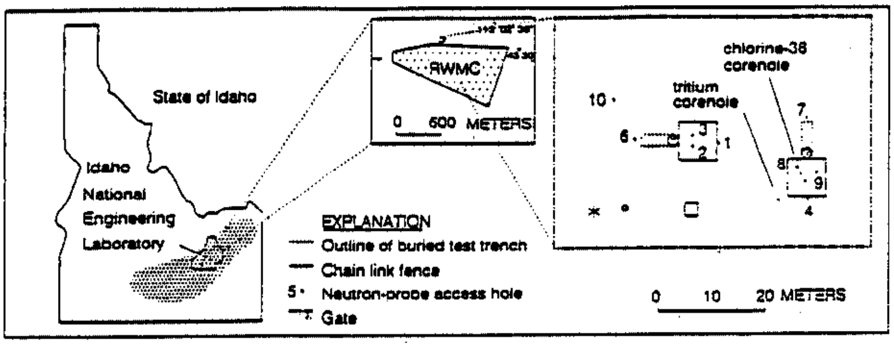

Figure 1.--Location of the test trench area near the Radioactive Waste Management Complex at the Idaho National Engineering Laboratory.

2 METHODS

A soil core for ^{36}Cl analyses was collected by hand in November 1990 using a 7.6-cm diameter auger bit and chamber. Sample intervals ranged from 0.1 to 0.15 m and were selected to ensure the collection of 800-1000 g of soil per sample. The borehole was completed to a total depth of 5.6 m and was discontinued at the sediment-basalt interface. The soil samples were stored in sealed containers, weighed in the field and transported to the laboratory for determination of volumetric water content. The samples were then transported to the DOE Environmental Measurements Laboratory (EML) where the ^{36}Cl and stable chloride were isolated and chemically purified to produce a AgCl precipitate (Conard and others, 1986). The ^{36}Cl concentrations were then determined using accelerator mass spectrometry (AMS) at the University of Rochester, New York (Kubik and others, 1987).

The soil samples for ^{3}H analyses were collected by air rotary drilling in November 1991. Sample intervals were selected in the same manner as those for ^{36}Cl. This borehole was completed to a total depth of 5.3 m; the basalt contact was not reached. Water was collected from the soil by distilling in a vacuum oven that was connected to ice-water and liquid-nitrogen traps. The water samples were then enriched and counted with a liquid scintillation detector (Thatcher and others, 1977, p. 79-81).

The neutron probe used to measure soil water content contains a source of fast, high-energy neutrons and a slow (thermal) neutron detector. The probe is lowered into a cased hole to specified depths and readings are recorded. Hydrogen present in the soil water slows down the neutrons for detection by the probe. The moisture data are displayed as raw hydrogen counts that can be correlated with the water content of the cores taken during construction of the probe access hole (Campbell Pacific Nuclear, 1984, p. 1).

3 RESULTS AND DISCUSSION

Chlorine-36 and ^{3}H concentrations for the soil cores are shown in Table 1 and graphs of ^{36}Cl ratios, ^{3}H concentrations, and volumetric soil water content as a function of depth are given on Figure 2. For annual average net infiltration rate calculations, a 4-year average volumetric water content of 0.19 over the entire length of neutron access hole 9 was used (Figure 2). The use of average volumetric water contents for the ^{36}Cl and ^{3}H soil samples collected in the fall of 1990 and 1991, respectively, would bias the rate calculation toward the dry time of the year.

3.1 *Chlorine-36*

Chlorine-36 is produced in the environment by cosmic-ray interaction with argon. Approximately two-thirds of natural ^{36}Cl production is in the stratosphere and the remaining one-third is in the troposphere (Faure, 1986, p. 416). Chlorine-36 also was produced during nuclear weapons tests conducted over the oceans during 1952-58 (Schaeffer and others, 1960). After a residence time in the troposphere of about 1 week, ^{36}Cl is removed in wet and dry deposition. For the latitude of the INEL, Bentley and others (1986) predicted that pre-weapons tests ^{36}Cl/Cl ratios in ground water should be in the range of $300\text{-}640 \times 10^{-15}$. The precipitation of ^{36}Cl produced during weapons tests in the 1950's

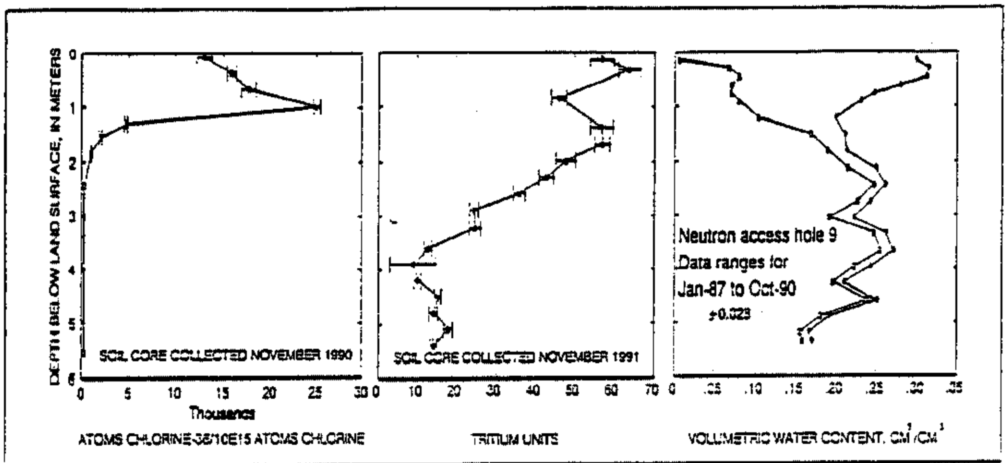

Figure 2.--Chlorine-36 ratios, tritium concentrations, and volumetric soil water content as a function of depth.

exceeded the natural production by three orders of magnitude in rainwater at Long Island, New York (Bentley and others, 1982). Phillips and others (1988) predicted that approximately 2.5×10^{12} atoms of ^{36}Cl m^{-2} were deposited on the Earth's surface at the latitude of the INEL. It is this pulse of ^{36}Cl produced during nuclear weapons tests that can be used in the unsaturated zone in semiarid climates to calculate net water infiltration rates.

An estimate of net water infiltration can be made using the penetration depth of the maximum ^{36}Cl or ^{3}H pulse in the soil column (produced during weapons tests in 1955 for ^{36}Cl and 1963 for ^{3}H) and an average volumetric water content over the profile. The movement of ^{36}Cl and ^{3}H through the unsaturated zone can be treated as piston flow modified by dispersion (Anderson and Sevel, 1974). The largest ^{36}Cl/Cl ratio ($25000 \pm 400 \times 10^{-15}$) at the RWMC occurs at a depth of about 1.1 m (Figure 2). A more accurate estimation of net infiltration rate may be calculated by determining the depth of the largest concentration of ^{36}Cl atoms per kilogram of soil from the volumetric water content, chloride concentration, and ^{36}Cl/Cl ratio (Table 1). That depth is centered at 1.3 m where the concentration is $30{,}200 \pm 1{,}100 \times 10^{6}$ atoms ^{36}Cl/kg soil. This concentration corresponds to 5.4×10^{12} atoms m^{-2} which is more than double the value predicted by Phillips and others (1988) for the latitude of the INEL as a result of fallout from nuclear weapons tests. By integrating the atoms of ^{36}Cl m^{-2} over the upper 2.5 m of the soil column, there is in excess of 44×10^{12} atoms. These excess ^{36}Cl atoms may be attributed to stack emissions from the Idaho Chemical Processing Plant (ICPP); these emissions began in 1953 and are discussed in another paper (Beasley and others, in preparation). The average net water infiltration rate calculated with the 4-year average volumetric water content of 0.19 is 0.71 cm/year.

3.2 *Tritium*

Environmental ^{3}H concentrations began increasing in 1953 in response to nuclear weapons testing; maximum concentrations were reached in 1963 after large-scale weapons testing conducted in 1962. Tritium concentrations in the vicinity of Idaho Falls reached a maximum of 3000--4000 Tritium Units (one Tritium Unit (TU) = one tritium atom/10^{18} hydrogen atoms) which exceeds natural levels by approximately three orders of magnitude (Michel, 1989). By using a local 39-year average annual precipitation rate of 22 cm/year (Clawson and others, 1989, p.77), the total deposition of ^{3}H at the site was 3500--4000 TU-meters. Since 1963, atmospheric ^{3}H concentrations have declined by two orders of magnitude.

An estimate of infiltration is obtained by locating the leading edge of the 1963 maximum. For this profile, the 1963 peak was centered at a depth of 1.65 m. By using the 4-year average volumetric-water content of 0.19, the average annual net infiltration rate is 1.1 cm/year. The decay-corrected ^{3}H mass balance inventory in these soil samples also was used to calculate an infiltration rate. The total

Table 1.--Volumetric water content and chloride, tritium, and chlorine-36 atom concentrations with depth

Depth, meters	Volumetric water content, cm^3/cm^3	Chloride, mg/L	Tritium, tritium units (TU)	$\frac{10^6 \text{ atoms anthropogenic } ^{36}\text{Cl}^*}{\text{Kg soil}}$	$\frac{\text{atoms } ^{36}\text{Cl}}{10^{15} \text{ atoms Cl}}$
Soil samples collected in November 1990 for chlorine-36 analyses					
0-0.14	0.04	4.2	–	1000±70	13000±800
0.3-0.43	0.13	3.8	–	1100±30	15800±500
0.6-0.73	0.14	3.8	–	1140±30	17600±800
0.91-1.1	0.13	15	–	7100±110	25000±400
1.2-1.4	0.13	355	–	32400±1100	4700±150
1.5-1.6	0.21	780	–	26600±220	2000±140
1.8-1.9	0.24	1070	–	14300±1500	1000±70
2.4-2.5	0.31	1290	–	**	290±14
3.9-4.0	0.30	1240	–	**	260±12
5.5-5.6	0.16	740	–	**	280±15
Soil samples collected in November 1991 for tritium analyses					
0-0.17	0.14	–	57.0±3.0	–	–
0.17-0.42	0.07	–	63.8±2.8	–	–
0.61-0.69	0.03	–	insufficient sample	–	
0.69-0.93	0.08	–	46.0±1.9	–	–
1.2-1.5	0.12	–	57.0±2.7	–	–
1.5-1.8	0.15	–	57.1±1.9	–	–
1.8-2.1	0.17	–	47.9±2.4	–	–
2.1-2.4	0.15	–	42.9±1.7	–	–
2.4-2.7	0.17	–	36.0±1.3	–	–
2.7-3.0	0.17	–	24.6±1.0	–	–
3.0-3.4	0.15	–	24.7±1.5	–	–
3.4-3.7	0.16	–	12.8±0.7	–	–
3.7-4.0	0.17	–	9.0±6.0	–	–
4.0-4.3	0.15	–	10.1±0.7	–	–
4.3-4.6	0.14	–	15.3±0.8	–	–
4.6-4.9	0.13	–	14.2±1.0	–	–
4.9-5.2	0.13	–	18.0±1.0	–	–
5.2-5.5	0.14	–	14.2±0.8	–	–

* A natural meteoric ^{36}Cl/Cl ratio of 300 x 10^{-15} was subtracted for INEL and was selected from Bentley and others (1986) and data presented here

**Concentrations are from natural production

^{3}H observed in the measured core was 35–40 TU-meters. Approximately 800 TU-meters would still be present if decay were the only process removing ^{3}H from the soil profile. A ^{3}H mass balance calculation gives an average annual net infiltration rate of 0.89 cm/year.

3.3 *Neutron logging*

Infiltration in semiarid climates has been studied by use of neutron logging in sandy soils at the DOE Hanford site near Richland, Washington (Gee and Kirkham, 1984) and in sandy soils near Socorro, New Mexico (Stephens and Knowlton, 1986). Estimated infiltration rates at the Washington site were 8.5 cm/year in a wet year, and 0.7 to 3.7 cm/year at the New Mexico site.

Neutron-probe access hole 9, nearest to the ^{36}Cl and ^{3}H core holes (Figure 1), was used to determine soil water content and variability with depth and time. Saturated and unsaturated hydraulic conductivities were estimated by the instantaneous profile and unit gradient methods using field data collected within 10 m of the study area (Kaminsky, 1991). Hydraulic properties of soil cores taken at neutron access hole 9 were also estimated from laboratory-determined saturated hydraulic conductivities and water retention curves. Pressure heads were estimated from the volumetric water contents at neutron-probe access hole 9 using the soil water retention function of van Genuchten (1980). Unsaturated hydraulic conductivities were obtained by the combination of the van Genuchten retention function with the pore-size distribution model of Mualem (1976). Hydraulic conductivities [K(θ)] were determined using the 4 year mean volumetric water contents below the root zone. The vertical soil-water flux (Q_z) below the root zone was calculated using the unsaturated form of Darcy's Law:

$$Q_z = -K(\theta)\left(\frac{dh}{dz} - 1\right) \qquad (1)$$

where ($dh/dz - 1$) is the total hydraulic head gradient with z increasing upwards. The neutron-probe infiltration rate estimate based on vertical head distributions from 3.7 to 5.2 m is 0.36 cm/year.

4 CONCLUSIONS

Net water infiltration rates were calculated from maximum penetration depths of two environmental tracers produced during atmospheric nuclear weapons testing. These rates were compared to Darcian flux calculations based on neutron logging data collected for 4 years. The average annual infiltration rates calculated from ^{36}Cl and ^{3}H profiles in the unsaturated zone adjacent to the Radioactive Waste Management Complex were 0.71 and 1.1 cm/year, respectively. The tritium mass balance method gives a rate of 0.89 cm/year. Neutron logging data in the same area were used to calculate an average annual infiltration rate of 0.36 cm/year. The range of infiltration rates (0.36-1.1 cm/year) represents 2-5 percent of the long-term annual average precipitation at the Idaho National Engineering Laboratory.

As determined in previous studies , tritium appears to move deeper in the soil column than ^{36}Cl, possibly by vapor transport of ^{3}H and not of ^{36}Cl. The infiltration rate for ^{3}H (1.1 cm/year) is larger than that calculated from the ^{36}Cl profile (0.71 cm/year), which is nearly double the rate calculated from neutron logging (0.36 cm/year).

The calculated infiltrations rate estimates should be considered site specific and may not represent the full range of infiltration rates in the study area. Further work utilizing all three techniques is needed. Future work at the site will include chloride mass balance calculations of net infiltration and a determination of the effects on infiltration rates of a caliche ($CaCO_3$) layer encountered at various depths to about 1 m. The effects on infiltration rates from stack emissions of ^{36}Cl and ^{3}H at the ICPP will also be determined. Future neutron logging work will include flux calculations from other neutron probe access holes located in undisturbed soils and simulated waste pits.

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