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Distribution of Plutonium and Americium Beneath a 33-yr-old Liquid Waste Disposal Site[1]

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ABSTRACT

The distribution of Pu, ^{241}Am, and water in Bandelier Tuff beneath a former liquid waste disposal site at Los Alamos was investigated. The waste use history of the site was described, as well as the previous field and laboratory studies of radionuclide migration performed at this site. One of the absorption beds studied had 20.5 m of water added to it in 1961 in an aggressive attempt to change the distribution of radionuclides in the tuff beneath the bed. Plutonium and ^{241}Am were detected to sampling depths of 30 m in this bed, but only found to depths of 6.5 to 13.41 m in an adjacent absorption bed (bed 2) not receiving additional water in 1961. After 17 yr of migration of the slug of water added to bed 1, 0.3 to 5.1% of the Pu inventory and 3.0 to 49.6% of the ^{241}Am inventory was mobilized within the 30-m sampling depth, as less than one column volume of water moved through the tuff profile under the bed. The results of similar lab and field studies performed since 1953 were compared with our 1978 data and site hydrologic data was used as a time marker to estimate how fast radionuclide migration occurred in the tuff beneath absorption bed 1. Most of the radionuclide migration appeared to have occurred within 1 yr of the 20.5-m water leaching in 1961. The implications of our research results to nuclear waste management were also discussed.

Additional Index Words: **americium, nuclear waste management, plutonium, soil chemistry, soil transuranics.**

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In late 1943, a site with the primary responsibility for the purification of Pu was established at Los Alamos, NM. Because of urgency, limited construction time, and the lack of information on the resulting radioactive wastes, it was initially decided to dispose of radioactive wastes in several ways. Untreated liquid wastes were at first discharged into canyons, underground storage tanks, and absorption beds filled with gravel and cobble (Abrahams, 1962; Rogers, 1977), such as the beds at Area T in the DP West site of the Los Alamos National Laboratory.

The interaction of radionuclides in these liquid wastes with local soils and geologic materials was initially studied in the laboratory. Cores of Bandelier Tuff collected at Los Alamos were contaminated with waste solutions of Pu, essentially all of which was retained in the top few millimeters of the core even after subsequent leaching (Christenson et al., 1958). In 1953, the U.S. Geological Survey (USGS) conducted a preliminary study to determine the vertical distribution of Pu beneath the absorption beds at Area T, because "past information indicated that this particular location has probably received more Pu contamination from liquid wastes than any other area" (Herman, 1954). Five 3- to 6-m deep holes were drilled in and around the absorption beds and an effort was made to gather samples at 30-cm intervals using a pick and shovel, a driven pipe, and a drilling rig with a core barrel. The results of this study indicated that the vertical migration of Pu occurred within 6 m of the surface of the absorption beds and that Pu is readily retained by the components in the bed.

A joint USGS-Los Alamos National Laboratory study (Abrahams, 1963; Christenson and Thomas, 1962) was begun in October 1959 at Area T with the construction of a 9.1-m deep, 1.8-m wide, 3.6-m long caisson on the northeast corner of absorption bed 1. Twelve horizontal holes were drilled at 61-cm depth intervals and long enough to terminate at about the center of the absorption bed. Liquid samples were collected from each hole under vacuum for radionuclide assays and soil water determinations were performed in each hole with a neutron moisture gauge. Cuttings from these horizontal holes were collected in the summer of 1959 to estimate the vertical distribution of Pu in absorption bed 1. Average gross alpha radiation readings in these samples at the 3-m depth were ". . .3003 cpm/g and gradually decreased to 28 cpm/g at 8.5 m, except for a high concentration (402 cpm/g) found at the 7.3-m depth"; the later observation was attributed to an acutal vertical fissure in the tuff at this depth, which allowed wastes from the absorption bed "to drain unchanged to a lower level" (Christenson and Thomas, 1962).

After documenting the vertical distribution of radioactivity in the 1959 to 1960 study, an aggressive attempt was then made to change the distribution of Pu beneath absorption bed 1 by adding 10.8 m of effluent containing Pu in July 1961 and 9.7 m of tap water 30 d later. During and immediately after the releases, a large effort was also expended to characterize the infiltration and movement of water and Pu in the tuff beneath the absorption bed. Unsaturated water flow was observed from 12 to 30 m beneath this absorption bed, with about 10 m of water-saturated tuff occurring above this layer after the addition of the tap water.

In 1967, another reconnaissance study was made of the Area T absorption beds (Purtymun, 1967). Water samples were again collected from the tuff beneath the bed and the moisture contents of the tuff were again logged at selected depths. Results showed the maximum tuff water contents moved from the 3.7-m depth in August 1961 to the 12-m depth in January 1967. It was again concluded that most of the Pu was retained in the upper 6 m of the absorption bed, with some Pu moving to greater depths through open joints.

In 1974, a detailed series of laboratory studies was initiated with crushed and intact Bandelier Tuff at Argonne National Laboratory, which showed that

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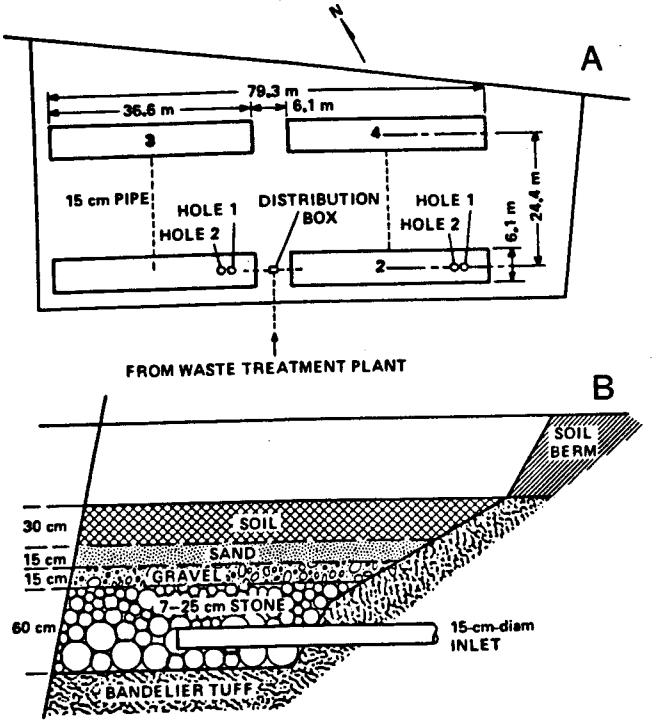

Fig. 1. Design of absorption beds at Area T; A, site plan of absorption bed system, B, cross section of an absorption bed.

waste and aqueous solutions of Pu and Am exhibited anomalous migration behavior (Fried et al., 1975; Fried et al., 1976; Fried et al., 1977; Fried et al., 1978). This research demonstrated that Pu appeared to exist in two forms, one which (probably the hydrolyzed form) migrated much more rapidly than the "ionic" form. The experimental results suggested a predicted penetration rate of the more mobile Pu phase of about 217 cm/yr when accompanied by unsaturated water flow in the tuff (Fried et al., 1975)!

The objective of this field study was to determine the distribution of Pu, ^{241}Am, and water beneath two absorption beds at Area T in Los Alamos as a function of depth and the waste use history of each absorption bed. The vertical distributions of radionuclides and water were related to the occurrences of fractures and geologic units of tuff in each profile. The findings of this field study are also compared with the results of other studies performed at this site and are discussed relative to the long-term migration of water and radionuclides in the tuff at this site.

WASTE USE HISTORY AND DESCRIPTION OF STUDY SITE

The absorption beds at Area T are the oldest used for the disposal of liquid wastes at Los Alamos (Fig. 1) and have been described in detail (Rogers, 1977). After the construction of these 1.2-m deep, 36.6-m by 6.1-m absorption beds was completed in 1945, they received untreated radioactive liquid wastes from 1945 through 1951 from DP site. The DP West liquid waste treatment plant was installed in 1952 (Fig. 1), largely because the volume of liquids discharged to the beds had exceeded the holding capacity of the absorption beds, despite the fact that the beds had been equipped with a distribution box, located between beds 1 and 2, which ensured that approximately equal volumes of wastes were discharged to these two beds. About 89% of the 69 260 m^3 of liquid effluents added to the Area T absorption beds were added between 1945 and 1960, with the remaining 11% added in rapidly decreasing amounts until 1967 (Rogers, 1977). A new treatment plant was built in 1967, which also infrequently discharged treated wastes into the absorption beds. However, almost all of these treated effluents were discharged to the canyon north of the plant.

Both the addition rate and the type of waste added to the absorption beds changed with time at Area T. About 98% of the estimated 370 000 MBq of Pu discharged to the absorption beds was added as untreated wastes between 1945 and 1952 (Anonymous, 1974; Rogers, 1977). The concentration of Pu in the estimated 53 000 m^3 of untreated effluents during this period has been estimated at about 2000 Bq Pu L^{-1}, with an average fluoride concentration associated with the wastes of 160 mg L^{-1}. However, about 40 m^3 of untreated effluents containing large concentrations of ammonium citrate were released into the beds from June 1951 to July 1952, and these wastes contained about 230 000 Bq Pu L^{-1} and 200 mg fluoride L^{-1}. The smallest contributions to the beds came between 1953 and 1967 when about 16 000 m^3 of treated effluents containing only about 3.3 Bq Pu L^{-1} were discharged to the absorption beds. Although most of the physical and chemical properties of these liquids were described (Rogers, 1977), the results of the 1961 study show the pH of the raw wastes usually ranged from 3.0 to 4.0, with about 50% of the alpha activity and 75% of the solids in the raw wastes passing through a 0.45 μm Millipore filter (Christenson and Thomas, 1962). These waste solutions contained an average of 1245 mg K L^{-1}, 197 mg Na L^{-1}, 57 mg Ca L^{-1}, 7 mg Mg L^{-1}, 200 mg F L^{-1}, 139 mg NO$_3$ L^{-1}, 70 mg Cl L^{-1}, and 36 mg SO$_4$ L^{-1} during 30 d of daily sampling in this study.

Detailed geologic descriptions of the Area T site were summarized in 1977 (Rogers, 1977). The absorption beds were excavated in Unit 3 of the Tshirege Member of the Bandelier Tuff. The upper 3.6 m of a typical geologic profile consists of a moderately welded, light brownish gray tuff. This layer is underlain by a 3-m thick layer of reworked tuff and pumice, which was emplaced contemporaneously with the upper unit in a channel or low relief cut into the lower tuff unit. A sharp contact zone is found between this reworked tuff and the lower, moderately welded, light gray tuff unit, which has an approximate thickness of 33 m. This later unit is underlain by a moderately to densely welded tuff, giving a total thickness of Bandelier Tuff in excess of 250 m. The top of the main zone of saturation is about 350 m below the surface of the mesa.

The saturated hydraulic conductivity of 20 intact tuff samples from the upper unit of tuff at Area T, collected adjacent to absorption bed 1, ranged from 0.05 to 0.29 cm h^{-1} (Nyhan, 1979). However, a more detailed description of the geohydrology of Bandelier Tuff was recently reported (Abeele et al., 1981).

METHODS

Our field study was initiated in 1978 to determine the vertical distribution of ^{241}Am, Pu, and water beneath the absorption beds at Area T. Two 30.5-m deep holes were drilled through each of the absorption beds 1 and 2 during 1978 (Fig. 1). Continuous core samples of the tuff were collected beneath the beds by driving a split spoon sampler, 60-cm long and 7.6-cm diam, through a 23-cm diam hollow-stem auger. The sampler was driven with either an 82-kg or a 181-kg drop hammer. At the end of each core run, to minify sample cross-contamination, the auger was advanced to the bottom of the core hole before the next core was collected.

Core samples were cut up into 15-cm segments as they were removed from the split spoon, immediately placed in glass jars, and brought back to the laboratory. Each sample was dried for 72 h at 110°C to determine soil water. The sample was then crushed in a plastic bag, mixed on a sheet of paper, and assayed for ^{241}Am and Pu using an L x-ray and gamma ray radionuclide assay system described previously (Trujillo et al., 1980; Nyhan et al., 1983). Radionuclide inventories for each hole were determined by multiplying the concentration of either ^{241}Am or Pu by the total oven-dry weight of tuff in the depth segment for every segment in the hole below the gravel-cobble bed bottom, and expressing this result as MBq of radioactivity for each hole.

The inventory of soil water in each hole was calculated from the gravimetric water content and bulk density of all the samples from each hole. The bulk density was calculated from the total oven-dry

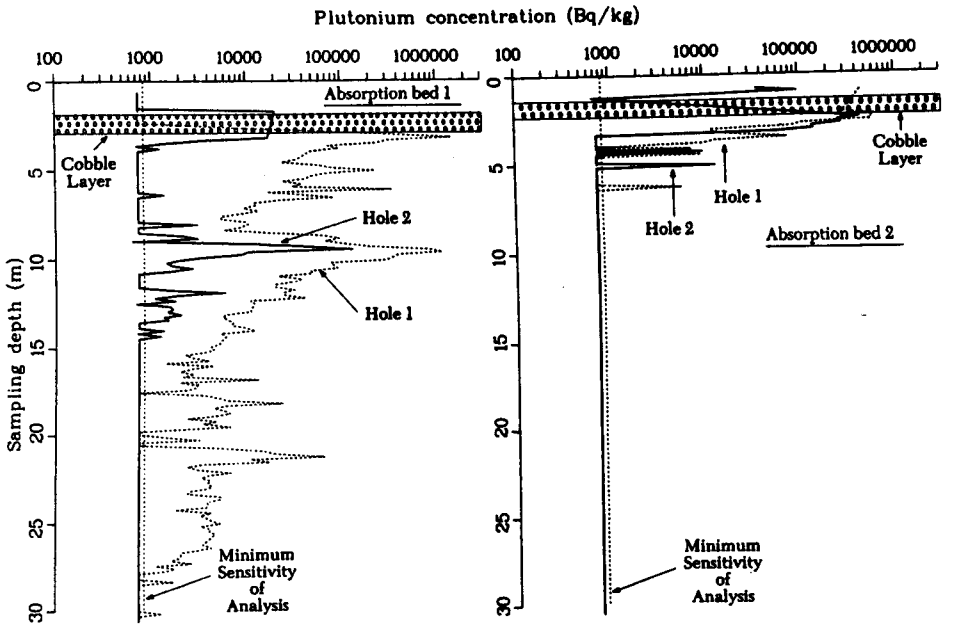

Fig. 2. Concentration of plutonium as a function of sampling depth for absorption beds 1 and 2 in 1978.

weight of the tuff samples from each 15-cm core segment and the known sampling volume of the split spoon sampler. The bulk density was multiplied by the gravimetric water percentage of each sample divided by 100, and by the sampling depth to calculate the total amount of water in each core segment.

Although the hydraulic conductivity (K) of the upper unit of tuff adjacent to bed 1 was determined (Nyhan, 1979), we were not able to collect undisturbed samples of tuff within the contact zone between the two tuff units to assay for K. Thus, we approximated the value of K for the contact zone using the data collected in the 1961 Area T study (Christenson and Thomas, 1962), by the constant-head method according to the following equation

$$K = \frac{Q}{At}\frac{L}{\Delta H}. \qquad [1]$$

The volume of water, Q, that passed through the contact zone tuff core of area A and length L was measured for a known time t and for a constant hydraulic head difference, ΔH. The value of Q was approximated from the difference of the neutron moisture gauge data collected on 26 July 1961 and 23 August 1961 below the contact zone (12.19- to 25.91-m depth), for a tuff core with a radius of 30 cm, corresponding to the estimated radius of investigation of the moisture gauge (Nyhan et al., 1983; Q = 18 615 cm^3 water). Thus, the estimated values for A and t were 2827 cm^2 and 672 h. The value for ΔH, 969 cm, was determined from the thickness of the tuff layer (above the contact zone) which was saturated with water, according to the 1961 neutron moisture gauge data. The thickness, L, of the contact zone was estimated from the drilling log data and the distribution of water beneath bed 1 in 1978 (L = 250 cm).

Soil pH was determined on a few samples of tuff using a 2:1 water-to-soil ratio (w w^{-1}) so that the effect of the nonradioactive components of the waste solutions on the tuff geochemistry could be partially evaluated.

RESULTS AND DISCUSSION

We will first examine the distribution of radionuclides beneath the absorption beds in 1978 and then, since the movement of water under the beds is the driving force for radionuclide migration, the distribution of water in the tuff will be analyzed. The inventories and vertical distributions of radionuclides and water will then be related to site geohydrologic characteristics and the pH of selected tuff samples. Our data will then be compared with the results of similar studies performed at Area T in the past, and the temporal changes in water and radionuclide distributions at this site will be discussed.

Vertical Distributions of Radionuclides and Water Beneath the Absorption Beds

The distribution of Pu and ^{241}Am concentrations as a function of sampling depth is presented for both absorption beds in Fig. 2 and 3. The cobble layer indicated in these figures marks the gravel-cobble layer in the bottom of the original absorption beds (Fig. 1). Although the individual data points are not plotted in these figures, the high degree of detail reflects the fact that this data set represents radionuclide assays on a total of 800 samples.

Radionuclides were generally detected to the bottom of both holes in absorption bed 1, which received the additions of large amounts of tap water and effluents in 1961 to provide the additional driving force for radionuclide migration (Fig. 2 and 3). More specifically, in hole 1, Pu was detected to a sampling depth of 30.33 m and ^{241}Am to a depth of 30.48 m. Although the hole 2 samples generally contained smaller radionuclide concentrations than the samples from hole 1, ^{241}Am was detected to a depth of 30.78 m and Pu was found to a sampling depth of 14.48 m in hole 2, indicating higher mobility for ^{241}Am than for Pu under these environmental conditions.

The Pu and ^{241}Am did not penetrate nearly as far into the tuff beneath absorption bed 2, because this bed did not receive additional water in 1961 (Fig. 2 and 3). The minimum sensitivity of analysis (depicted in these figures as a sample plotted to the left of the minimum sensitivity of analysis line) was reached for tuff samples collected at sampling depths greater than 6.55 m for Pu and 13.41 m for ^{241}Am in hole 1. Plutonium was only

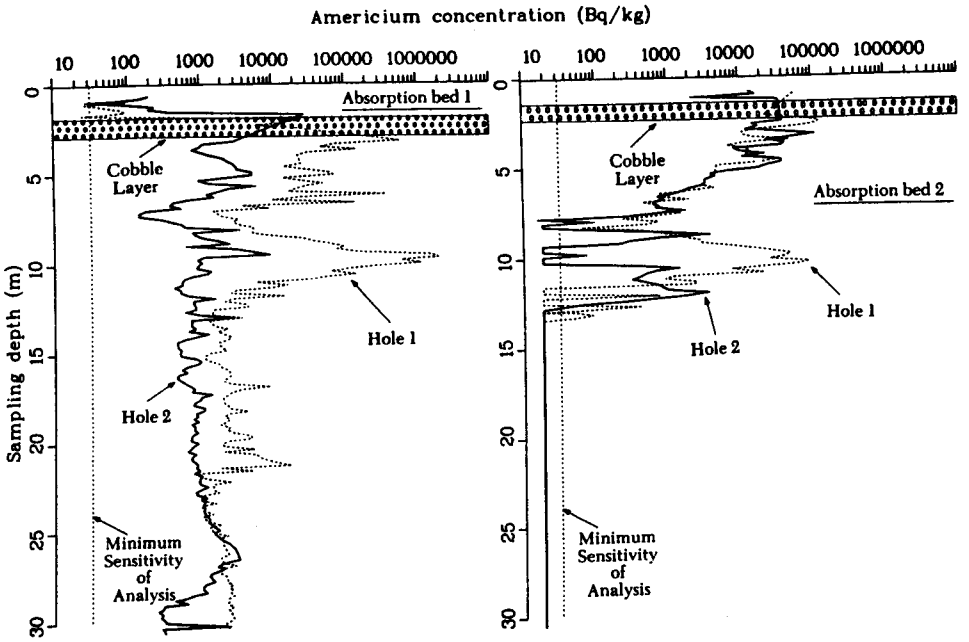

Fig. 3. Concentration of ^{241}Am as a function of sampling depth for absorption beds 1 and 2 in 1978.

detected to a depth of 5.18 m in hole 2 and ^{241}Am to a sampling depth of 12.80 m in this hole, indicating a higher degree of mobility of ^{241}Am than Pu in this absorption bed, just as for absorption bed 1. This observation was also shown for the Area T field data described previously (Fried et al., 1977).

The distribution of water in the tuff (gravimetric water content) beneath the Area T absorption beds is shown as a function of depth below the current land surface (Fig. 4). The absorption bed 1 profiles showed generally higher tuff water contents than similar profiles in absorption bed 2, with some samples attaining gravimetric water contents as high as 0.30 kg kg^{-1} (Fig. 4), a value very close to saturation 0.38 kg kg^{-1} for Bandelier Tuff. The water content of most of the tuff samples collected in bed 1 was obviously still enhanced relative to the bed 2 samples due to the additions of water made in 1961. For example, considering only the samples collected from the 25.15- to 30.48-m depth in the hole 1 profiles, the absorption bed 1 average tuff water content was 0.077 kg kg^{-1}, whereas the average gravimetric content for similar bed 2 samples was only 0.059 kg kg^{-1}.

Correlations of Radionuclide and Water Content Distributions with Site Geohydrology and Geochemistry

Although all of the variations in soil water content (Fig. 4) and radionuclide concentrations (Fig. 2 and 3) can not be explained, several changes can be accounted for based on the geohydrologic data collected in this and other studies at Area T.

The first high concentrations of radionuclides and water encountered in the tuff beneath the gravel-cobble layer in absorption bed 1 were found at sampling depths of 4 to 5.5 m (Fig. 2–4), where a highly weathered, light orange-gray tuff layer with a high clay content was found. This layer, previously described as Bed B (Rogers, 1977), would be less permeable than the rest of the surrounding tuff and probably resulted from the severe chemical and hydrologic tuff weathering processes brought about by the acidic liquid wastes added to this absorption bed.

The next major increase in tuff water content and radionuclide concentrations occurred at a sampling depth of about 8 to 9 m in absorption bed 1 (Fig. 2–4). At this depth we encountered a change in tuff units from the upper-lying, light brownish-gray, moderately welded tuff to the lower-lying, light gray, moderately welded tuff. This contact zone was identified on the basis of color changes and the change in the total amount of force required to drive the split spoon sampler into the tuff. For example, in hole 1 of absorption bed 1, the upper tuff unit required 102 to 157 blows of a 181-kg hammer to extract a 61-cm core sample, whereas the unit under the contact zone only required 23 to 60 blows to collect a similar sample. Contact zones such as this exhibit increased welding, decreased porosity (Abeele et al., 1981), and, thus, decreased conductivity relative to the adjacent tuff units. Using the neutron probe data collected in the 1961 study (Christenson and Thomas, 1962), we estimated the saturated hydraulic conductivity, K, of this layer to be 0.0025 cm h^{-1}, which corresponds to both a 10-fold lower water transmission rate and a 10-fold longer contact time between soluble radionuclides and tuff than for that of the overlying unit of tuff (Nyhan, 1979).

Fractures, originally formed by cooling of the tuff ash flows, commonly divide the tuff into irregular blocks and accounted for some of the variations in tuff water content shown in Fig. 4. Although a few fractures occurring from 3 to 12 m at this site could have received saturated flow of liquids directly from the large amounts of effluents discharged to absorption bed 1 (Abrahams, 1963), fractures usually act as barriers for

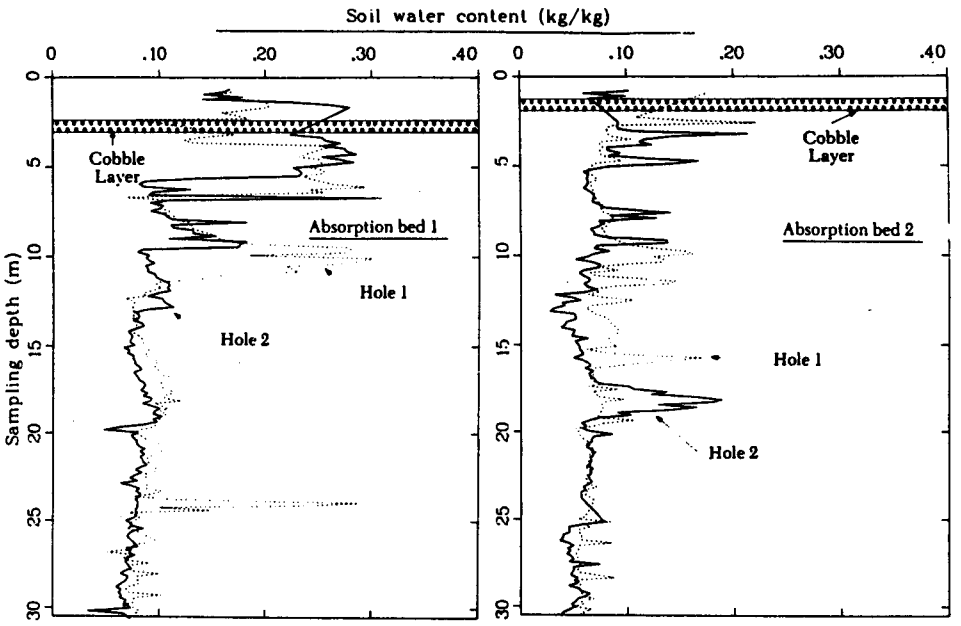

Fig. 4. Gravimetric water content as a function of sampling depth for absorption beds 1 and 2 in 1978.

unsaturated liquid flow (Abeele et al., 1981). For example, a fracture was found in hole 1 of absorption bed 2 at a depth of 10.06 to 10.21 m. The fracture fillings had a water content of 0.125 kg kg^{-1}, compared with a value of 0.162 kg kg^{-1} in the adjacent tuff sample collected at the 9.91 to 10.06 m depth. The water contents of tuff samples collected a the 23- and 24-m depths in hole 1 of absorption bed 1 were also elevated, probably indicating the presence of fractures close to and below the hole but not detected in the hole.

Since previous studies at Area T emphasized the role of fractures in promoting vertical radionuclide migration (Abrahams, 1963; Christenson and Thomas, 1962), we collected samples of fracture fillings and analyzed them for radionuclide concentrations. No significant differences were found in radionuclide concentrations between fracture fillings and adjacent tuff samples in eight out of 10 cases, where fractures were encountered at sampling depths ranging from 2 to 18 m in both absorption beds. Both cases involving higher radionuclide concentrations in the fine-textured fracture fillings than in the tuff adjacent to the fractures were found in the upper unit of tuff, i.e., at sampling depths of 6.6 m in absorption bed 1 (two-fold difference in radionuclide concentrations) and 3.5 m in absorption bed 2 (three-fold difference in radionuclide concentrations). Thus, these results tend to support the idea that fractures in the tuff generally act as barriers to unsaturated flow of migrating waste solutions (Abeele et al., 1981); however, fractures may play a role in conveying waste solutions through the tuff close to the bottoms of the absorption beds, where saturated flow conditions were more commonly found.

During the final week of the addition of 10.8 m of untreated, acidic (usually pH 3 to 4) wastes to absorption bed 1 in 1961, the pH of water samples extracted from the tuff at sampling depths of 0.9 to 8.5 m was studied (Christenson and Thomas, 1962). The pH values of these water samples usually ranged from 4 to 5, but returned to pH 8 to 9 after the subsequent addition of 9.7 m of tap water 4 weeks later, reflecting a dilution of the acidic wastes initially added to the tuff and the natural buffering capacity of the tuff.

We further evaluated these 1961 study results by performing pH determinations on a few of the tuff samples collected in our 1978 study (Table 1). The average pH of tuff samples collected in absorption bed 2 ranged from 7.0 to 8.0 below sampling depths of 16 m, which corresponded to sampling depths not receiving detectable levels of Pu and ^{241}Am wastes (Fig. 2 and 3). Samples collected in sampling locations other than these and beneath both beds, which obviously received large volumes of wastes (Fig. 2 and 3), exhibited no significant differences in pH (Table 1). This observation indicates that the natural buffering capacity of the tuff was still maintaining the pH of this geochemical system in 1978 (and not the acidity of the waste solutions added in the past), just as it did after the additions of water to absorption bed 1 in 1961.

Inventories of Radionuclides and Water Beneath the Absorption Beds

Since radionuclides were not found below 11.28 m beneath bed 2 (Fig. 2 and 3), the resulting inventory (for the projected area of the hole) was then summed for all the samples from depths of 0 to 11.28 and 11.28 to 27.13 m below both of the absorption beds (Table 2). For absorption bed 2, total Pu inventories ranged from 1.22 to 2.27 MBq and total ^{241}Am inventories ranged from 0.58 to 0.97 MBq. For absorption bed 1, hole 1 contained 7.85 MBq Pu and 8.25 MBq ^{241}Am, and 0.26 MBq Pu and 0.21 MBq ^{241}Am was found in hole 2. Coefficient of variation (standard deviation of mean/mean) estimates of radionuclide inventories were three- to four-fold higher for absorption bed 1 than for bed 2, probably reflecting enhanced variation in liquid waste deposition patterns close to the point of entry of the waste solu-

Table 1. Average of tuff samples collected beneath absorption beds at area T in 1978.

Sampling depth, m	Average and standard deviation of pH of tuff from absorption bed number†	
	1	2
Immediately below gravel cobble layer	7.6 (0.5)	6.8 (0.50)
10.06-10.21	7.6 (0.78)	7.0 (0.25)
16.00-16.15	8.7 (0.06)	7.3 (0.47)
19.51-19.66	7.4 (0.40)	7.5 (0.91)
25.60-25.76	7.5 (0.01)	7.0 (0.11)
30.02-30.18	7.5 (0.06)	8.0 (0.73)
30.18-30.33	8.1 (0.12)	7.6 (0.91)

† Average pH of one sample collected at each depth from each of two holes.

tions into the absorption bed (Fig. 1), as shown previously (Herman, 1954; Abrahams, 1963; Christenson and Thomas, 1962). Thus, even though the bed 1 samples were collected closer to where the wastes were added to the bed than the bed 2 samples were (Fig. 1), no significant differences in total radionuclide inventories could be found between these two absorption beds; this observation supports the idea that approximately equivalent amounts of wastes were added to each bed through the distribution box between beds 1 and 2 (Fig. 1).

To better understand the relationship between radionuclides and the major radionuclide redistribution factor, water, we calculated the total inventory of water in the tuff samples from each hole. The total centimeters of water in each sample were calculated by multiplying the sample's gravimetric water content (percent water divided by 100) by the bulk density of the sample and the sampling depth (cm). Because the maximum penetration of radionuclides beneath absorption bed 2 was 11.28 m below the gravel cobble layer at the bottom of the absorption bed, the inventory of water was calculated for the same depths as for the radionuclide inventories (Table 2). The water inventory calculations clearly demonstrate that even 17 yr after the 1961 additions of water to absorption bed 1, elevated water contents can still be found under this bed. Holes 1 and 2 in absorption bed 1 contained 435 and 380 cm of water, respectively, whereas holes 1 and 2 in absorption bed 2 only contained 286 and 260 cm of water, respectively. In addition, total inventories of water and radionuclides were directly correlated within each absorption bed, indicating enhanced radionuclide migration with elevated levels of water in the tuff (Table 2).

The most important information to be gleaned from Table 2, however, is related to the inventories of radionuclides and water found at the 11.28- to 27.13-m depths of both holes in absorption bed 1. Thus, in hole 1 of absorption bed 1, 0.396 MBq Pu and 0.247 MBq ^{241}Am were translocated to this depth (Table 2), representing 5.1 and 3.0%, respectively, of the radionuclide inventories in this hole. The samples from hole 2 of this absorption bed, which contained very low radionuclide inventories, exhibited only 0.00081 MBq Pu and 0.103 MBq ^{241}Am within the 11.28- to 27.13-m depth increment (Table 2), representing 0.3 and 49%, respectively, of the radionuclide inventories. The relative distribution of the water inventory with depth demonstrated an amazingly consistent pattern for this bed. The

Table 2. Inventories of plutonium, americium, and water beneath absorption beds at area T in 1978.

Depth below bottom of absorption bed, m	Inventory			
	Absorption bed 1		Absorption bed 2	
	Hole 1	Hole 2	Hole 1	Hole 2
	Plutonium (MBq)			
0-11.28	7.45	0.257	2.27	1.22
11.28-27.13	0.396	0.00081	0.00	0.00
Total inventory	7.846	0.258	2.27	1.22
	Americium (MBq)			
0-11.28	8.00	0.105	0.966	0.579
11.27-28.13	0.247	0.103	0.00	0.00
Total inventory	8.25	0.208	0.966	0.579
	Soil water (cm)			
0-11.28	241	211	135	120
11.28-27.13	194	169	151	140
Total inventory	435	380	286	260

top 11.28 m of the profiles below absorption bed 1 contained 55% of the inventory of water, with only 45% of the inventory found at the 11.28- to 27.13-m depth (Table 2). Thus, these data also indicate that 17 yr after the addition of a large slug of water to absorption bed 1, the tuff located 11.28- to 27.13 m below this bed contained significant radionuclide inventories and a 25% higher average water inventory than did the tuff at a corresponding depth below absorption bed 2, which did not receive a large addition of water in 1961.

DISCUSSION OF RADIONUCLIDE MIGRATION WITH TIME AT AREA T

Since the long term migration of radionuclides in the porous materials of a burial site is an important issue, we examined the question of what happened to the distribution of radionuclides with time at Area T. In the succeeding subsections we first demonstrate the results of previous studies of the vertical distribution of radionuclides at Area T, and compare them with the results of our 1978 study. We will then use the hydrologic characteristics of the tuff at Area T and at Mesita del Buey, NM as a time marker to more precisely estimate what happened with time to the vertical distribution of radionuclides at Area T.

Vertical Radionuclide Distributions at Various Sampling Dates

Estimates of the distribution of Pu beneath absorption bed 1 were made for the 1953 (Herman, 1954) and 1960 (Christenson and Thomas, 1962) studies and compared with our 1978 results. The results of the 1953 study used here were for the DPW-4 hole, which was closest to our hole 1 in absorption bed 1. In 1953, peak concentrations of 603 000 and 759 000 Bq Pu kg^{-1} were found above the tuff in the sand and gravel bed bottom, with 200 Bq kg^{-1} found at the maximum sampling depth of 6.1 m. The general vertical distribution pattern of Pu with depth at this time was similar to the 1978 data.

The 1960 data represent the average estimated Pu concentrations found in from 4 to 10 core samples per depth collected directly under absorption bed 1 when

Table 3. Inventory of water in the tuff below absorption bed 1 at three sampling times.†

epth below bottom of absorption bed, m	June 1961	August 1961	1978
0–11.28	221	401	241
11.28–27.13	211	350	194
Totals	432	751	435

† Inventory estimates from the neutron moisture gauge data of Christenson and Thomas (1962). The moisture gauge data were collected in June 1961 (before the addition of water to the absorption bed) and in late August 1961 (immediately after the last addition of water to the absorption bed).

horizontal holes were drilled into the northeastern corner of the absorption bed to a maximum sampling depth of 10 m. The original gross alpha radiation determinations (cpm g^{-1}) were intended to represent Pu concentrations, which we estimated, but were also known to reflect alpha-radiations by ^{241}Am. However, these 1960 data exhibited decreases in radionuclide concentrations from 125 000 Bq kg^{-1} immediately beneath the cobble-gravel layer to about 629 Bq kg^{-1} at the 6.71 m sampling depth, only to increase to 16 800 Bq kg^{-1} at 7.3 m and then decrease to 1110 Bq kg^{-1} at 10 m. The 1978 data we collected showed very similar trends, such as at the 7- to 8-m depth where the contact zone was encountered as well as throughout the rest of the profile.

However, both the 1953 and 1960 studies only sampled the upper 10 m of the absorption bed profile, so that no direct comparisons can be made with the data collected below this depth in our 1978 study.

Use of Site Hydrology as a Time Marker

Since the distribution of radionuclides 10 m beneath absorption bed 1 has only been determined in our 1978 study, we decided to estimate how fast radionuclide migration occurred beneath this bed from estimates of the movement of water, the driving force for radionuclide migration.

Besides our 1978 study, the only other comprehensive study of water distributions in the tuff below the absorption beds at Area T was done in 1961 (Abrahams, 1963; Christenson and Thomas, 1962). Hole 2 of this 1961 study was found to be adjacent to our hole 1 in absorption bed 1, and consisted of the location where a neutron moisture gauge access tube extended almost 30 m below the bed (this 1961 access tube was removed with subsequent waste operations at this site). We estimated water inventories in the tuff beneath bed 1 for two sampling dates in 1961 (using the methods described previously) and compared this neutron moisture gauge data with the results of our 1978 study (Table 3). In late June 1961, before the large addition of water to this absorption bed, the total water inventory beneath the absorption bed was 432 cm. In late August 1961, immediately after the addition of water to the absorption bed, the inventory of water in the tuff increased to 751 cm. Our data, which was collected in 1978 for hole 1, showed a total water inventory of 435 cm, a value very similar to the estimated inventory observed in June 1961, before the water was added to this absorption bed. Thus, 316 cm of water drained out of this tuff profile some time between 1961 and 1978, resulting in a 42% decrease in water inventory.

Table 4. Inventory of water in tuff at three sampling times in the Mesita del Buey study.†

Sampling date	Inventory of water in tuff, cm
20 Apr. 1960	99
6 Dec. 1960	160
12 Oct. 1961	88

† These data represent estimates from the neutron moisture gauge data of Christenson and Thomas (1962). The moisture gauge data were collected in June 1961 (before the addition of water to the absorption bed) and in late August 1961 (immediately after the last addition of water to the absorption bed).

Since no additional data were collected after August 1961 in Christenson and Thomas's 1961 study to follow the drainage of water out of the tuff, we decided to estimate the drainage rate from an infiltration-drainage experiment performed at Mesita del Buey (Abrahams, 1963), a site with similar geohydrologic characteristics. In this study, water was continuously added to a 0.91- by 0.91- by 0.91-m pit for 230 d at a rate of about 0.2 m d^{-1}, similar to the water addition rate in the Area T study (Christenson and Thomas, 1962). Neutron moisture gauge readings were collected to a depth of 11 m before and immediately after water additions to the pit, as well as after 286 d of drainage of the tuff (Table 4). The results show that 72 cm of water drained out of this tuff profile after only 286 d, with the result that the inventory of water in the tuff profile was back to what the original water inventory was under the Mesita del Buey pit.

The results of the Mesita del Buey study demonstrate that it only takes about 286 d of drainage for the inventory of water in the tuff profile to be reduced approximately to its original pre-addition value. This implies that most of the water and, thus, radionuclides, probably migrated between 1961 and 1962 at Area T, and not between 1961 and 1978. However, after 1962, the water and radionuclides probably did continue to migrate, but very slowly, since reduced levels of soil water result in unsaturated conductivities that are many orders of magnitude less than they were in 1961 when the water was originally added to this Area T absorption bed (Abeele et al., 1981).

SUMMARY AND IMPLICATIONS

The distribution of Pu, ^{241}Am and water beneath two absorption beds in a former liquid waste disposal site at Los Alamos was investigated 33 yr after the establishment of the site. Plutonium and ^{241}Am were detected to sampling depths of 6.5 to 13.4 m in absorption bed 2, which received 98% of its liquid wastes before 1952. However, absorption bed 1, which received 20.5 m of waste liquids and water in 1961 in an attempt to change the distribution of radionuclides in the tuff, exhibited detectable levels of Pu and ^{241}Am to maximum sampling depths of 30 m. These field observations are in sharp contrast with the results of the early laboratory studies of Pu solutions (with and without complexing agents) in tuff cores, which demonstrated that essentially all of the Pu was retained within the top few millimeters of the tuff core (Christenson et al., 1958). Whether or not

these differences in radionuclide behavior can be explained by physical and chemical differences in the liquid waste streams is unclear at this time and not within the scope of this study.

One of the major motivations for performing our field study involved the findings of the laboratory studies at Argonne National Laboratory, which demonstrated a distinct peak concentration of mobile Pu migrating 10 times faster than the bulk of the Pu in tuff. Whereas our 1978 field data did not show an analogous peak concentration band for either Pu or ^{241}Am, we did observe elevated levels of water and radionuclides within layers of tuff with large amounts of clay and within contact zones between tuff units. These heterogenous geologic layers (which did not exist in the lab studies) were shown to contain hydrologic characteristics which were different than those of the adjacent tuff, which in turn probably changed the kinetics of flow of water and radionuclides through the tuff.

A previous field study (Christenson and Thomas, 1962) attributed the major portion of the vertical migration of transuranics in tuff to be due to flow of liquid wastes through fractures in the tuff. Our results indicate that radionuclide concentrations in the tuff were generally correlated to the water content of the tuff and not just in sampling locations involving fractures. The ^{241}Am data, for example, suggest a continuous flow of water and ^{241}Am through almost 30 m of intact tuff and around fractures in the tuff (Fig. 3), which usually acted as barriers to unsaturated liquid flow.

Since earlier studies with our tuff had demonstrated that Pu could move with the water front in both unsaturated (aqueous Pu solutions; Fried et al., 1975) and saturated (probably a mixture of complexed and aqueous Pu species; Christenson and Thomas, 1962) flow conditions, we decided to estimate what fraction of our radionuclide inventories had been mobilized beneath absorption bed 1 (Table 2). After demonstrating that no significant differences in total radionuclide inventories could be detected between absorption beds 1 and 2, we demonstrated that from 0.3 to 5.1% of the Pu and from 3.0 to 49% of the ^{241}Am was mobilized under absorption bed 1 (Table 2). Whether or not these mobilized radionuclides are currently water soluble, potentially existing as various complexed, non-complexed, and colloidal chemical species, are questions outside the scope of our field study, but are definitely important to the nuclear waste management community.

We also estimated that the radionuclides translocated beneath absorption bed 1 were mobilized with the addition of less than 1 column volume of water (316 cm of water drained out of the tuff profile [Table 3], whereas one column volume of water, covering the area of one absorption bed to a sampling depth of 30.5 m, would contain 1067 cm of water). In contrast, although a different waste solution was probably used in a laboratory study with intact tuff cores, one study reported that 1% of the Pu inventory and only 0.022% of the ^{241}Am inventory could be mobilized after the addition of 1000 column volumes of water (Fried et al., 1976).

Since previous sampling programs at Area T had only attained maximum sampling depths of 10 m, we could not compare our 1978 radionuclide inventory estimates below 10 m with those observed at a previous date. Thus, we used site hydrology as a time marker to estimate that most of the radionuclide mobilization below 10 m in absorption bed 1 probably occurred within a year after the large slug of water was added to this bed.

By gaining an appreciation for the amounts of long-lived radionuclides migrating in soils and geologic materials and the geohydrology of a site, we can factor this information into our environmental monitoring and disposal practices. Thus, the probability that the public will be exposed to significant adverse health risks will be reduced. Data generated in this study are also currently being used to design field sampling programs to evaluate radionuclide migration at Area T in much more detail and to evaluate hydrologic models dealing with unsaturated water flow in Bandelier tuff. Information similar to that gathered in this study at Area T is needed for many sites with varying environmental conditions before the long-term behavior of long-lived radionuclides can be more clearly understood.

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