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Possible Chemical Analogues
transuranic Nuclides in the
9-24.

DISTRIBUTION OF PLUTONIUM AND AMERICIUM AT A FORMER
LOS ALAMOS WASTE DISPOSAL SITE

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ABSTRACT

A thirty year old liquid waste disposal site at Los Alamos Scientific Laboratory has been cored and a portion of each six inch increment analyzed for plutonium and americium. The site received unprocessed low level waste solutions from 1945 to 1951, and treated waste effluents from 1952 to 1967. The actinide content of each core increment was determined by low energy photon spectroscopy utilizing the characteristic L x-rays of each nuclide. The data obtained are presented as a histogram and compared to gross activity measurements made at the site.

A laboratory simulation of the site was also prepared using simulated waste solutions and a specimen of disposal site's geologic material, tuff, a welded volcanic ash. This simulation was scaled and proportioned according to archival records of the original site. The results of this experiment are compared with measurements from the actual core sample.

INTRODUCTION

One of the most serious problems confronting the nuclear energy industry is the long term isolation of radioactive wastes. Transuranic elements contained in such waste contribute significantly to the hazard, both in toxicity and persistence. Whatever method is finally chosen to contain these wastes, access of the actinides to the environment must be restricted. Deep terrestrial

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TABLE 1
COMPOSITION OF EFFLUENTS

Date	Volume (gallons)	Chemical Composition	Plutonium Concentration
1945 - 1950	1.4×10^7	160 ppm F^- 13 ppm NH_4^+	60 c/m/ml
1951 - 1952	1.0×10^4	"Conc" ammonium citrate 200 ppm F^-	7×10^3 c/m/ml
1953 - 1967	4.0×10^6	0.1M K_2NO_3	Trace

produces immediate surface runoff; the annual precipitation of 460 inches; distribution of the waste solution; for modeling purposes, it was assumed collected received ten percent of

Cores were taken at six inch intervals accomplished without water for it was packaged in small polyethylene Laboratory. Each sample was ground to prevent cross contamination into a sample holder. Sample holes square 4 cm on a side with a 5/16 cellulose tape on each side for 0.4 ml of powdered material. The Several analytical methods for detection before choosing low energy photon by alpha proportional counting was samples to insure complete recovery offers the greatest sensitivity, and, although promising, offers a

The spectrometer consists of a plate by 5 mm thick. Its shallow design energy photons. Its small diameter 350 eV in the L x-ray region, and entire detector-cryostat assembly background shield with a graded L x-ray region is 0.27 c/m. A mu controlled real time data acquisition

In operation, all samples were counting was accomplished by initial subtraction from a sample spectrum. ^{241}Am the keV photopeak and that value used in turn was subtracted from the x-ray contribution from that point ^{239}Pu was then determined by summing spectrometer efficiency was calibrated in identical manner to the sample.

A laboratory simulation of this validity of our modeling technique taken near the site was cut into a surface was waxed to restrict later was attached to a glass tube which were to percolate the rock. Waste ^{237}Pu tracer; the latter in place

produce immediate surface runoff. A small fraction, perhaps less than 5%, of the annual precipitation of 460 mm infiltrates the waste trenches. The actual distribution of the waste solutions within the four trenches is not known. For modeling purposes, it was assumed that the trench from which the core was collected received ten percent of the total discharged.

Cores were taken at six inch intervals to a depth of 30 feet. Drilling was accomplished without water for lubrication or cooling. A portion of each core was packaged in small polyethylene bottles and shipped to Argonne National Laboratory. Each sample was ground in a new mortar which was discarded after use to prevent cross contamination, dried four hours at 110° C., and loaded into a sample holder. Sample holders were constructed from 2 mm thick cardboard squares 4 cm on a side with a 5/8 inch hole in the center. A single strip of cellophane tape on each side formed a thin, vial-shaped cavity which contained 0.4 ml of powdered material. The average weight of each sample was 0.35 g. Several analytical methods for determining actinide content were considered before choosing low energy photon spectroscopy. Chemical separation followed by alpha proportional counting was rejected because of difficulty in dissolving samples to insure complete recovery of the actinides. This method, however, offers the greatest sensitivity. Neutron activation analysis was also tried and, although promising, offers no greater sensitivity than our chosen method.

The spectrometer consists of a planar lithium drifted germanium detector 2 cm² by 3 mm thick. Its shallow design results in a low background from high energy photons. Its small diameter is a compromise between high resolution, 350 eV in the L x-ray region, and good geometric efficiency, about 8%. The entire detector-cryostat assembly was contained in a large 6 inch thick steel background shield with a graded x-ray liner. The resulting background in the L x-ray region is 0.27 c/m. A multichannel analyzer coupled to a computer controlled real time data acquisition system completed the spectrometer.

In operation, all samples were counted for a minimum of 15 hours. Data processing was accomplished by initial subtraction of a normalized background count from a sample spectrum. ²⁴¹Am then was determined by integrating under the 60 keV photopeak and that value used to normalize a standard ²⁴¹Am spectrum which in turn was subtracted from the sample spectrum. This step eliminated the Am x-ray contribution from that portion of the spectrum due to the Pu x-ray. ²³⁹Pu was then determined by summing the integrals of the three L x-rays. The spectrometer efficiency was calibrated using synthetic standards prepared in an identical manner to the samples.

A laboratory simulation of this site was prepared primarily as a test of the validity of our modeling techniques (fig. 2). A specimen of low Alamosa tuff taken near the site was cut into a cylindrical plug 18 x 75 mm. The curved surface was waxed to restrict lateral flow of liquid. The cylinder of rock was attached to a glass tube which served as a reservoir for liquids which were to percolate the rock. Waste solutions were synthesized from ²⁴¹Am and ²³⁷Pu tracer; the latter in place of ²³⁹Pu to facilitate low level determination.

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Calculation	Plutonium Concentration
7×10^3	60 c/m/ml
7×10^3	7×10^3 c/m/ml
	Trace

Solution volumes and concentrations were proportional (as closely as possible from available Los Alamos records) to permit an arbitrary scale of 2.2 ft/cm. The synthetic waste solutions were then delivered to the test specimen in the same sequence as at the site at a controlled rate of 2 ml/hour using a metering pump. Disposal at the site, however, was not continuous but rather sporadic. Since no record of the cycle is available, no attempt was made to reproduce it. We fully realize the complications such simplification of the model would produce but felt the chance to compare even a crude model to an actual site justified this approach. The effluent of the column was collected and prepared for analysis. At the conclusion of the experiment, the specimen was removed from the apparatus, dried, cut into 0.5 cm sections, and also analyzed by gamma spectrometry to determine the distribution of the actinides in the rock.

RESULTS

Examination of the actinide content in the core (fig. 3) showed that detectable amounts of these nuclides did indeed migrate downward under the prevailing conditions to 20 feet below the bottom of the trench. Elevated concentrations of both nuclides are found at less than 1 foot and at 13 feet. The bulk of the americium is centered around the 9 foot mark. Noteworthy is the abrupt diminution of plutonium and americium below 13 feet; in fact, only 0.2% of the detected plutonium was below that level. On the other hand 45% of the plutonium was 1 foot or less from the bottom of the trench.

The distribution of actinides in the simulated model (fig. 4) is in excellent agreement with our earlier modeling work using tracers and simulated rain (Fried et al., 1976). Characteristic of these data are strong retention of actinides at the near surface with exponential tailing downward. In addition, plutonium exhibits a second concentration band beyond this tail. The percent of plutonium in this band is a function of the prior chemical treatment of the tracer material. The chemistry of plutonium is complex, existing in a IV, V, and a VI oxidation state. Polymeric states are also known at high pH. The strongly retained large peak at the surface is most likely Pu(IV). The faster moving species is not so readily characterized. The amount of fast moving plutonium in the model here depicted is much smaller than earlier models, reflecting, perhaps, the lower pH of the feed solution. It appears in the histogram as a slight rise at 6 cm with a rapid drop. An examination of the eluent (fig. 5) from the model shows no readily discernable pattern. It contains no americium and only 5% of the plutonium. It does indicate the mobilizing influence of the various waste solutions upon plutonium. Most interesting is the rapid drop in the elution of plutonium once water has rinsed the waste solutions from the rock and is the only eluting agent. This agrees with previous work which shows that distribution of plutonium is dependent upon the conditions of loading on the rock such as chemical composition and rate. Once plutonium is adsorbed by the rock, the actual water induced migration is extremely slow.

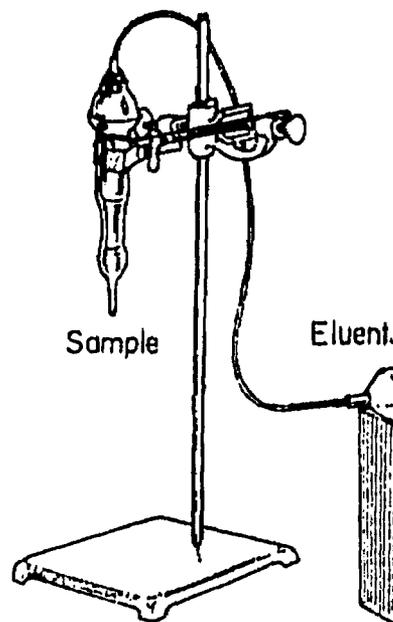


FIGURE 2. Experiment

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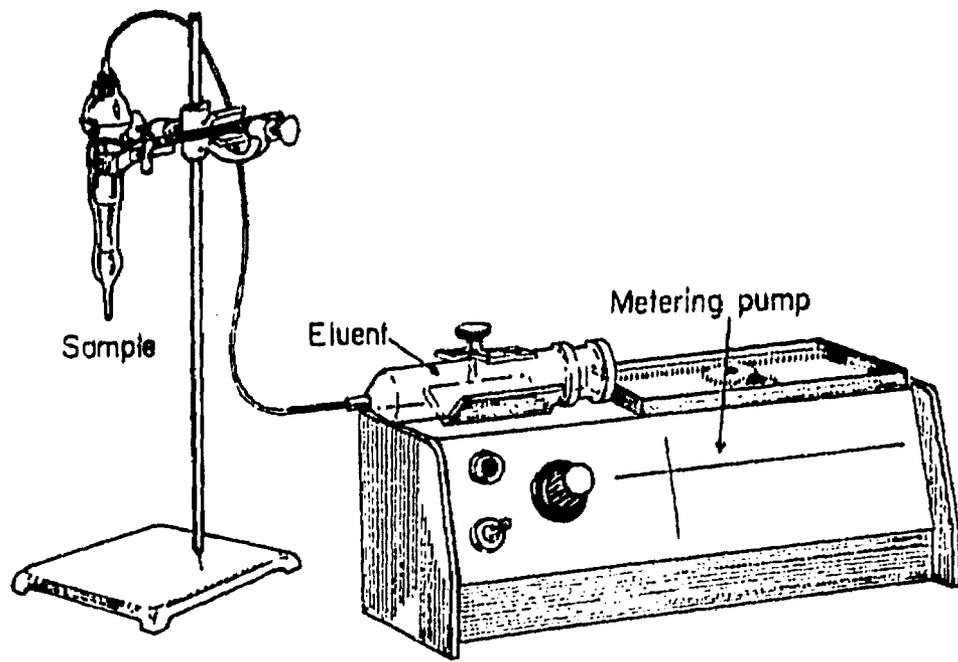
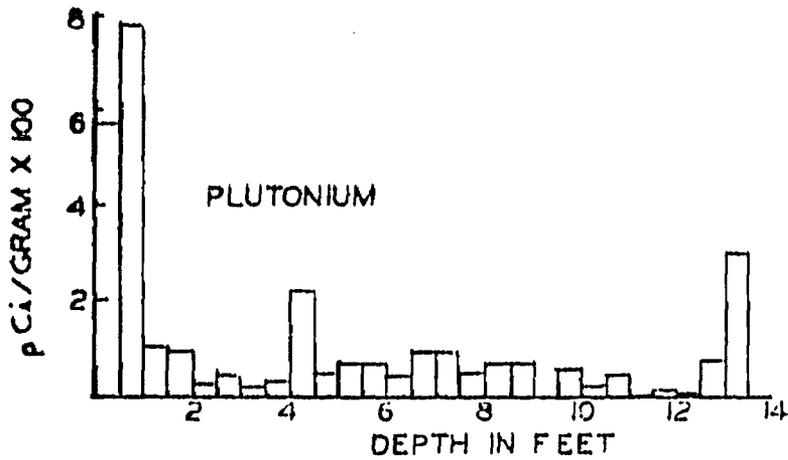
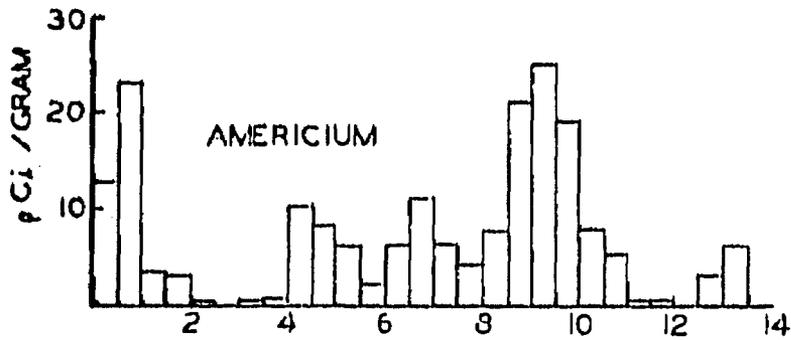


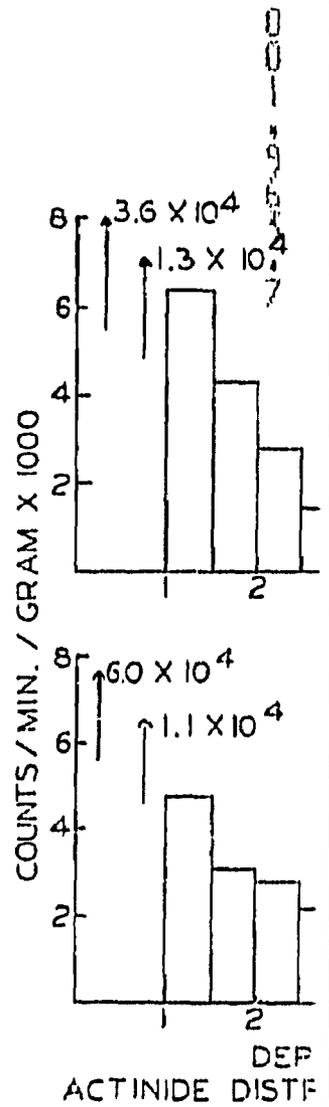
FIGURE 2. Experimental modeling apparatus.

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ACTINIDE DISTRIBUTION AT DISFOSAL SITE

Figure 3



ELUTION OF PLUTONIUM
FROM LABORATORY MODEL

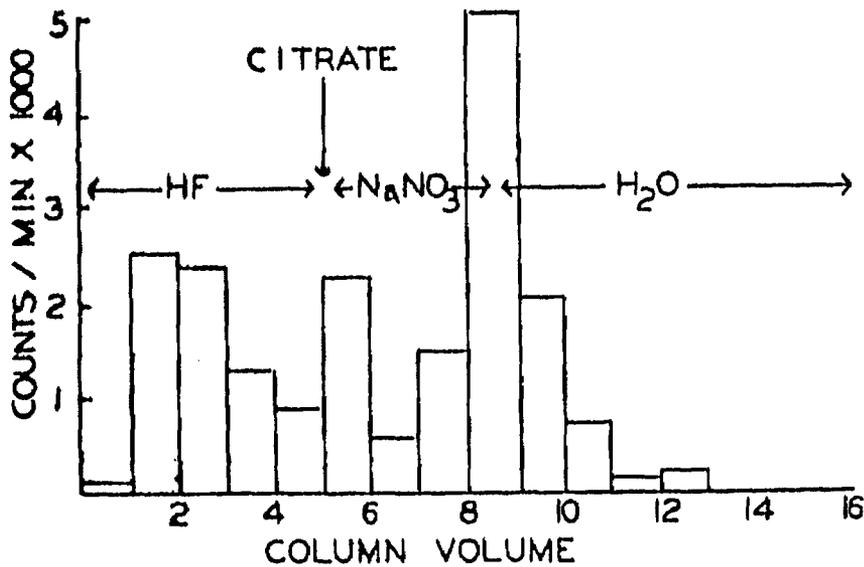


Figure 5

In order to compare results of certain reservations must be un-
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 depth of penetration of 14 feet
 2.2 ft/cm. Our predictions for
 of the near surface peak predic
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This work was supported by the
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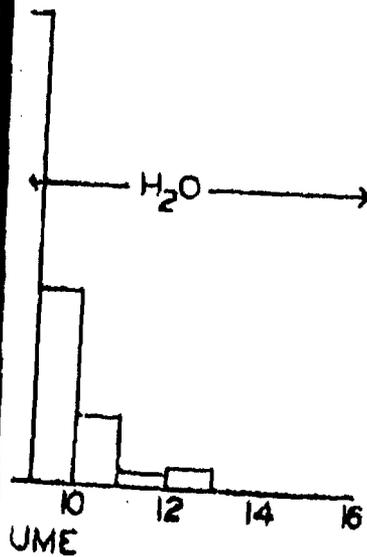
CONCLUSIONS

In order to compare results of the model to results from the actual site, certain reservations must be understood. The model constructed in the laboratory was primarily a chemical model. It could not reproduce geologic conditions of the site, the most prominent of which are fractures in the rock which can greatly accelerate or retard the flow of solutions through a stratum depending upon whether they are void or clay filled. Such geologic features contribute to the character of the site data and, in fact, may outweigh the chemical effects. For example, examination of the core indicated the presence of a clay-filled fracture at approximately 16 feet. This fracture appeared to be restricting downward flow of moisture, possibly accounting for the reduced activity below that depth in the core. Nevertheless, the model correctly predicts the large surface concentration for plutonium as well as a maximum depth of penetration of 14 feet. The latter is based on the modeling scale of 2.2 ft/cm. Our predictions for americium did not fare nearly so well. Instead of the near surface peak predicted by the model, americium was found at the site in a diffuse band near 9 feet. The danger of inferring too much from a single coring experiment is obvious. Therefore, more corings and evaluations are planned and under way with the hope of resolving the discrepancies and confirming the predictions.

ACKNOWLEDGMENT

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PLUTONIUM MODEL



002-3-63

REFERENCE

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THE RELATIONSHIP OF THE
FATE OF TRANSURANIC

R. E. Wildung, R. Dri
Bat:
Pacific Northwest
Richland,

ABS

This review considers the influence of processes on the long-term solubility of plutonium and other transuranic elements in the soil cycle. Emphasis is placed on chemical and microbial processes affecting solubilization and transformation of insoluble in soils strictly on their characteristics.

Soluble, diffusible Pu in soils is to be largely present as particulate. Evidence suggests that microorganisms and that the nonparticulate plutonium solution by inorganic or organic processes. The role of soil microorganisms in the plant-availability of the transuranic elements is discussed in terms of (1) known chemistry of organic ligands, (2) known microflora, and (3) principal mechanisms of direct alteration (valence state changes, metabolite interactions, influence on cycling processes (biological uptake).

The toxicity of Pu to microorganisms is however, soil microorganisms are apparently due to radiation rather than to the chemical nature of the organisms have been shown to be altered and altering its form in the cell. Pu complexes tend to be of higher valence (Pu-DTPA) and negatively charged. The toxicity of Pu, as defined, is dependent upon organochemical exposure during growth. These factors are discussed in terms of their source, soil properties, and soil processes. The relative influence of these factors on predicting the long-term behavior of Pu in the terrestrial environment.

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