LETTER REPORT

TITLE: Review of the paper, "Distribution of Plutonium and Americium Beneath a 33-year-old Liquid Waste Disposal Site", J. Environ. Qual. 14, 501-9 (1985), by J. W. Nyhan, B. D. Drennon, W. V. Abeele, M. L. Wheeler, W. D. Purtymun, G. Trujillo, W. J. Herrea, and J. W. Booth, Los Alamos National Laboratory, Los Alamos, New Mexico

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SUMMARY

Significant fractions of the americium (3.0 to 49.6%) and plutonium (0.3 to 5.1%) in an old waste disposal bed were much more mobil in the underlying Bandelier tuff than had been predicted based on laboratory experiments. Since the migration in Bandelier tuff was under unsaturated conditions, similar overestimation of americium and plutonium retardation based on laboratory experiments could be possible in predicting retardation at the Yucca Mountain candidate repository site in Topopah Spring tuff; see, for example, Kerrisk, LA-10414-MS. The results in the paper illustrate the importance of verifying laboratory migration information by field studies.

1. DESCRIPTION AND SUMMARY OF THE PAPER

This paper describes a field study of the migration of americium and plutonium in Bandelier tuff beneath old liquid waste disposal beds at Los Alamos. Two disposal beds were studied, which had received large amounts of americium- and plutonium-containing liquid wastes in the 1950s. Water was added to one of the disposal beds in 1961 in an aggressive attempt to change the distribution of plutonium in the tuff beneath the bed. The volume of water added was such that unsaturated conditions were maintained in the underlying tuff. Core samples for radionuclide analysis were taken from beneath the two beds in 1959-1960 prior to water addition, and from beneath the treated bed in 1967. Additional core samples were taken from beneath the two beds in 1978 after the initial findings suggested significant migration of some of the plutonium and americium through the Bandelier tuff.

The 1978 samples showed different migration behavior for the two beds. In the absence of added water, detectable plutonium was not found below the 6.5-m depth or americium below the 13.4-m depth in tuff beneath the disposal bed. After the 17-year migration period, the waste disposal bed, which had been treated with additional water, showed that 0.3 to 5.1% of the plutonium and 3.0 to 49.6% of the americium had been mobilized in the tuff beneath the bed. For this treated bed, water (and americium and plutonium) had moved to at least the 30-m depth in tuff, the deepest core samples taken. Water movement appeared to be by unsaturated flow, since fracture fillings contained the same levels of radioactivity and water content as did the adjacent tuff samples.

The field observations are described as being in sharp contrast to early laboratory column studies with plutonium solutions and cores of Bandelier tuff, which showed that all the plutonium was retained in the top few mm of the tuff core. Another laboratory study showed that only 1% of the plutonium and 0.022% of the americium would be mobilized by 1000 column volumes of water. In contrast, the water added to the treated disposal bed was less than 1 column volume. Thus, americium and plutonium were much more mobile in the field than in the laboratory. The paper states that it is not possible to say at this time whether the migrating americium and plutonium are water soluble species, either complexed or uncomplexed, or if they are colloidal forms.

2. *RELEVANCE TO REPOSITORY PERFORMANCE MODELING*

In modeling the predicted performance of candidate repository sites, sorption of some radionuclides by the host rock is often considered to be an important barrier to migration to the environment. For example, distribution coefficients for americium and plutonium of 1000 and 500 L/kg, respectively, were used in a recent study which evaluated the Yucca Mountain site (Kerrisk, LA-10414-MS, October 1985). Such relatively large distribution coefficient values result in very large calculated retardation factors, and that report concluded that travel times for these radionuclides to the environment would be in the order of 10^5 to 10^7 y, or greater.

The distribution coefficients for americium and plutonium used for the Yucca Mountain evaluation are based on laboratory experiments. No field studies of radionuclide migration in the repository horizon (Topopah Spring tuff) have been reported. The results of the study of migration in Bandelier tuff given in the paper under review, however, suggest caution in the extrapolation of laboratory data to describe repository performance. Since migration of significant quantities of americium and plutonium occurred under unsaturated conditions with less than one column volume of water in the Bandelier tuff, in contrast to the high retention anticipated based on laboratory measurements, it seems possible that the laboratory data from drill core samples used in laboratory experiments to develop distribution coefficient values relative to the Yucca Mountain site could similarly overestimate the retardation to be expected in the field at Yucca Mountain.

The work with Bandelier tuff also illustrates the time and expense involved in conducting large-scale field studies. The results described in the paper cover work conducted over almost two decades. It seems possible that a similar effort could be required to conduct field-scale studies in the exploratory shaft at candidate repository sites.

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