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Long-Term Risk from Actinides in the Environment: Modes of Mobility

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Research Objective

The mobility of actinides in surface soils is a key issue of concern at several DOE facilities in arid and semiarid environments, including Rocky Flats, Hanford, Nevada Test Site, Idaho National Engineering Laboratory, and Los Alamos National Laboratory and the Waste Isolation Pilot Plant (WIPP). Key sources of uncertainty in assessing Pu mobility are the magnitudes of mobility resulting from three modes of transport: (1) wind erosion, (2) water erosion, and (3) vertical migration. Each of these three processes depend on numerous environmental factors and they compete with one another, particularly for actinides in very shallow soils (~ 1 mm). The overall goal of the study is to quantify the mobility of soil actinides from all three modes. Our study is using field measurements, laboratory experiments, and ecological modeling to address these three processes at three DOE facilities where actinide kinetics are of concern: WIPP, Rocky Flats, and Hanford. Wind erosion is being measured with suite of monitoring equipment, water erosion is being studied with rainfall simulation experiments, vertical migration is being studied in controlled laboratory experiments, and the three processes are being integrated using ecological modeling. Estimates for clean up of soil actinides for the extensive tracts of DOE land range to hundreds of billion $ in the U.S. Without studies of these processes, unnecessary clean-up of these areas may waste billions of dollars and cause irreparable ecological damage through the soil removal. Further, the outcomes of litigation against DOE are dependent on quantifying the mobility of actinides in surface soils.

Research Progress and Implications

This report provides a summary of work for the first year of a 3-year project; subcontracts to collaborating institutions (Colorado State University and New Mexico State University) were not in place until late December 1997, and hence this report focuses on the results of the 5 months from January through May 1998. Our major result to date is a review of literature on the potential for using soil concentrations of ^{137}Cs and ^{241}Am as tracers for plutonium in soil. Measurements of ^{239}Pu contamination in the environment are expensive and time consuming, requiring radiochemical analysis and alpha spectroscopy. We evaluated the literature for measurements of ^{137}Cs and ^{241}Am, both of which are more cost-effectively measured by gamma spectrometry, as tracers for Pu in soil. Our results indicate that: significant positive correlation exists between Pu, Cs, and Am in soils and sediments at several locations including Rocky Flats, Los Alamos, and Hanford; atmospheric transport of Pu and Cs from worldwide fallout is essentially the same; the attachment of Pu and Cs to soil particles of various size is very similar; both Pu and Cs movement in the environment correlate well with soil and sediment particle movements; a significant correlation between Pu, Cs, and Am was found in soil as a function of depth, indicating similar vertical migration behavior (most of the activity of these radionuclides is confined to the top 10-20 cm of soil at virtually all locations); most Pu and Cs are strongly absorbed onto clay and organic matter in soils and there is essentially very little leaching of Pu, Am and Cs through soil columns. Based on the above information, we believe that ^{137}Cs and ^{241}Am are excellent tracers for both ^{239}Pu and soil particle transport processes in clay, mineral bearing and/or organic soils. Therefore, Cs and Am would be good tracers for our proposed water erosion and vertical migration work, at least for both Rocky Flats and Hanford. The correlation between Pu and Cs may not be as strong in sandy soil (e.g. WIPP site), however, we need to examine more data.

Planned Activities

An all-investigators meeting was held at Colorado State University in January, at which time experimental designs were evaluated and tasks were identified and scheduled. Most of the field measurements that we will obtain will be during Summer months when winds are potentially high and soils are dry (expected to lead to higher erosion rates). We therefore decided to conduct field work for FY 1998 at the WIPP site, where logistics are less restrictive. The measurements for the WIPP site will actually be obtained on adjacent BLM land, where the Carlsbad Environmental Monitoring and Research Center is conducting measurements. Measurements will be obtained from Rocky Flats in Summer of 1999 and at Hanford in summer of 2000. Plots for the BLM/WIPP site have been selected and surface characteristics are being measured. An intensive period of field measurement will take place at the site in early June.

Rainfall simulation experiments will be conducted on plots with low and high amounts of vegetation cover. The low cover plots will be disturbed by recent burning, whereas the high cover plots will be undisturbed. The simulator experiments will yield hydrographs and sediment loads for both plot types as a function of ground cover and rainfall intensity. The rainfall simulator has been refurbished, tested, and transported to the field site.

Measurement of transport rates of plutonium contaminated soil through wind erosion will be made and correlated with micrometeorological, soil, and vegetative conditions. These relationships will be investigated with measurements that are averaged over longer periods and short-term, instantaneous, measurements. Longer-time averaged aerosol concentration measurements will be made using passive Bagnold type samplers (for surface creep and saltating soils) and using retrospective and cascade impactor air samplers (for suspended soils). Highly frequent aerosol measurements will be made using up to 26 optical particle counters and sizers. Meteorological conditions will be measured using a 3-dimensional sonic anemometer to measure wind velocity components at rates up to 60 Hz, wind vanes and cup anemometers, air humidity and temperature probes, and rain gauges.

Soil samples for the vertical migration studies will be collected in-tact or in representative layers in early June and returned to Colorado State University for manipulative experiments in soil columns. One set of experiments will focus on the effects of physical factors (wetting/drying cycles and freezing/thawing cycles) on vertical migration; the treatment will be soil columns without rocks, with 25% rocks, and with 75% rocks. A second set of experiments will focus on the effects of biological factors on vertical migration; treatments will include bare soil, vegetated with grass, and grass with introduced earthworms.

Results from all three studies will be integrated using a systems modeling approach. We are currently reviewing a version of the PATHWAY model that was used to evaluate actinide mobility at Rocky Flats. We are identifying processes that will need to be added to the model to compare all three pathways of concern (wind erosion, water erosion, and vertical migration) and evaluating alternative formulations of those processes.