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Title: LONG-TERM RISK FROM ACTINIDES IN THE
ENVIRONMENT: MODES OF MOBILITY

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

Mid-Year Progress Report

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SUMMARY

The overall goal of this project is to assess the relative importance of three modes of actinide mobility in semiarid environments: wind erosion, water erosion, and downward vertical migration. An understanding of the relative roles of these three processes and how they differ under varying environmental conditions is needed to assess dose associated with decreases in the concentrations of actinides in the surface soils of sites such as Rocky Flats and Hanford, where litigation over these issues is underway. Our study will focus on three sites adjacent to major DOE facilities: the Waste Isolation Pilot Plant (WIPP), Rocky Flats, and Hanford. In the past year we have focused on obtaining field measurements at WIPP. Our data indicate that water erosion is not likely to be very high due to the sandy texture of the soils. Wind erosion can be substantial and appears to be related to thresholds of wind velocity. We have made progress in developing techniques for measuring downward vertical migration in the laboratory and experiments are proceeding. Although our results are only preliminary, we believe that wind erosion is probably the most important contaminant process at WIPP. In addition, our results show how surface disturbances that decrease ground cover (implemented as a fire at near WIPP) greatly increase both wind erosion and water erosion. Recent studies on wind and water erosion indicate that relative changes in wind velocity have greater effects on changes on wind erosion than effects of relative changes in precipitation of the same magnitude on water erosion. We are evaluating models such as WEPS and EPIC for use in integrating our study results across processes and across sites.

Goals of Project:

The overall goal of this project is to assess the relative importance of three modes of actinide mobility in semiarid environments: wind erosion, water erosion, and downward vertical migration. An understanding of the relative roles of these three processes and how they differ under varying environmental conditions is needed to assess dose associated with decreases in the concentrations of actinides in the surface soils of sites such as Rocky Flats and Hanford, where litigation over these issues is underway. Our study will focus on three sites adjacent to major DOE facilities: the Waste Isolation Pilot Plant (WIPP), Rocky Flats, and Hanford. In the past year we have focused on obtaining field measurements at WIPP.

Technical Description of Work:

Our study includes measurements on undisturbed and disturbed sites. We are studying wind erosion via numerous measurements of meteorological conditions, Bagnold samplers for collection of saltating and resuspended material, and air samples for air concentrations and particle size distributions; we are studying water erosion via rainfall simulation experiments; and we are studying vertical migration via laboratory column studies.

Accomplishments to Date:

Wind Erosion

Measurements to determine resuspension rates as a function of meteorological conditions at sites near the WIPP site began in June 1998 and continued until May 1999. In addition, comparisons of dust fluxes between a vegetated site and burn site (from a 1400-acre fire that occurred the week prior to the start of the measurements) are being investigated. Both sites are east of the WIPP site by approximately 10 miles, the vegetated site in the SE direction and the burned site in the NE direction from the WIPP site. Site characterization was done by collecting soil and vegetation samples at both the burned and unburned sites. In addition, the site's aerodynamic characteristics, in terms of surface roughness and friction velocity were determined based on micrometeorological measurements using a 3-dimensional wind sensor.

Resuspension rates (R) were measured by multiplying the eddy diffusivity coefficient (K_z) by the concentration gradient with height.

$$R = K_z \frac{dC}{dz}$$

The eddy diffusivity coefficient was calculated as:

$$K_z = k_v z u_*$$

where:

k_v = von Karman's constant (assumed 0.4),

z = the measurement height (3 m), and

u_* = the friction velocity.

Finally, the friction velocity (u_*) was calculated as:

$$u_*^2 = \overline{u'w'^2} + \overline{v'w'^2} \sqrt{\quad}^{1/2}$$

where:

u' = the instantaneous wind velocity in the x-direction,

v' = the instantaneous wind velocity in the y-direction, and

w' = the instantaneous wind velocity in the z-direction.

The measured friction velocities at both the burned and unburned sites were strongly correlated to the horizontal wind velocity and were similar to those found in other studies of similar landscapes. The following relationships were found:

Burn Site: $u_* = 0.09 * (\text{horizontal wind velocity}) - 0.04$ ($r^2 = 0.93$)

Vegetated Site: $u_* = 0.11 * (\text{horizontal wind velocity}) + 0.03$ ($r^2 = 0.93$)

Concentration gradients with height (dC/dz) were measured sampling aerosol concentrations at 1 and 3 meters. The sampling inlets used were designed to sample efficiently regardless of wind direction, velocity, and particle size. Early results showed that the aerosol concentrations measured at 1 m were generally greater than those measured at 3 m. The final samples are still being analyzed and the effects of environmental conditions, such as wind velocity, on the concentration gradients are being investigated.

Particle concentrations as a function of particle size were also measured in "real-time" using laser optical counters and correlated to wind velocities. These measurements were conducted in 1-minute sampling intervals, substantially less than the weekly sample exchange for the active samplers. These measurements were designed to look at the time-resolved structure of dust fluxes. The measurements at the vegetated site suggested that a threshold wind velocity of around 7 m/s exists, that once exceeded, fluxes increased in a non-linear fashion. Interestingly, the effect was most pronounced for the larger particles ($>10 \mu\text{m}$) and a much reduced effect on the smaller particles ($<1 \mu\text{m}$). Also, we found evidence that even when the threshold velocity was exceeded, there was a delay of several minutes before the aerosol concentrations significantly increased.

Dust flux measurements were obtained at both sites using Bagnold-type samplers (six at each site). These samplers orient to wind direction and sample at six different heights ranging from 0 to 68 cm above the ground. Sampled dust was generally collected every two weeks. Preliminary results show significantly greater dust fluxes at the burn site compared to the vegetated site ($8 \pm 1 \text{ mg/d}$ at the burn site, $0.1 \pm 0.06 \text{ mg/d}$ at the vegetated site) for the summer sampling period from June 10 to September 1. Analyses to determine the time dependence of the collection rates and correlations of dust collection rates to meteorological conditions are ongoing.

Preliminary findings were presented in a poster session as part of the Undergraduate Research Semester Program at Los Alamos National Laboratory, and at a scientific colloquium at the Carlsbad Environmental Monitoring and Research Center.

Water Erosion

The water erosion component of the project at WIPP was conducted 40 miles SE of Carlsbad NM on May 30-June 12, 1998. This Chihuahuan Desert study site receives about 20 cm of precipitation per year and is covered primarily with grass species including black grama (*Bouteloua eripoda*), purple threeawn (*Aristida purpurea*) and several annual forbs that appear early in the summer. Soils at the study site were very sandy, well drained, and contained only about 5% clay. Average bulk density and hydraulic conductivity were about 1.4 g/cc and 3×10^{-4} , respectively. Little visual evidence of serious water erosion was evident around the study site. The purpose of this component of the project was to evaluate the hydrologic response of natural and disturbed plots using a large, rotating boom rainfall simulator to apply large, short duration rainstorm events.

Six plots, which were each 3.1 m x 10.7 m and an average 6% slope, consisted of two treatments (3 Natural cover and 3 Burned) were established on the study site. Plots were paired so that precipitation could be applied to two at a time at rates of 60 mm per hour. The sequence of rainfall applications were 60 mm applied over 1 hour followed by a 24 hour wait when 2 30 mm applications (at a rate of 60mm.hr) were applied sequentially to evaluate the effects of increasing antecedent soil moisture on runoff, erosion, and cesium transport in runoff. A total of 120 mm of precipitation was applied to each plot over a 2 day period.

Many measurements were made to fully characterize the physical and biological characteristics of the site as well as develop parameter estimates for use in a computer simulation runoff and erosion model. Total canopy cover on Natural plots averaged about 80% while about 70% of the ground surface was covered with organic material and the rest with exposed soil or rock. Burning the plots removed all of the canopy cover and changed ground cover by exposing a factor of 2 more bare soil because litter was reduced by a factor of two by the fire.

Results show that runoff, and fallout cesium-137 transport from all of the plots was generally very low reflecting the sandy soil and high infiltration rates. For example, none of the plots generated runoff during the first rainfall simulation event (designated as the Dry Run) when 60 mm of precipitation was added over a one hour period. During the second rainfall simulation event when 30 mm of precipitation was added during a 30 minute period (designated as the Wet Run), the Natural plots still did not produce runoff whereas 2 of the 3 Burned treatments did generate some runoff. During the last rainfall simulation event when an addition 30 mm

of precipitation was added over 30 minutes (designated as the Very Wet Run), most of the plots generated some runoff.

However, even though amounts were very low, runoff, erosion, and fallout cesium-137 transport were all clearly a function of plot treatment. Runoff from Burned treatments, for example, averaged a factor of 10 higher than plots with Natural cover. About 6% of the 120 mm total precipitation applied to Burned treatments contributed to runoff while only about 0.4% of the precipitation contributed runoff from Natural treatments.

Likewise erosion from all of the plots was very low reflecting the lack of runoff or the very low runoff velocities. However, total sediment yields from the Burned treatments was about a factor of 5 times higher than Natural treatments reflecting the loss of protective cover and the increased exposure of the soil to impacting raindrops. On a mass basis, at least 90% of the sediment leaving the plots was $>45\mu\text{m}$. Erosion on the Burned plots (after application of 120 mm of precipitation over a 2 hour period) averaged 0.1 T/ha (metric tonnes) while the rate from the Natural treatments was about 0.02 T/ha.

As expected, measured fallout cesium-137 concentrations in soils and eroding sediments were very similar. Additionally, cesium concentrations in the $<45\mu\text{m}$ size fraction of both soils and sediments averaged about 3 times higher than in the $>45\mu\text{m}$ size fraction. However due to the low mass of the smaller size fraction in both soils and eroding sediments, most of the cesium-137 inventory was associated with the coarser fractions.

Cesium loss from the plots was a function of both sediment particle size and treatment. Even though higher cesium concentrations occurred in the $<45\mu\text{m}$ size fraction, most of the cesium eroding from the plots was associated with the coarser fraction because of the large mass of this fraction. About 25% of the total cesium loss was associated with the $<45\mu\text{m}$ size fraction on the Burned treatments and 29% from the Natural treatments. In contrast, the $<45\mu\text{m}$ fraction in study plot soils comprises about 5-9% of the soil mass suggesting that an enrichment of fines in sediments has taken place.

This enrichment of fines in eroding sediments, compared to the soil, is the basis of the well known fact that many chemicals, including radionuclides, are also enriched in eroding sediments because of the preferential transport of fines during the erosion process. The enrichment ratio (i.e., the concentration of contaminant in eroding sediments relative to the soil) for fallout cesium transport in runoff at WIPP averaged 2-3 for both the Natural and Burned treatments. This compares well with enrichment ratios for plutonium of 2-5 for other locations in the U.S. Due to this enrichment, relatively more cesium was eroded from the plots than would be predicted by gross soil losses alone. The total cesium loss from the top 5 cm of WIPP site study plots averaged about 0.051% on the Burned plots and 0.004% from the Natural plots.

A sensitivity analysis of the WEPP model was undertaken to ensure that field data collection address critical information needs for using the model. This analysis showed that rainfall intensity, soil thickness, and litter on the ground surface were important determinates of the amount of runoff and erosion generated by the soil surface.

In summary, sediment and cesium loss from the WIPP study site soils by runoff and erosion was very low due to the sandy soil and high infiltration rates. Only under wet or very wet antecedent soil moisture conditions, did runoff and erosion occur. In fact, two storms totaling 90 mm of precipitation applied over 1.5 hours were required to generate any runoff from the Natural treatment demonstrating the effects of soil type and the important role vegetation plays in reducing runoff and erosion. The losses that did occur were a factor of 5-10 higher on Burn treatments than on undisturbed treatments covered with native vegetation. Hydrologic erosion rates on all plots was very low indicating that study area soils would likely be resistant to degradation by runoff events.

Vertical Migration

Measurements of vertical migration of ^{134}Cs in soil cores from the WIPP site are in progress. A ^{134}Cs -spiked WIPP soil was evenly distributed (to about 0.5 cm thickness) across the surface of each soil core. The objective is to determine the affects of physical processes on the downward migration of ^{134}Cs . The physical processes are: 1) continuous saturated flow of water; and 2) alternating periods of saturated flow and drying cycles (to promote soil cracking and increase the potential for downward migration). Our hypothesis is that any vertical migration of ^{134}Cs in the soil columns is likely to be the result of colloidal transport, or possibly the transport of larger soil constituents through macropores such as soil cracks. The ultimate goal is to use data from this portion of the study to estimate a site-specific loss rate constant for use in an equation that will model the transport of ^{134}Cs (our surrogate for ^{239}Pu) via wind and water erosion at the WIPP site. The loss rate constant

would be based on an assumed scenario of alternating periods of prolonged drought and heavy precipitation. The wind and water erosion portions of the model will be based on the respective results of those portions of the overall study at the WIPP site.

It has taken several months to obtain or construct the necessary equipment, and to develop an accurate methodology for taking both beta and gamma measurements non-destructively. Because beta emissions are readily attenuated, beta measurements from above the soil surface should be able to detect very small changes in the distribution of the spiked soil (such as downward migration). Beta measurements involve two different detectors, each of which have certain advantages / disadvantages. Measurements from both detectors will be taken throughout the study. Distance from the soil surface is critical for these measurements and thus great care is taken to set each detector at a consistent distance from the soil surface. The first detector is a pancake G-M detector with scaler-readout meter. The second is a surface barrier detector with scaler-readout meter. Although the pancake detector has much greater efficiency due to its larger size, its size limits how closely it can be placed above the soil surface and also, it is sensitive to a small percentage of gamma radiation. The surface barrier detector can be placed much closer to the soil surface and has almost no sensitivity to gamma radiation, but it is also less efficient.

Gamma measurements are taken using a BGO detector with pre-amp and multichannel analyzer. Gamma measurements involve unshielded top views of the soil surface, and side views along the outside of the column walls, with the detector housed in lead shielding constructed as a collimator (Figure 1). The collimator slit (about 4 cm wide by 3 mm tall) is oriented with its width parallel to the soil surface in order to view thin horizontal planes or slices of the soil cores. Readings at any given soil profile depth are taken on four sides of the column and averaged. The resolution of the side view measurements has proved to be excellent, discriminating changes in activity to within 1mm. We believe that between all of the methodologies we have developed to measure vertical migration, we will be able to accurately detect very small movements of ^{134}Cs , as a function of time and experimental treatments.

The nature of these soil column experiments in the lab, particularly with such large soil cores (30 cm $\times$ 20 cm diameter), is a very time consuming project. Similar column experiments by other researchers have taken up to several years to complete. We are doing everything possible to speed up the physical processes involved, but even with a special oven we have constructed, it will take many weeks or possibly even months to dry the columns enough to promote cracking to a maximum extent. At this time, thousands of measurements have been made to document initial conditions and establish initial ^{134}Cs depth distributions for both completely dry and saturated soil columns. Thus far, there are no apparent indications of movement of ^{134}Cs after initial additions of water were allowed to percolate through, and completely saturate, the soil columns. The leachate contained no activity significantly greater than background, and thus, there was no evidence of leaching of liquid phase ^{134}Cs .

Figure 1:

Additional column studies will also be conducted to investigate the efficacy of using non-radioactive tracers as analogs for investigating the dynamics of ^{137}Cs and Pu in soils in 1998. This activity is lead by New Mexico State University. Our original plans were to obtain field measurements at Rocky Flats the first year and then move to WIPP, but problems in getting permission to do the field studies at the Rocky Flats site resulted in the field work being rescheduled for the WIPP site to year 1. Therefore, much of New Mexico State's efforts to date focussed on supporting the field work for the wind and water erosion studies for the WIPP site. This included helping with the initial setup of the meteorological stations and the Bagnold erosion samplers, helping set up permanent erosion stakes at the Cactus Flats sampling area, and of a continuing task to download the meteorological data and collect the soil from the Bagnold samplers on a regular basis, identifying the location where the rainfall simulation experiment was conducted, making the local arrangements with the Bureau of Land Management for permission to conduct the experiment, arranging with the BLM to conduct the burning of three of the six plots in the experiment, locating and arranging for the delivery of a tank of 10,000 gallons of water for use in the experiment, helping find a local firm to manufacture the erosion barriers for the plots, and general support during the conduct of the experiments.

Construction of the columns needed for these studies has started. PVC pipe identical to that used for the column studies at CSU was obtained and cut to length. In addition, we located a site, owned by El Paso Gas, from which we obtained soil having very low background levels of Pu and ^{137}Cs . Soil was obtained from beneath the concrete slab of the porch of a building built prior to 1945. A layer of fill dirt was removed to the approximate level of the soil surface. This fill dirt was indistinguishable from the surrounding soil. The PVC tubes were then driven into the soil to obtain soil cores. Additional soil was excavated and placed in sealed containers. These soils are very different in texture than those nearer to the WIPP site, and their fine texture may pose a problem with using them in our columns. In particular, of approximately 6 cm of water placed on the surface of a core at the field site, approximately 4 cm remained after 6 hours. This is in sharp contrast to the soils near the WIPP site, where infiltration of water was very rapid.

Our plan is to start the soil column studies after a methodology is developed at CSU for reducing edge flow in columns subjected to freeze-thaw cycles. Space for conducting the experiments has also become an issue at the CEMRC, due to changes in the scope of our WIPP related work. However, we have a plan in place that will provide us with space in our sample preparation area later this summer. We also plan to collect soil columns from areas more typical off the WIPP site, which should provide a significant contrast to the fine texture soils that we have currently.

Modeling

Our efforts in the past year have focused primarily on obtaining field data that can be used parameterize or test model predictions. We are in the process of evaluating different codes for use in integrating the modeling work. These include WEPS, EPIC, WEPP, and KINEROS. These models contain tradeoffs with respect to how wind erosion vs. water erosion is modeled; none of the models contained detailed mechanisms for both processes and none of the models contains a component for downward vertical migration. Hence, we will probably integrate these codes in a framework such as TIME-ZERO and add components for downward vertical migration. In addition, we will evaluate alternative modes of implementing surface disturbances. Recent sensitivity studies of EPIC indicate that relative changes in wind velocity have greater effects on changes on wind erosion than effects of relative changes in precipitation of the same magnitude on water erosion. Our field data indicate that wind and water erosion will be highly sensitive to changes and ground cover, and these recent studies indicate that variations in climatic conditions will also be important, particularly for wind erosion. In addition, we have located several studies on erosion from the Jornada site in southern New Mexico, which is near WIPP, that may provide valuable data for model testing.

Projections:

In June we will initiate field activities at a site adjacent to Rocky Flats and expect to collect data there during the remainder of the fiscal year and completing the scope of work planned for this year.

Funding:

Spending levels have been steady throughout the fiscal year and should remain on track through the remainder of the fiscal year, allowing the scope for this year to be completed within budget.

Issues/Problems and Corrective Actions:

Impediments to our progress on this project have included (1) a several month delay during the initial year in implementing the subcontracts to Colorado State University and New Mexico State Universities, which required that we reorder our study sites in order to get field data the first year, and (2) lack of access to Rocky Flats for our field measurements, which we have addressed by gaining access to land adjacent to Rocky Flats.