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

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Executive Summary

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

> *"There is scientific uncertainty about the levels of risk to human health and the environment at the end stages of the DOE cleanup effort. Accurate risk analyses require thorough knowledge of ... basic ecological processes and principles (and) rates at which contaminants move through ecosystems...In particular, better knowledge of ... transport dynamics (is needed) to assist DOE in protecting the public, workers, and the environment." (EMSP 2000 Science Priorities, Health/Ecology/Risk Category).*

The mobility of actinides in surface soils is the major driver of risks to human health and the environment for DOE facilities in arid and semiarid environments. Understanding actinide mobility is an EMSP priority in the category of Health/Ecology/Risk for addressing numerous and extensively contaminated DOE sites. Uncertainty in actinide mobility is the fundamental driver for remediation decisions, litigation issues, and long-term stewardship strategies. Indeed, actinide mobility is already a high-visibility issue at Rocky Flats and Hanford, upon which current litigation and clean-up decisions are pending, and public groups are increasingly concerned about changes resulting from environmental disturbances such as fire that increase mobility. Similar issues exist at other DOE facilities in arid/semiarid environments (i.e., Nevada, Idaho, Los Alamos, Pantex, and WIPP). At all of these facilities, uncertainty in actinide mobility hinges on the relative roles among three modes of transport: (1) wind erosion, (2) water erosion, and (3) vertical migration, each of which depends on several interrelated environmental factors. The objective of our study was to quantify the mobility of soil actinides from all three modes using a combination of field studies, laboratory studies, and modeling for several sites. We have developed a data set that quantifies actinide mobility by the three processes and have integrated our results by developing initial assessment tools that will provide the basis for incorporating strong technical information into the cleanup decision-making process. Our goal is to provide

DOE with data and tools that improve risk assessments, cut cleanup costs, and facilitate technology transfer. Our accomplishments to date include development of advanced measurement techniques for assessing the three pathways; new data on each pathway based on site-specific field and laboratory measurements at three major DOE facilities; and multi-pathway, multi-site assessments of mobility based on existing long-term transport models. Our measurement and assessment results point the way to future payoffs for DOE. First, estimates from equilibrium-type models indicate that the relative importance of actinide transport by wind erosion, water erosion, and vertical migration differ among and between sites and that these differences exceed more than an order of magnitude within a site and across sites. These results can be used to prioritize efforts to improve risk assessments and remediation and are some of the first multi-pathway, multi-site estimates for DOE facilities. Second, field studies demonstrate that disturbances that reduce ground cover, such as fire or heavy grazing, can increase wind and water erosion by more than two orders of magnitude. These results demonstrate the need to factor in disturbance events and recovery rates into long-term assessment of actinide mobility. Third, all three pathways are driven by low frequency, extreme climatic and disturbance events that greatly increase transport rates relative to long-term averages. These results highlight the need to account for extremes in climate and disturbance that occur on a finer time-scale but may contribute most to long-term risks. Improved risk assessment for addressing remediation, litigation, and long-term stewardship will require a more mechanistic understanding and predictive capability for low frequency, high impact events.

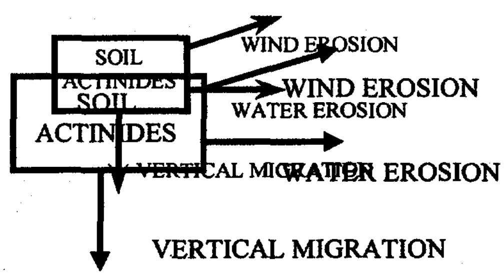
1. Research Objectives

The mobility of actinides in surface soils remains a key issue of concern at several DOE facilities in arid and semiarid environments, including Rocky Flats, Hanford, Nevada, Idaho, and Los Alamos. Over the last 50 years, nuclear research and development programs have resulted in releases of plutonium to both on-site and off-site locations. Most of this plutonium and other actinides are currently in soils where it is tightly bound to soil particles (Watters et al. 1983), but these particles themselves are subject to redistribution. Research indicates that actinide redistribution is driven primarily by physical and biological processes associated with ecosystem dynamics, rather than by chemical processes.

Actinide mobility is a high visibility issue at Rocky Flats and Hanford due to pending litigation and clean-up decisions. The potential for redistribution has lead plaintiff groups to sue the DOE and its contractors at Rocky Flats and Hanford by claiming that past releases of plutonium have occurred and that these releases have exposed off-site human populations to large amounts of plutonium with consequent negative health risks to humans (Goble 1996). Plaintiffs also claim that these exposures will continue to occur because of chronic releases from contaminated soil from on-site sources (Goble 1996, Smallwood 1996a, b). The concern about past and potential releases of contaminants from Rocky Flats appears to have reduced property values in nearby communities (Flynn et al. 1998). In addition, public groups focusing on past and potential off-site transport from Rocky Flats are expressing concern about increased erosion-driven transport associated with disturbances such as fire.

A central issue in arguing these cases has been whether the plutonium presently in soils is immobile (Litaor et al 1996) or whether it is subject to transport by biological, physical, or chemical processes. Plaintiffs claim that wind erosion of soil is producing in large chronic releases of plutonium to off-site locations (Goble 1996, Smallwood 1996a, b). While past and current air and soil monitoring data would suggest that these releases are very unlikely (Price 1989, Dirkes and Hanf 1996), definitive scientific data are not available to defend a position on the long-term spatial changes in the plutonium distribution in soils due to biological, physical, or chemical processes, including effects of site disturbances. Past data reflect site management practices (e.g., fire suppression, erosion control) and climatic conditions that may not be relevant to sites in the future (Swetnam et al. 1999). There is now evidence that climate change is likely to impact the return frequencies of extreme events such as droughts, fires, and floods, in many areas (Olsen et al. 1998).

The key source of uncertainty in assessing actinide mobility is the relative importance of transport by: (1) wind erosion, (2) water erosion, and (3) vertical migration:



Each of these three processes depends on several environmental factors and they compete with one another. A scientific assessment of the long-term risks associated with actinides in surface soils depends on better quantifying each of these three modes of mobility. ***The objective from our EMSP study was to quantify the mobility of soil actinides by wind erosion, water erosion, and vertical migration at three semiarid sites where actinide mobility is a key technical, social and legal issue.***

Our EMSP project was the first to evaluate all three factors at a site. Our approach has been to investigate both short- and long-term issues based on field and lab studies and model comparisons. Our results demonstrate the importance of incorporating threshold responses into a modeling framework that accounts for environmental factors and natural disturbances that trigger large changes in actinide mobility.

Our project directly addressed EMSP Science needs in Health, Ecology and Risk:

> *"There is scientific uncertainty about the levels of risk to human health and the environment at the end stages of the DOE cleanup effort. Accurate risk analyses require thorough knowledge of contaminant characteristics, basic ecological processes and principles, (and) rates at which contaminants move through ecosystems...In particular, better knowledge of radionuclide and toxic chemical transport dynamics is needed to assist DOE in protecting the public, workers, and the environment."*

In addition, our work is relevant to stated EMSP priorities in Environmental Restoration:

> *" Fundamental improvements are needed in the abilities to ...assess data, and to predict the movement and fate of contaminants. New materials and designs must be developed and proven for caps and covers for buried waste and closed facilities in order to develop robust systems to insure waste isolation over the range of climate conditions and extremes. Better understanding (is needed) of vadose zone processes which are complicated by the possibility of multi-phase flow...by competition between fluids for wetting of surfaces, by effects of alternating wet and dry periods and other factors. Basic data or methods (are needed) to support long-term decisions and to understand processes that affect the validity of remediation decisions and long-term effectiveness of remediation alternatives.*

Further, our proposal provides tools for addressing priorities in Deactivation and Decommissioning, particularly with respect to developing effective methodologies for large environmental sites.

The vast majority of actinide contaminants at DOE sites consist of low concentrations of radioactivity sequestered in soils. The first important question to ask about sites with extensive actinide contamination is not how to clean them up, but rather whether or not to clean them up at all. One of the major options being considered—physical removal of the soils—is costly and requires both virtual destruction of the contaminated ecosystem and the availability of a new site licensed to dispose of the contaminated materials that are removed. Soil substrate removal only translocates the problem and may add significant health risks to the clean-up workers. In contrast, if the long-term risk from actinides in surface soils of the environment is sufficiently low, contaminants may be left in place. However, leaving the contaminants in place requires that our assessments of this cleanup option take into account low frequency, high impact disturbances that change thresholds between significant and non-significant transport of soil contaminants. Until sound technical data and knowledge are available to accurately address long term fate and effects of soil actinides, public, scientific, and regulatory confidence in DOE cleanup decisions will be limited.

Soil actinide transport by wind erosion, water erosion, and vertical migration affects cleanup decisions at nearly all DOE sites in arid/semiarid environments, including Rocky Flats, Hanford, Idaho, Los Alamos, and Pantex, as well as potential future contamination at the WIPP facility near Carlsbad. There are few peer-reviewed journal publications of risk assessments at these any of these sites that take into account all three modes of transport. Further, to our knowledge no one has ever addressed the impact of the linkages between these processes in affecting long-term actinide fate and risks, or the effects of low frequency high impact disturbances on changing the rates of transport. Credible assessments must address these issues and heighten stakeholder confidence in cleanup decisions.

2. Methods and Results

2.1. Overview

We are developing a data set on wind erosion, water erosion, and vertical migration though an integrated set of field, laboratory, and modeling studies. Wind erosion rates are being quantified using spatially-distributed aerosol measurements, including finely time-resolved measurements, and correlated with meteorological and groundcover conditions. Water erosion rates are being quantified using rainfall simulator experiments in the field and a large number of ancillary measurements to characterize the sites and to estimate horizontal changes in fallout cesium-137 distribution an analogue for actinide mobility. Vertical migration is being studied in the laboratory to quantify the effects of soil water content and wetting-drying cycles. These studies are conducted in soil columns that were collected in situ from the field. Our initial studies focus on WIPP, Rocky Flats, and Hanford, three semiarid DOE sites that differ in climate, soils, vegetation, and actinide sources. The relative importance of the three pathways is also being evaluated using existing models that predict average long-term responses for a total of seven DOE sites in arid and semiarid sites (the three sites where we are collecting data plus four other sites). Data for the three field sites have been used to parameterize and run models and to compare predictions with measured field data.

Our studies (Breshears, et al. 1998, 1999, Breshears 2000) demonstrate three key ways in which risk assessments can be improved. (1) Estimates of transport based on both field studies and equilibrium-type models for average long-term responses indicate that the importance of wind erosion, water erosion, and vertical migration can differ by more than an order of magnitude within and across sites. These results provide a rationale for prioritizing efforts to improve risk assessments, remediation, and long-term stewardship. To our knowledge, they are also the first multi-pathway, multi-site estimates for long-term mobility of surface actinides at arid and semiarid DOE facilities. (2) Field studies demonstrate that disturbances that reduce ground cover, such as fire or heavy grazing, can increase wind and water erosion by more than two orders of magnitude—these results demonstrate the need to factor in low frequency, high impact disturbance events into long-term assessment of actinide mobility. This issue has largely been ignored to date. (3) Wind, water, and vertical transport are all driven by very short phase, low frequency disturbance effects events that greatly increase transport rates—these results highlight the need to include finer time-scale events in determining long-term risks. The effects of these short events largely have been masked to date by longer-term averages.

An important theme emerging from this project is that each of the three pathways exhibits threshold responses to a suite of environmental conditions, resulting in non-linear increases in contaminant transport. By environmental conditions, we mean factors such as precipitation intensity, wind velocity, wetting-drying cycles, surface heterogeneity (vegetation and ground cover), and disturbances that impact the surface heterogeneity. These results highlight the importance of finer-scale processes that could dominant the overall risk estimates associated with the long-term mobility of actinides. Yet these processes are not generally evaluated in concert and their threshold response is largely unrecognized and/or overlooked. *Improved risk assessment for addressing remediation, litigation, and long-term stewardship will require a more mechanistic understanding and predictive capability of these processes.* Our work contributes to improved risk assessment at arid and semiarid DOE facilities.

2.2. Analogues for Actinide Mobility

Measurement of low-levels of ^{239}Pu contamination in the environment requires radiochemical analysis and alpha spectrometry, which are expensive, time consuming and destructive. In contrast, gamma spectrometry is cost-effective, rapid, and can measure in situ activity levels. We reviewed the literature to determine if ^{137}Cs and ^{241}Am, two raidonuclides readily measured via gamma spectrometry counting, could be used as tracers for Pu in soil

We found the following: (i) significant positive correlation exists between Pu, Cs and Am levels in soil and sediment at several locations, including Rocky Flats, Los Alamos, and Hanford; (ii) atmospheric transport of Pu and Cs from worldwide fallout is essentially the same; (iii) the preferential attachment of Pu and Cs to soil particles of various size is very similar; (iv) both Pu and Cs movement in the environment correlate well with soil and sediment particle movements; (v) significant correlation between Pu, Cs and Am exists as a function of soil depth, indicating similar vertical migration behavior, (vi) most of the activity of these radionuclides is confined to the top 10-20 cm of

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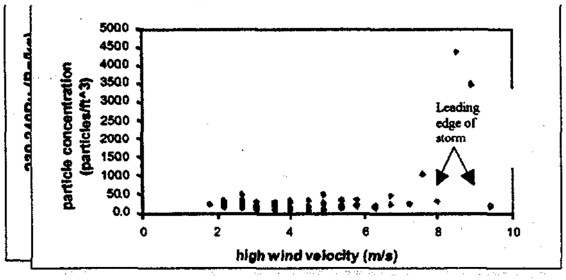


virtually all locations; (vii) most Pu, Am and Cs strongly absorbed onto clay, minerals and matter in soil and there is essentially very little of Pu, Am and Cs through soil columns; and distribution coefficients K_d for Pu and Cs are and in the range of 10^4 to 10^5.

In addition, soil samples recently collected by CEMRC for monitoring studies at WIPP also showed strong correlations between ^{137}Cs and both ^{239,240}Pu (r=0.97, n=96) and ^{241}Am (r=0.74, n=72) (CEMRC 2000). Soil texture was found to explain much of the variability in activity concentrations of fallout raidonuclides even within the narrow range of soil textures (sand or loamy sand) found at WIPP. The CEMRC studies also showed that the concentrations of the fallout radionuclides were strongly correlated with concentrations of Pb, another anthropogenic contaminant, and Al, a primary component of many clays.

Based on the information above, we believe that ^{137}Cs and ^{241}Am are excellent tracers for ^{239}Pu and that their transport is closely associated with the behavior of small particles in soils. We have applied this finding to our studies of wind erosion, water erosion and vertical migration.

2.3. Wind Erosion

Wind erosion is often estimated in risk assessments using resuspension factors reported in the literature. Yet resuspension factors can vary over several orders of magnitude. Hence, an average value has a larger degree of uncertainty and may not be accurate for other DOE sites.

We have obtained wind erosion measurements at representative locations for WIPP and Rocky Flats. The effect of disturbances in vegetation cover was investigated at each site by simultaneously measuring control and disturbed sites (a recently burned area for WIPP and heavily grazed pasture for Rocky Flats). We measured resuspension rates and micrometeorological conditions over time intervals that were both short (minutes to hours) and long (weeks to months). We measured (i) resuspension rates using the meteorological flux gradient method, (ii) aerosol concentration using total particulate samplers and optical particle counters positioned at different heights, (iii) eddy diffusivity coefficients using sonic anemometer data, and (iv) saltation rates using Bagnold-type samplers. The optical particle counters, which provided near real-time measurements of aerosol concentrations stratified by particle size area, are typically used for indoor aerosol studies (Whicker et al 1997). We are among the first to apply these sensitive indoor instruments for environmental measurements. We also applied a new aerosol sampling inlet, originally developed for non-radiological purposes, for detection of wind erosion spikes. Our data suggest that this inlet is far more sensitive in detecting spikes of resuspension than conventional inlets.

Our results show that episodic, high-wind events disproportionately increase resuspension. In fact, we found that the mass of resuspended soil during windstorms can be many orders of magnitude greater than that under average wind conditions. This result agrees with other studies that found thresholds and non-linear relationships between air concentrations and wind velocity or friction velocity (Anspaugh et al. 1975, Cahill et al. 1996). Our short-term measurements at WIPP suggest that a threshold in resuspension rates is triggered when wind velocities exceed 7m s^{-1}, and that this trigger is dependent on particle size. Concentrations of 5 µm particles increased by more than an order of magnitude after this threshold was exceeded (Whicker et al. 1999, 2000). Particle size dependencies in resuspension rate, which also have been observed in other studies (Garger et al 1998), could have important implications in risk estimation.

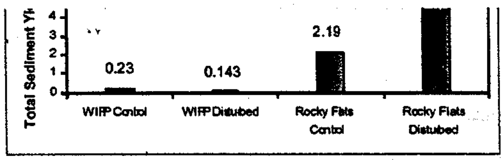
We also found evidence of the importance of wind gusts on long-term resuspension. Total resuspension from longer intervals (weeks) was correlated more highly with peak gust wind velocities ($r^2 = 0.52$) than with averaged weekly wind velocities, whether expressed linearly (r^2=0.27) or as a power function ($r^2 = 0.21$). Our results are significant in that current resuspension data is often limited to longer-term averages that can mask extreme episodic wind events.

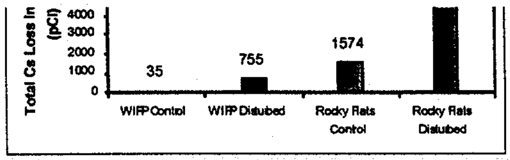
Our results yield strong evidence that wind erosion rates from disturbed sites are significantly greater than those from undisturbed sites, and that the difference can be dramatic. For example, saltation rates over the first months after the burn near WIPP (measured with Bagnold-type samplers) were about two orders of magnitude greater than those at the control site. In addition, measurements at WIPP of friction velocities and roughness lengths—key descriptors of wind shear characteristics near the ground—were significantly less at the disturbed site, relative to the control site, indicating the greater potential for resuspension at the disturbed site (Whicker et al. 1999). The control site had an the average friction velocity (normalized to a wind velocity of 3 m s^{-1}) of 0.35 m s^{-1} and a roughness length of 9 cm, whereas the disturbed site had a normalized friction velocity of 0.23 m s^{-1} and a roughness length of 2 cm. Measurement and analysis of these variables for Rocky Flats and Hanford are underway. Preliminary results from Rocky Flats also suggest that erosion rates are greater at the disturbed (heavily grazed) site. Collectively, these results demonstrate the importance of low-frequency thresholds and disturbances in ground cover in determining actinide transport by wind erosion. In addition, they provide site-specific data needed for ongoing risk assessments.

2.4. Water Erosion

Water erosion is a second major processes that effects contaminant transport, and yet major knowledge gaps exist, particularly site-specific data for semiarid DOE sites. Intense convection thunderstorms often play a major role in generating runoff and erosion in these environments. Key factors governing runoff, erosion, and associated transport of actinides are the amount and intensity of rainfall, soil type, vegetation and groundcover, and surface slope (Lane et. al., 1983). Past research has shown that vegetation and ground cover are especially important in mediating runoff, erosion, and radionuclide transport (Essington and Ronmey, 1986). Disturbances that remove surface cover, such those associated with fire, can dramatically increase soil erosion and the transport of soil contaminants (Gonzales, et. al., 1994). However, there is a lack of quantitative data on the hydrologic response of disturbed vs. undisturbed sites as it affects the fate of soil actinides at DOE sites.

Our studies measured erosional losses of sediment and fallout cesium (an actinide analogue) from field plots located near WIPP in 1998 and Rocky Flats in 1999 (Hanford studies are scheduled for summer 2000). At each site a 15.6 m diameter rotating boom rainfall simulator was used at each site to apply 60mm/hr rainstorms to six plots consisting of two treatments (Simanton et al., 1986). Three of the plots were burned to remove vegetation and litter while the remaining three were left unburned as control plots. We characterized key factors governing erosion and actinide mobility for each plot including soil types, vegetation covers, and hydrologic variables. For both disturbed (burned) and control sites, we estimated erosion and Cs-137 migration rates for each plot at three different levels of initial soil water content. In addition, for each plot we quantified the enrichment ratio—an index of radionuclide transport differences associated with the preferential mobility of fine soil particles. Fine soil particles usually have much higher radionuclide concentrations than coarser particles (Lane and Hakonson, 1982), and the enrichment ratio is used to define the ratio of fine soil particles and Cs-137 in eroding soil compared to the underlying soil. In





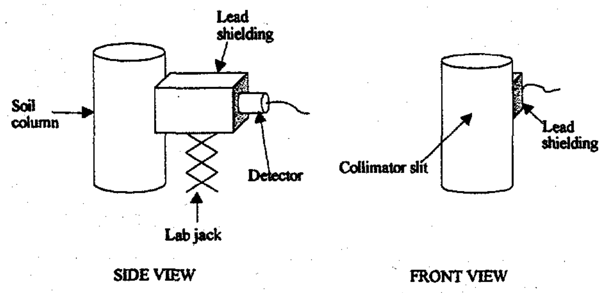
addition, we quantified key parameters and transfer rates for initializing and validating models used to predict actinide fate and effects.

Our results to date highlight the large effect of burning as a disturbance on contaminant transport and mobility via runoff and erosion. Average erosion rates at both WIPP and Rocky Flats were about three times higher from disturbed than from control plots. Cesium-137 yields from disturbed plots exceeded that from paired control plots by a factor of 3 to 13 Further, the enrichment ratios of disturbed plots (1.4 with SD=0.1 for Rocky Flats, 3.5 with SD=2.8 for WIPP) exceeded that of control plots at the same site (1.2 with SD =0.3 for REFTS, 2.1 with SD =2.1), although these differences associated with these small samples sizes were not statistically significant. Soil texture also had a pronounced effect on runoff and erosion. Relative to the Rocky Flats site, very little erosion was measured from either the disturbed or control treatments at WIPP. WIPP soils had a much higher sand content in contrast to those at Rocky Flats. In summary, our results to date show that runoff, erosion, and actinide transport are (1) strongly site specific—differences in radionuclide transport between WIPP and Rocky Flats differed by a factor of twelve because of soil and vegetation differences, and (2) are strongly impacted by disturbances such as fire, which can increase runoff, erosion, and actinide transport by more than an order of magnitude.

2.5. Vertical Migration

Vertical migration is an important potential mechanism for loss of surface actinides that could also lead to groundwater contamination. We are using ^{134}Cs tracer as an actinide analogue in laboratory studies designed to develop site-specific loss rates for actinide transport models. Vertical migration is being investigated as a function of soil type, water percolation, and wetting / drying cycles that promote macropores (cracks) in soil profiles. Because Pu solubility under normal environmental conditions is known to be extremely low, our hypothesis is that any vertical migration of Pu is likely to result from either the downward transport of soil particles through macropores in the profile, or from the transport of colloidal materials through the soil matrix itself. A recent article by Hascke et al. (2000) suggested that although Pu could be more mobile in an oxidizing surface environment, its movement in soil is likely facilitated by colloidal transport. This reinforces the need to understand the role of colloidal transport in surface and near-surface environments and explicitly include such transport in risk models.

We developed a new measurement system for studying vertical migration. Existing technology was insufficient for measuring vertical migration in soil columns with high spatial resolution. Hence we constructed, developed, and are in the process of testing a non-destructive methodology using both beta and gamma measurements. Our measurement system involves use of two complimentary beta detectors: a pancake G-M detector and a surface barrier detector The pancake detector has much greater efficiency due to its larger size, but its size limits how closely it can be placed above the soil surface and it is sensitive to gamma radiation. The surface barrier detector is less efficient but can be placed much closer to the soil surface and has almost no sensitivity to gamma radiation. In addition, gamma measurements with a BGO detector are used to obtain unshielded top views of the soil surface and side views along the outside of the soil column walls. The gamma detector has a lead shielded collimator allowing measurements of thin horizontal planes or slices of the soil cores 4 cm wide by 3 mm tall. The resolution of the side view measurements is excellent, discriminating changes in activity to within 1mm. With the methodologies we developed, we will be able to accurately detect very small movements of ^{134}Cs as a function of time and experimental treatments; we will verify the resolution of this technique by destructively sampling soil columns following completion of in situ soil column measurements.



In situ counting system developed for the EMSP project to monitor the mobility of ^{134}Cs in soil columns.

We collected intact soil cores (20 cm diameter x 30 cm depth) from field sites (WIPP and Rocky Flats to date) for use in the lab studies. Additional surface soils obtained near our study sites were irreversibly labeled with ^{134}Cs and evenly distributed on the surface of soil cores. Changes in ^{134}Cs depth profiles as a function of wetting or wetting / drying cycles are being monitored from outside the walls of the columns (in-situ) with our detector. Prior to subjecting soil columns to wetting/drying cycles to induce soil cracking we quantified initial distributions of ^{134}Cs. To minimize ^{134}Cs migration through cracks along the edges of the soil column, a latex foam sealant was injected into any cracks formed between the soil and the column walls after each drying cycle. Soil columns were subjected to a series of wetting/drying cycles in a drying oven designed to allow for the soil columns and detection system. Our treatment can induce soil cracking in a period of weeks to months, thereby accelerating this process relative to field conditions, which may produce soil cracking only under limited environmental conditions during each year. We also verified that Cs was not leached from the columns during soil wetting.

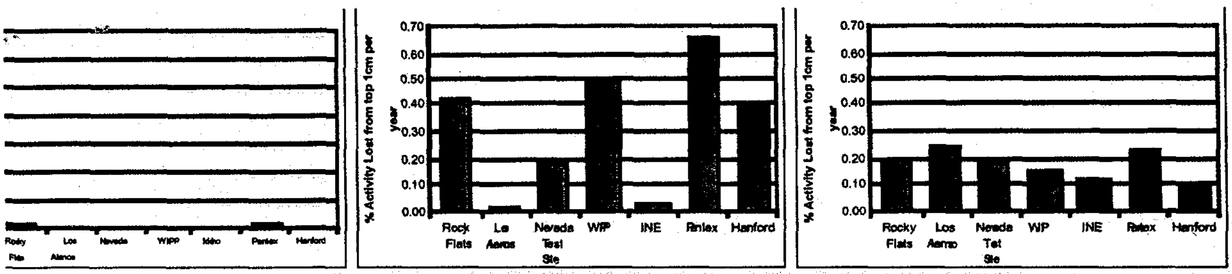
Soil cracking has been far more pronounced in the soils from Rocky Flats soils than those from WIPP. Extensive networks of large cracks (up to 0.5 cm in width) have developed when drying Rocky Flats soils. This difference is likely due to the much higher clay content in Rocky Flats soils. The vertical migration studies demonstrate trends of downward migration in both treated and control soil columns. Our data suggest increased migration in soils subjected to wetting/drying. In addition, vertical migration appears to be greater in the treated soils from Rocky Flats relative to WIPP; again, this likely relates to differences in soil texture between sites. Collectively, these results highlight the impact of low frequency events such as soil cracking on vertical migration.

2.6. Modeling

We used existing models of long-term transport to conduct a preliminary assessment for each of the three modes of transport: RUSLE for water erosion, RWEQ for wind erosion, and the radionuclide leaching component of RESRAD for vertical migration. The quantity of contaminants transported depends on many environmental factors including ground cover, soil texture, rainfall, soil permeability, slope, and contaminant type. These factors vary greatly between DOE's arid and semiarid facilities, and hence in the dominant mode of transport and the magnitude of transport rates.

The preliminary assessment for the three pathways was carried out for seven DOE sites (Los Alamos, Hanford, Pantex, WIPP, Idaho, Rocky Flats, and Nevada). To compare the relative importance of the three components, we assumed 1 Bq/m^2 of the contaminant was in the top .01 m of soil (bulk density of 1.5 g/cm^3). The resulting values (fraction of contaminant lost per year) vary in magnitude within the individual sites as well as between the different sites. The results showed that Pantex has the greatest long-term potential for migration due to wind and water erosion and Los Alamos has the greatest long-term potential for migration due to vertical infiltration. The models used for this assessment are designed to estimate long-term averages at a coarse spatial scale. However, because the majority of contaminants redistribute during brief events, spatially and temporally coupled estimates at finer time scales and smaller spatial scales are needed.

A sensitivity analysis was completed using Crystal Ball for each of the three pathways. Water erosion is most sensitive slope angle of the land and soil texture, wind erosion is most sensitive to range vegetation type and



amount, and vertical migration is most sensitive to soil texture and land contour. The results of this preliminary assessment and sensitivity analysis allows for a more focused effort on research efforts and cleanup of areas contaminated with radionuclides, thus saving time and money.

We also developed a transport model for simulating dynamic contaminant transport in soils. The model has an object-oriented design that provides a "structurally adaptive" framework for creating code in which the soil is represented by a matrix of adjoining soil columns with each column subdivided into layers. Ordinary differential equations are used to describe the vertical and horizontal movement of contaminants in the soil system. The framework handles all bookkeeping aspects of solving a system of simultaneous differential equations, data input and output, and management of sampling stochastic parameters. Specific rate equations are derived from a generic class and attached to the soil compartments. With this design it is easy to modify the form of the rate equation used to represent a transport process and add new transport pathways. Because these equations are derived from a generic "template" class for rate equations it is even feasible to design a model in which the rate equations are dynamically assigned as the simulation runs. For example, it would be possible to add rate equations for representing infrequent but significant events, such as floods or fire, for short intervals of simulated time, and then later remove them and revert to equations that better represent the typical dynamics of contaminants in the system. In this way the model can be easily customized for site specific processes.

The model's design enables the user to define the dimensionality of the model at run time. Currently, the vertical dimension is defined by n_l layers. The rate of vertical transport is defined as a gradient-controlled rate of dispersion, i.e.

where r is the rate constant for vertical transport and L_i is the concentration in layer L. The model permits the users to define the spatial extent as a grid of n_x x n_y soil columns. Surface transport between columns is defined by a set of linear, donor controlled rate equations between a column and all adjoining columns. The rate constant for each equation can be defined independently, thus providing a mechanism for representing differential flow due to factors such as slope and aspect. This code provides the framework for proposed future modeling efforts that will fully couple all three pathways and incorporate low frequency, high impact contaminant transport events.

2.7. Other Related Recent Research

Our efforts on the EMSP project leverage off and interrelate to other ongoing research that we have been conducting. In particular, we have published recent relevant work on contaminant inventories and dynamics at Rocky Flats, on aerosol measurement and on ecosystem dynamics and erosion thresholds.

In studies of radionuclide inventories at and near Rocky Flats, we found that Pu and Am concentrations decrease rapidly with soil depth at all undisturbed locations, that the rate of decrease is essentially independent of location and that 98% of the activity resides in the upper 21 cm of soil (Webb et al., 1997; Ibrahim et al., 1996). The primary plume of contamination extends nearly due east of the contaminated 903 Pad and decreases rapidly with distance from the pad. Both Pu and Am decrease at similar rates in the soil profiles and were strongly correlated at all locations. The total Pu attributable to Rocky Flats that could be accounted for in the primary study domain (a 60 - 120° true arc from 0.2 - 19 Km from the 903 Pad) was about 50 GBq with about 95% of this inventory residing on DOE property between the 903 Pad and Indiana Street. The Pu from Rocky Flats relative to that from global fallout may have extended to ≥ 19 km off-site in the eastly direction (Ibrahim et al., 1997). Because of the consistency between Pu and Am, we inferred that approximately 10 GBq of 241Am exists within our study domain, with about 95% of the inventory to be the Rocky Flats on-site. Comparisons with earlier studies (Little and Whicker, 1978) suggest that the Pu depth profile has changed very little in 25 years. We also measured the geochemical association of Pu in selected mineralogical and chemical phases of Rocky Flats soil (Litaor and Ibrahim, 1996). In the surface soil horizons, Pu was primarily associated with the organic carbon (45 - 65%), sesquioxides (20 - 40%), and the silicate fraction (10 - 15%). A small portion of Pu was found in the soluble or exchangeable fractions (less than 1%). Our results suggest that under normal pH and oxic conditions, relatively little Pu is available for geochemically induced transport processes. Gamma measurements were also used to estimate ^{137}Cs concentrations in soil profiles around Rocky Flats due to global fallout (Hulse et al., 1999). Soil concentrations of ^{137}Cs typically decreased with depth at nearly an exponential rate and the levels correlated strongly with both Pu and Am from the same area. This information supports a physical transport mechanism for the movement of all these radionuclides with soil particles and reinforce our finding that Cs and/or Am can be used to estimate Pu mobility in soil.

We have also been improving the state of the art on measuring aerosols. Traditional EPA methods for aerosol collection suffer from significant dependence of collection efficiency on direction and velocity of the wind and particle size (Wedding et al. 1977; Garland and Nicholson 1991). For this EMSP project, we used the inlet design described in Liu and Pui (1981) to construct new sampling inlets. These inlets are being tested in a wind tunnel this year and the results could have a significant impact on how DOE sites collect environmental air samples.

In addition, we have quantified disturbance-induced changes in vegetation and related them to erosion thresholds. Work at Los Alamos National Laboratory and on adjacent lands has quantified the most rapid landscape-scale shift of a woody ecotone ever documented. During the 1950s, the ecotone (boundary) between semiarid ponderosa pine forest and piñon-juniper woodland shifted extensively (2 km or more) and rapidly (less than 5 years), through mortality of ponderosa pines in response to a severe drought (Allen and Breshears 1998). This shift has persisted for 40 years. Forest patches within the shift zone became much more fragmented, and soil erosion greatly accelerated (Davenport et al. 1998). The rapidity and the complex dynamics of the persistent shift point to the critical importance of considering ecosystem dynamics in assessing contaminant mobility. The spatial heterogeneity in ground cover fundamentally affects this threshold. Runoff and erosion rates differ significantly at the scale of vegetation patches (beneath tree canopies, in grassy patches, and in bare patches —Reid et al. 1999), and degree of storage within and connectivity between these patches is thought to be a key determinant of erosion thresholds (Davenport et al. 1998; Reid et al. 1999). Land management and climate both can impact the proportions of these different patch types (Breshears and Barnes 1999) and thereby influence hillslope runoff, erosion, and contaminant transport. Vegetation changes and associated changes in erosion highlighted here need to be factored into risk assessments in the future.

3. Relevancy, Impact, and Technology Transfer

Quantifying the thresholds that determine changes in the rates of soil actinides transport from wind erosion, water erosion, and vertical migration over longer time frames provides the basis for a scientifically defensible risk assessment. Such an assessment may justify leaving the contaminants in place, thereby saving DOE billions of dollars. At a minimum, a sound technical basis for cleanup decisions will certainly increase stakeholder confidence in selected strategies and build confidence in plans for long term stewardship of contaminated lands. Table 1 indicates the many ways in which this project contributed to improved risk assessment, potential cost reductions, and technology transfer.

Table 1. Accomplishments and benefits to DOE to date from the current EMSP project.

Accomplishments	Improved Risk Assessment	Potential Cost Savings	Technology Transfer
Determination of use of Cs and Am distributions in soil as an **analogues for Pu transport** in semiarid environments	Uncertainty can be reduced through greater sample sizes and tying to previous work on wind erosion instead of directly measuring Pu	Analyses for Cs concentrations and sediment loads costs less than 1/10 that for Pu	Presentations: EMSP 1998, EMSP 2000, and Health Physics 2000 Symposium †
Application of a **new aerosol sampling inlet** for environmental monitoring of radionuclides	This sampling inlet could provide for more accurate aerosol sampling given variations in wind speed and direction	Improved risk estimates can save costs related to monitoring, remediation, and litigation	Results compared with sampling techniques used at DOE sites (WIPP and Rocky Flats). †
Application of **optical particle counters** for outdoor environmental measures	This sampling system detects critical wind erosion spikes that are damped out by existing technology	Improved risk estimates can save costs related to monitoring, remediation, and litigation	Improved risk estimates can save costs related to monitoring, remediation, and litigation
Development of **advanced techniques for measuring vertical migration** in soil columns using ^{134}Cs tracer.	Allows for process-based studies such as effects of wetting-drying cycles on vertical migration that are not feasible in the field	Improved risk estimates can save costs related to monitoring, remediation, and litigation	Presentations: EMSP 1998, EMSP 2000, and Health Physics 2000 Symposium †
Application of **erosion bridge measurements** for quantifying the dynamics of soil micro-topography	Allows estimation of long-term (months to years) dynamics in soil microtopography	Extremely low cost approach for measuring long-term (months to years) soil micro-topography	Process has been incorporated into DOE-funded monitoring effort; presentation to CEMRC / DOE in 1999; scheduled for EMSP 2000 and Health Physics 2000 Symposium †
Development of **object-oriented modeling core** to allow for analysis of alternative means of modeling each transport process.	Allows for ready analysis of alternative means of modeling each process, including higher temporal and spatial analyses proposed in this renewal	Precludes need to write new code each time an alternative model component is considered; provides basis for analysis at all semiarid DOE sites	Future code distribution planned following development

(Table 1: Continued)

Accomplishments	Improved Risk Assessment	Potential Cost Savings	Technology Transfer
Site-specific data for wind erosion at WIPP, Rocky Flats, and Hanford as a function of current and disturbed conditions	Provides critical data indicating thresholds related to wind velocity and disturbance; only existing wind erosion data for WIPP, only high-temporal resolution data for Rocky Flats and Hanford .	Improved risk estimates can save costs related to monitoring, remediation, and litigation	Wind erosion data being used to assess fire affects at Los AlamosPresentations at EMSP 1998 and to CEMRC / DOE in 1999; EMSP 2000 and Health Physics 2000 Symposium scheduled †
Site-specific data for water erosion at WIPP, Rocky Flats, and Hanford as a function of current and disturbance conditions	Provides critical data on thresholds related to precipitation intensity, soil type, and disturbance; only existing water erosion data for WIPP, only data for parameterizing runoff and erosion models for Rocky Flats and Hanford.	Improved risk estimates can save costs related to monitoring, remediation, and litigation	Water erosion estimates provided for Rocky Flats Actinide Migration Group modeling effort; WIPP data used by USDA-ARS for validation; Los Alamos data used to assess post-fire affects; presentations at EMSP 1998, 2000 and Health Physics 2000 Symposium scheduled †
Site-specific data for vertical migration at WIPP, Rocky Flats, and Hanford as a function of current and disturbance conditions	Provides critical data on thresholds related to soil texture, and disturbance; only existing vertical migration data in surface soils (vadose zone) for WIPP, only estimates of effects of wetting-drying cycles for Rocky Flats and Hanford	Improved risk estimates can save costs related to monitoring, remediation, and litigation	Presentations: EMSP 1998; EMSP 2000 and Health Physics 2000 Symposium scheduled †
Cross-site, multi-pathway "steady-state" assessment of contaminant transport for 7 DOE arid/semiarid sites	Provides risk estimates for the relative importance of the three pathways within each DOE site, the relative importance of a pathway among DOE sites, and sensitivity of parameters determining those priorities within and among sites, pointing to the need to incorporate threshold effects into more advanced models	Allows for prioritization of DOE funds across sites and with respect to transport processes	Presentations: EMSP 2000 and Health Physics 2000 Symposium scheduled †

† Publication submitted or in progress

4. Project Productivity

Nearly all the original project goals were completed, although modeling not proceed as far as targeted. The project was impacted by (1) a 3 month delay in receiving initial funding for all participating institutions, (2) related activities resulting from the cerro Grande fire at Los Alamos, (3) changing the third study site from Hanford to Los Alamos due to the Cerro Grande fire.

5. Personnel Supported

Los Alamos National Laboratory:

David D. Breshears
Jeffrey J. Whicker
Susan R. Johnson
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P. T. Wasiolek
R. Tavani,

Colorado State University:

Shawki A. Ibrahim
Thomas E. Hakonson
F. Ward Whicker
Mat Johansen
Randy Whicker

New Mexico State University:

Thomas B. Kirchner
Dave Schoep
Mark Walthall
David Ganaway
Jim Yahr
Carl Schloesslin
Cheryl Schloesslin

6. Publications

Numerous publications are in review or progress:

- Johansen, M., T. E. Hakonson, J. R. Simanton, F. W. Whicker. Hydrologic response and radionuclide transport following fire at semi-arid sites. Submitted to Journal of Environmental Quality.
- Whicker, J. J. Performance evaluation of several environmental aerosol inlets at high wind velocities. Submitted to Aerosol Science and Technology
- Whicker, R. D., S. A. Ibrahim, and F. W. Whicker. Vertical migration of plutonium in soils from from the Waste Isolation Pilot Plant and the Rocky Flats Environmental Technology Site. Manuscript in preparation
- Johansen, M., T. E. Hakonson, J. R. Simanton, J. J. Stone, D. D. Breshears, and F. W. Whicker. Hydrological and surficial responses to rainfall simulations following the Cerro Grande fire, Los Alamos, New Mexico. In preparation.

- Whicker, J. J., D. D. Breshears, P. T. Wasiolek, R. Tavani, D. Schoep, T. B. Kirchner, and J. C. Rodgers Wind erosion and resuspension in burned and unburned semiarid shrublands. Manuscript in preparation.

- Whicker, J. J., and D. D. Breshears. Comparison of Wind Driven Resuspension among Several Department of Energy Sites. Manuscript in preparation.

- Johnson, S. R., D. D. Breshears, and T. B. Kirchner. Comparing steady state models of wind erosion, water erosion, and vertical migration across semiarid DOE sites. Manuscript in preparation.

Abstracts from a symposium at the 2000 Health Physics meeting are published in Health Physics, Vol. 78, no.6, supplement 3, pages S156-S158 (ISSN 0017-9078) for the following:

- Breshears, D. D. Cross-cutting themes for contaminant transport in semiarid ecoystems.

- Chromec, F. W., G. A. Wetherbee, C. Dayton, J. Meyers, I. Paton, and K. Spitze. Predicting surface water impacts and cleanup levels for actinides in surface soils.

- Johansen, M. P., T. E. Hakonson, and F. W. Whicker. Actinide mobilization by surface water erosion at Department of Energy sites.

- Shinn, JH. A comparison of dustborne radiocontaminant fluxes from Tonopah, Nevada, Palomeres, Spain, and Marilinga, South Australia.

- Whicker, J. J., D. D. Breshears, P. T. Wasiolek, R. Tavani, D. Schoep, T. B. Kirchner, and J. C. Rodgers. Effects of episodic high-wind events and fire on resuspension: measurements near the Waste Isolation Pilot Plant.

- Whicker, R. D., Shawki A. Ibrahim, and F. W. Whicker. Vertical migration studies of Pu-239 in soils from WIPP and Rocky Flats.

- Janecky, D. R. Plutonium contamination: isotopics and composition as tracers.

- Johnson, S. R., D. D. Breshears, and T. B. Kirchner. Multi-pathway, multi-site contaminant transport: assessing vertical migration, wind erosion, and water erosion at semiarid DOE sites.

- Meyer, C. R. Coupling wind and water erosion models.

- Whicker, F. W. Biologically-mediated mechanisms of actinide migration.

Los Alamos National Laboratory Report:

Tavani, R. 1999. The effect of wind on the transport of potentially contaminated soil at WIPP. Los Alamos National Laboratory Report LAUR-99-4887.

7. Interactions

See published abstracts listed under Publications and interactions listed in Table 1.

8. Transitions

See technology transfer listed in Table 1.

9. Patents

None.

10. Future Work

Our proposed follow-up work to this project, submitted as an EMSP renewal proposal, was not funded. We believe additional development of a modeling framework that accounts for disturbance events is essential for DOE, particularly given that Hanford, Idaho, and Los Alamos all experienced wildfires in 2000. We will be seeking funding at Los Alamos and elsewhere to follow up on this topic.

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