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

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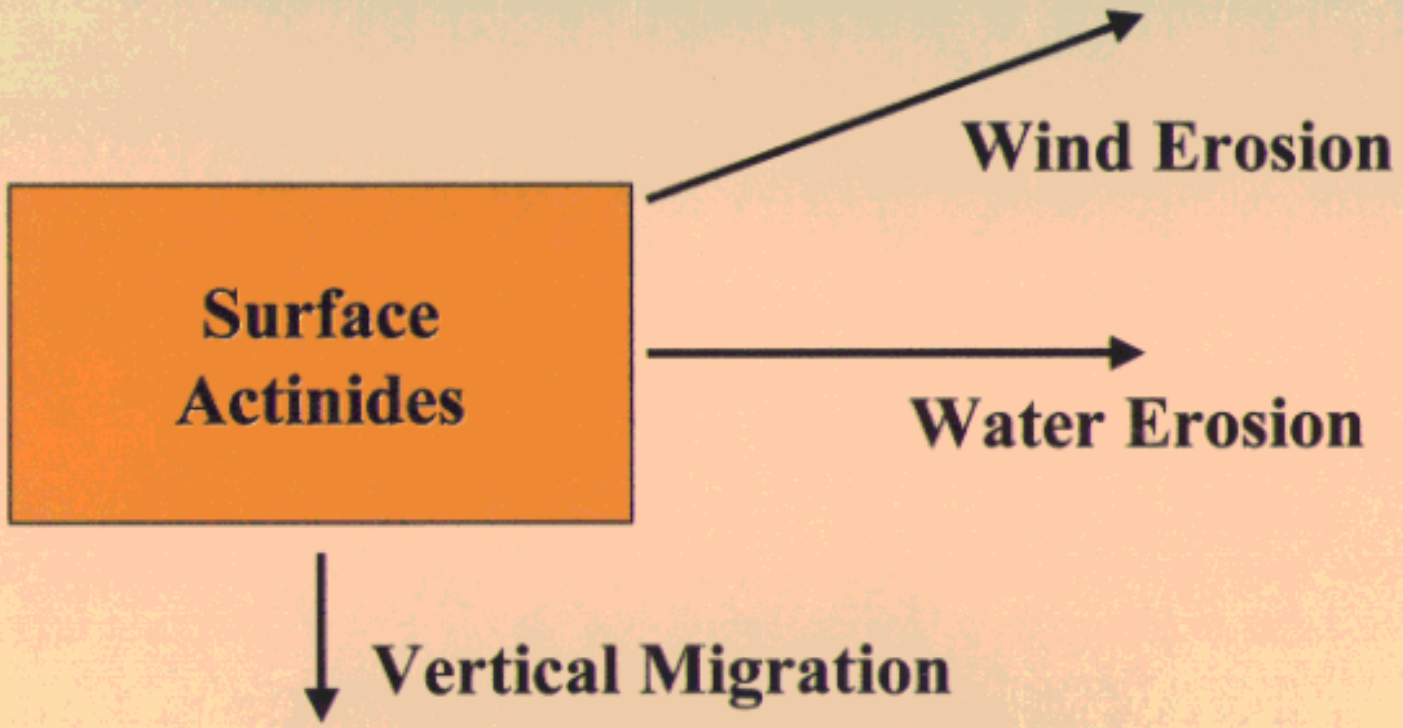
Problem Statement

- ***Actinide mobility:*** The mobility of actinides in surface soils, particularly $^{239,240}Pu$, is of concern at several DOE facilities in arid and semiarid environments. Most of these actinides are currently tightly bound in surface soils but nevertheless are potentially subject to redistribution by physical and biological processes.
- ***Litigation:*** Actinide mobility is a high visibility issue at Rocky Flats and Hanford, with current litigation and clean-up decisions pending on assessment of the risks resulting from the long-term mobility of actinides. The potential for redistribution has lead plaintiff groups to sue the DOE and its contractors at Hanford and Rocky Flats.
- ***Scientific data needed:*** Is the $^{239,240}Pu$ presently in soils immobile or is it subject to transport by physical and/or biological processes? Definitive scientific data are lacking.

Modes of Mobility for Surface Actinides



Objective

To quantify the mobility of soil actinides from all three transport modes:

- ***wind erosion***
- ***water erosion***
- ***vertical migration***

at a suite of semiarid sites where actinide mobility is a key issue of concern for DOE.

Fire:
A Surface Disturbance

On BLM land adjacent to WIPP, a control burn removes vegetation. This type of process is expected to increase surface mobility, especially for wind and water erosion.

Surface Disturbance

- Surface disturbances that cause changes in vegetation and the upper soil profile are critical processes for determining long-term mobility of actinides in the environment.
- Types of surface disturbances include fire, drought, tilling, and burrowing animals.
- Our study will include an evaluation of the effects of surface disturbances on the modes of mobility. At our first site (BLM land adjacent to the WIPP site) the disturbance was fire.

Study Sites

- ***WIPP*** (Adjacent BLM land) - 1998
  - Chihuahuan desert shrub-grassland
  - Annual precipitation: ~ 20 cm / year
  - Soils: sandy
- ***Rocky Flats*** (Tentative) - 1999
  - Shortgrass steppe - grassland
  - Annual precipitation: ~ 42 cm / year
  - Soils: variable—loamy, rocky
- ***Hanford*** (Tentative) - 2000
  - Palouse Prairie / Great Basin Sagebrush
  - Annual precipitation: ~ 15 cm / year
  - Soils: sandy
- ***Related sites:*** Other DOE facilities where our results will be applicable include ***Los Alamos***, ***Nevada***, and ***Idaho***.

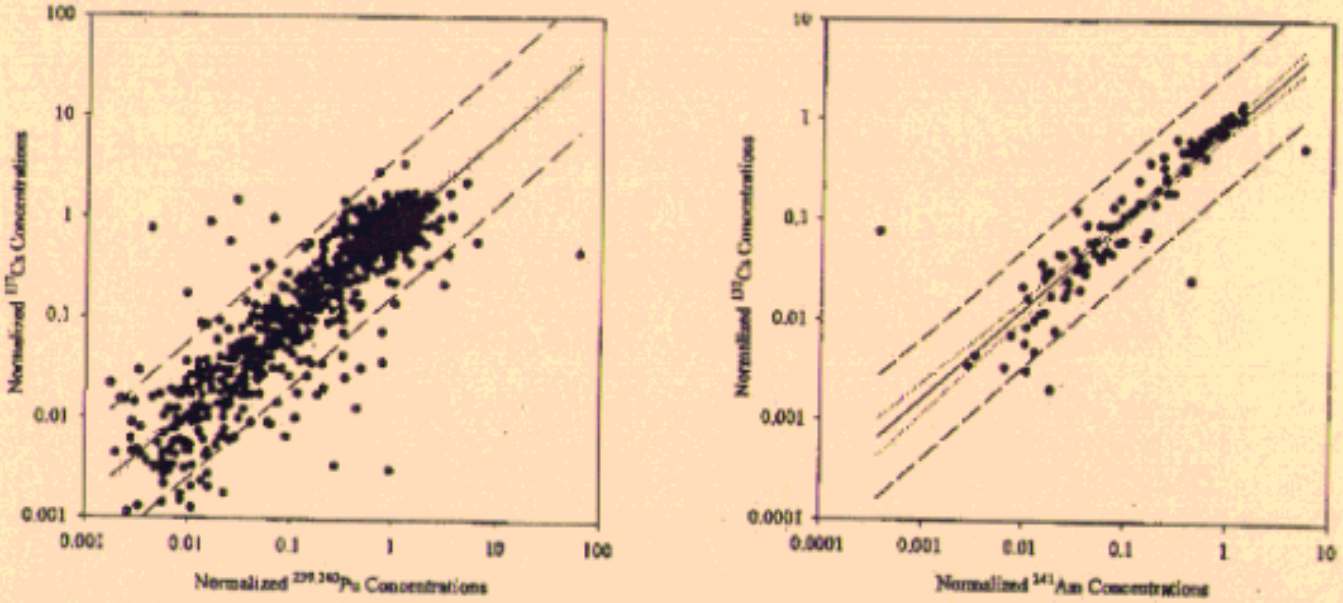
Methods

- ***Actinide tracer***
  - Literature review of ^{137}Cs and ^{241}Am as *in situ* $^{239,240}Pu$ tracers in the field
- ***Wind erosion***
  - Erosion measurements correlated with local meteorological conditions on undisturbed and disturbed plots
  - Short-term (minutes-weeks) measurements with active air samplers
  - Long-term (months) measurements with passive samplers
- ***Water erosion***
  - Rainfall simulation on undisturbed and disturbed plots
  - Estimation of several key hydrologic parameters
- ***Vertical migration***
  - Soil column studies in the laboratory
  - Treatments: wet/dry, roots, and earthworms
- ***Modeling***
  - Integration of modes of mobility building on the PATHWAY model

Actinide Tracers

- Analysis for $^{239,240}Pu$ is very expensive, whereas that for ^{137}Cs and ^{241}Am is relatively inexpensive.
- Can *in situ* ^{137}Cs and ^{241}Am be used as tracers for $^{239,240}Pu$ in the field?
- Yes—results of a literature review:
  - Significant positive correlation exist between $^{239,240}Pu$, ^{137}Cs and ^{241}Am in soils and sediments at several locations (e.g., Rocky Flats, Los Alamos, and Hanford).
  - Atmospheric transport of $^{239,240}Pu$ and ^{137}Cs from worldwide fallout is very similar.
  - Attachment of $^{239,240}Pu$ and ^{137}Cs to soil particles of various size is very similar.
  - Both $^{239,240}Pu$ and ^{137}Cs movement in the environment correlate well with soil and sediment particle movements.
  - A significant correlation between $^{239,240}Pu$, ^{137}Cs and ^{241}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.
  - Most $^{239,240}Pu$, ^{241}Am, and ^{137}Cs in soil is strongly absorbed onto clay, minerals, and organic matter and leaches very little through soil columns.

^{137}Cs and ^{241}Am as $^{239,240}Pu$ Tracers



Correlations for ^{137}Cs and $^{239,240}Pu$ and for ^{137}Cs and ^{241}Am in soil from the 3-cm depth interval on-site and the 21-cm depth interval off-site for Rocky Flats (normalized to means for samples in the 3 cm interval)

From: Hulse, S.E., S.A. Ibrahim, F.W. Whicker, and P.L. Chapman. Comparison of ^{241}Am, $^{239,240}Pu$, and ^{137}Cs concentrations in soil around Rocky Flats. Submitted to *Health Physics:* May 1998.

Chihuahuan Desert Shrub-Grassland

Study site on BLM land adjacent to WIPP

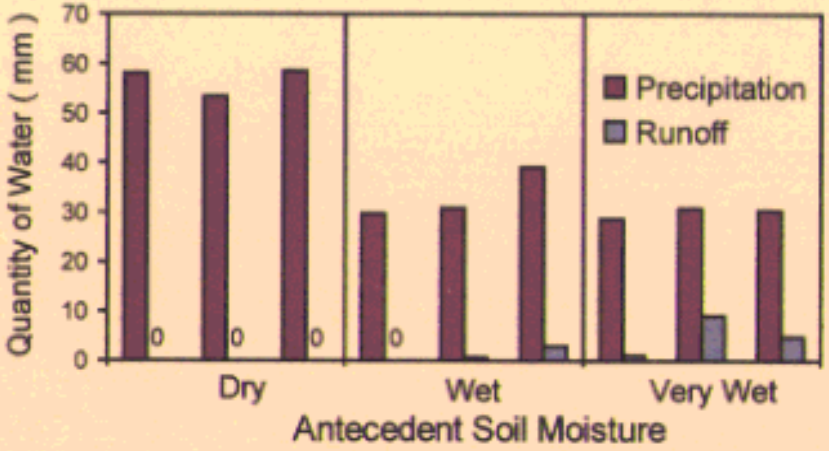
Disturbed - burned

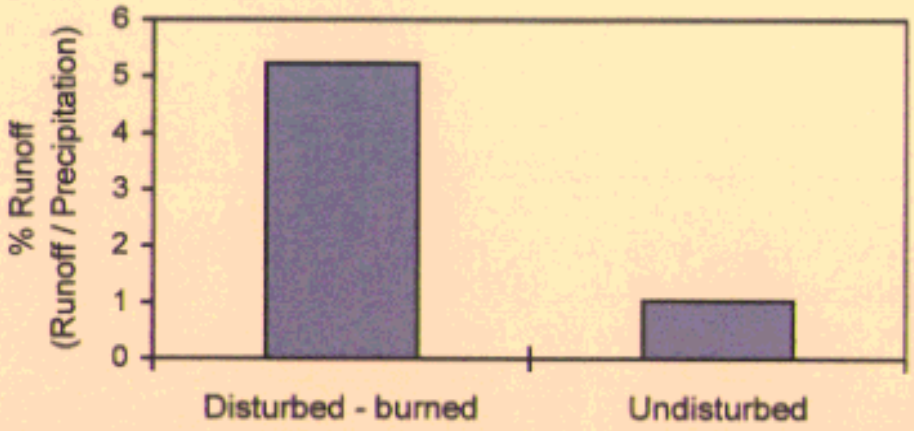
Undisturbed

Water Erosion

- Disturbed (burned) and undisturbed plots; 3 each
- Rainfall simulations for dry, wet, and very wet soils sequentially
- Runoff and erosion measured through time

Rainfall Simulation Results





- The first ~ 60 mm of precipitation, which was applied to the dry plots, yielded no runoff.
- There was very little runoff from any of the plots: < 6% of the precipitation in all cases, as compared to ≥ 10% typical for many sites.
- The disturbed plots yielded about five times as much runoff as the undisturbed plots: 5% disturbed vs. 1% undisturbed.

Wind Erosion

Seasonal integrals in wind erosion are being measured on undisturbed and disturbed plots using passive modified Bagnold samplers.

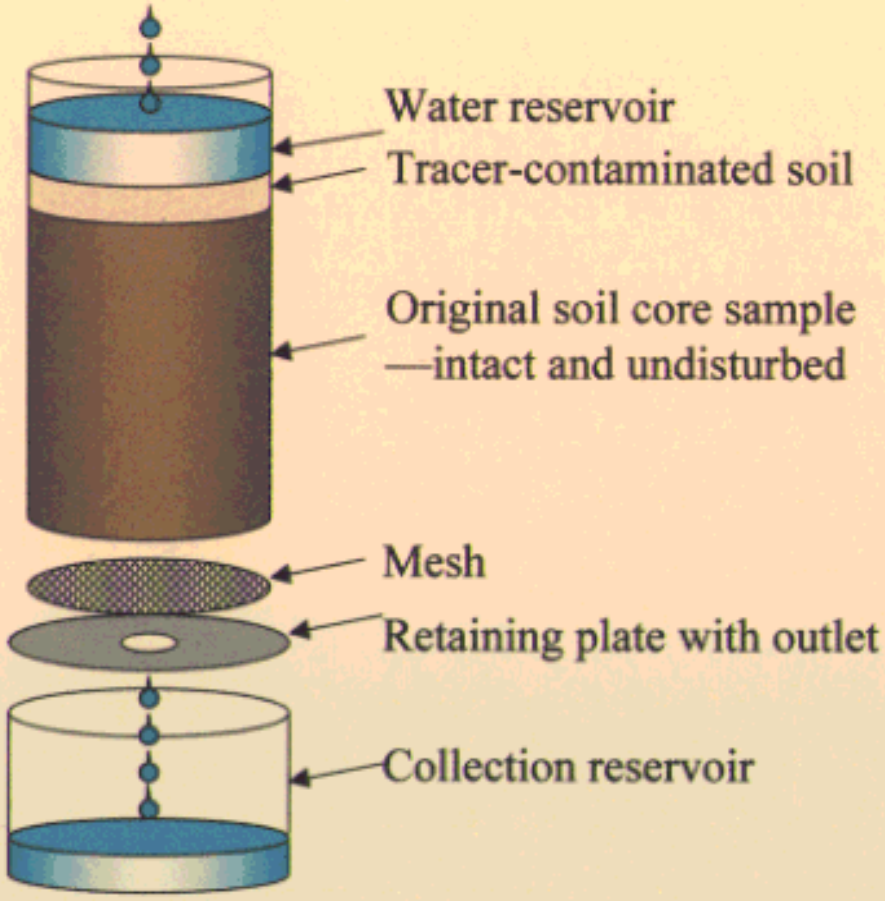
Experimental design:

- Disturbed (burned) and undisturbed plots (2 per treatment)
- Short-term fluxes of wind erosion, including particle size distributions, being measured using active air samplers (1 per treatment)
- Longer-term (monthly) fluxes of wind erosion are being measured using passive modified Bagnold samplers (3 samplers per plot)
- Local meteorological conditions being measured (1 per treatment)

Vertical Migration

Experimental design:

- Duplicate soil cores collected in tact from study sites for soil column studies
- Tracer applied to measure vertical migration
- Physical processes evaluated: wetting and drying cycles applied to promote cracking
- Biological processes evaluated: introduction of plant roots and earthworms to develop macropores



Modeling

- To include a combination of first order kinetics and discrete event processes.
- Based on the PATHWAY model (Whicker et al. 1990) and modifications initiated for Rocky Flats by Higley (1994).
- To include model development, uncertainty analysis, sensitivity analysis, and validation for a variety of sites.

Higley, Kathryn A. 1994. Vertical movement of actinide-contaminated soil particles. Ph.D. Dissertation. Colorado State University, Fort Collins, Colorado.

Whicker, F. W., T. B. Kirchner, D. D. Breshears, and M. D. Otis. 1990. Estimation of radionuclide ingestion" the "PATHWAY" food-chain model. Health Physics 59: 645-657.

Conclusions to Date

- ***Surface processes:*** Surface processes are likely to be a key factor in determining the long-term mobility of actinides in the environment; our study is one of few addressing this issue.

- ***Actinide tracers:*** Our literature review indicates that ^{137}Cs and ^{241}Am are appropriate *in situ* tracers for assessing $^{239,240}Pu$ mobility in the field.

- ***Modes of mobility at WIPP:*** The low amounts of runoff that were documented in the rainfall simulations for the WIPP site, in conjunction with low apparent clay content at that site, suggest that wind erosion may be the key mode of mobility for WIPP. Ongoing data collection will be used to assess this very preliminary conclusion.

Applications and Implications for DOE

- ***Contaminant transport and dose assessment:*** Contaminant transport and dose assessment at DOE sites is highly dependent on the modes of mobility that we are quantifying—the relative importance of the three modes of mobility directly effects risks.
- ***Litigation issues:*** Ongoing litigation at DOE sites fundamentally relates to the three modes of mobility that we are studying—litigants want to know about off-site transport of surface actinides by wind and water erosion and about risk from ground water contamination due to vertical migration.
- ***Comparison of remediation alternatives:*** Remedial alternatives for many contaminated sites are fundamentally dependent on the relative importance of the three modes of mobility—the results can promote cost savings through effective remediation based on understanding key processes.

Participants

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  - Thomas B. Kirchner; Analysis
- ***Collaborators***
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  - Steve Daly , BLM

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Related reprints: Reprints from recent related studies by the investigators are available below. For additional information contact PI David D. Breshears at daveb@lanl.gov, (505) 665-2803, or at the address above.