

**Department of Energy**

ROCKY FLATS FIELD OFFICE
P.O. BOX 628
GOLDEN, COLORADO 80402-0628

JUN - 6 1997

97-DOE-02499

Dear Community Member:

As a follow up to the last Actinide Migration Meeting on March 31, 1997, I am sending you the following three documents regarding the Actinide Migration Study:

- Proposed Path Forward for the Actinide Migration Studies, June 1997
- Development of Conceptual Model of Potential Pathways for Migration of the Actinides Uranium, Plutonium, and Americium at the Rocky Flats Technology Site, June 1997
- Work Scope Document for 'Actinide Migration Studies at the Rocky Flats Environmental Technology Site', May 26, 1997

The Proposed Path Forward lists all planned activities over the next two to three years. This will help us gain a better understanding of actinide migration and build a defensible conceptual model which will help guide future remedial activities. The Development of Conceptual Model document explains what a conceptual model is and what types of information are needed to better understand actinide migration at the Rocky Flats Environmental Technology Site. Finally, the Work Scope document details Bruce Honeyman and Peter Santschi's work scope for fiscal year 1997. This work is a component of the overall Path Forward for Actinide Migration Studies.

I welcome your comments. Comments that can be reasonably incorporated into this year's scope of work will be. On the other hand, if specific comments are beyond this year's scope of work they will be considered for next year's activities. Please feel free to call me with any questions at 966-6246.

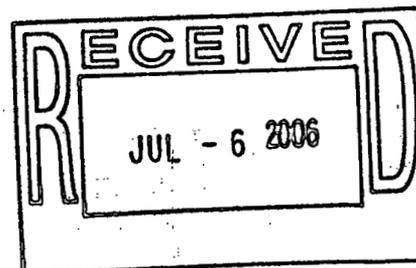
Sincerely,



John J. Rampe
Environmental Restoration
Program Coordinator
Rocky Flats Field Office

Enclosures

cc w/o Enclosures
J. Legare, AMEC, RFFO
J. Karpatkin, OCED, RFFO
N. Castaneda, CD, RFFO
C. Dayton, K-H



ADMIN RECORD

SW-A-005705

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PROPOSED PATH FORWARD FOR THE ACTINIDE MIGRATION STUDIES
JUNE 1997

1.0 BACKGROUND AND REGULATORY FRAMEWORK

The evaluation of remedial alternatives for actinides (plutonium [Pu], americium [Am], and uranium [U] isotopes) at Rocky Flats Environmental Technology Site (RFETS) (the "Site") must consider migration and mobility (transport) along environmental pathways. Transport pathways include: soil (surficial and subsurface), groundwater, surface water, and air. A conceptual model of actinide transport at RFETS is being developed to better understand the relationship among these transport pathways and the potential effects of actinides on surface water and air quality. In 1996, the Actinide Migration Expert Panel was formed to make an initial assessment of the Conceptual Model. As recommended by the Panel, this Actinide Migration Study has been initiated to further the understanding of the Conceptual Model.

1.1 1996 Work and Results

During 1996, the Actinide Migration Expert Panel, which consisted of nationally recognized experts on actinide chemical behavior and migration/mobility in the environment, reviewed existing data on actinide migration at RFETS and made recommendations for further work. Their recommendations included: 1) develop a defensible conceptual model for actinide transport, based on a thorough understanding of chemical and physical processes; 2) investigate the long-term impacts of actinide geochemical mobility on remedial requirements; and (3) evaluate the long-term protectiveness of the Rocky Flats Cleanup Agreement (RFCA) soil action levels on surface water quality.

The RFCA action levels and standards were also developed during 1996 by an interagency work-group. Radionuclide soil action levels were calculated based upon dose limits for selected land use scenarios, and surface water standards were assigned at points of compliance. The Conceptual Model will also assist in assessing the long-term protectiveness of the actinide soil action levels on surface water, as required in the RFCA.

1.2 Regulatory Framework

Attachment 5 of the RFCA (the Action Level and Standards Framework for Surface Water, Groundwater, and Soils) sets forth the standards and action levels for environmental media which incorporate the RFCA Vision, and land- and water-use controls in the RFETS cleanup. Actinide surface water standards are based upon human health risk as a consequence of human ingestion of surface water and are assigned at compliance points as water leaves the site, so that it is of acceptable quality for any use. The soil action levels for radionuclides are based upon radiation dose limits for designated land-use scenarios for RFETS and did not consider the transport of soil containing actinides to surface water because no consumption of groundwater or surface water was assumed. It is anticipated that the dose-based soil action levels for remedial activities will clean up the most significant contamination, but it is recognized that the long-term protectiveness of surface water quality requires confirmation by the Actinide Migration Studies.

RFCA states that protection of surface water uses with respect to the long-term Site condition will be the basis for making soil and groundwater remediation and management decisions, and that additional groundwater or soil remediation or management may be required for the protection of surface water quality or ecological resources. Development of a thorough conceptual model will allow RFETS to better understand the relationship of actinide levels in soil and the effect of remedial activities on the long-term protection of surface water. As indicated in RFCA, additional remediation and/or institutional/management controls may be warranted for the long-term protection of surface water quality.

2.0 LONG-TERM GOALS OF THE ACTINIDE MIGRATION STUDIES

Members of the 1996 Actinide Migration Expert Panel are assisting the RFETS in the multi-year Actinide Migration Studies. The Studies' mission is to develop a better understanding of the chemical and physical mechanisms of actinide mobility and potential impacts to surface water, in order to build a defensible conceptual model that may guide remedial activities for Site closure. The associated goals are:

- Assess the long-term protectiveness of the actinide soil action levels on surface water;
- Design remedial actions that minimize the environmental migration of actinides after Site closure and that will meet RFCA goals for surface water quality; and
- Understand the main actinide environmental transport mechanisms in order to better understand the Conceptual Model (see Attachment 1 for a detailed discussion of the Conceptual Model).

These goals will be supported by:

- Inventorying total quantities of actinides in environmental media at the Site;
- Preparing mobility distribution maps for soils, sediments, and groundwater;
- Estimating the quantities and distributions of actinides in soil and sediments at the time of Site closure; and
- Assessing the quantity of sediments that may reach each drainage basin (mass loading limits) that will be protective of surface water quality.

3.0 FISCAL YEAR 1997 WORK-SCOPE OF THE ACTINIDE MIGRATION STUDIES

The work for the 1997 fiscal year in support of the long-term goals consists of:

- Finalizing the *Evaluation of Existing Data on Actinide Migration at the Rocky Flats Environmental Technology Site Report* (Draft, September 1996) (referred to herein as

Evaluation of Existing Data Report) in order to present currently available data and to identify potential data deficiencies in the understanding of the Conceptual Model;

- Collecting initial sediment, soil, and water data from discrete locations containing actinides for phase association and soil/water partitioning of Pu;
- Preliminary assessment of the loading of actinides to surface water from soil erosion and preferential flow in drainages leading from the industrial area, and assessment of the long-term impacts to surface water quality; and
- Provide recommendations on what additional data is needed to meet the long-term goals.

The Evaluation of Existing Data Report is planned to be available in August 1997, and the remaining work (which is described in Attachment 2) is planned to be summarized in a report available in October 1997.

4.0 PRIORITIZATION OF INFORMATION COLLECTION AND EVALUATION

Additional information needed to develop a thorough conceptual model has been identified and prioritized. Actions needed to obtain this information have been prioritized to facilitate the remediation of the 903 Pad, and the Solar Ponds plume and other remedial actions. The following table lists the needed information, the actions planned to obtain the information, and the fiscal year (FY) in which they will be completed.

INFORMATION NEEDED	FISCAL YEAR	ACTION
Evaluate existing knowledge on the movement of actinides from vegetated and unvegetated surface soils to drainages.	1997	Review previous erosion studies for the 903 Pad, Lip Area, and Walnut Creek. Summarize review in the revised <i>Evaluation of Existing Data Report</i> .
Determine Site-specific chemical behavior of actinides (speciation and distribution coefficient [Kd] values, see Attachment 1 for description) using contaminated soils and groundwater from RFETS for the purpose of assessing the long-term protection of surface water.	1997	Initial laboratory studies of speciation and determination of Kd values for soil and pond sediment samples.
	1998	Additional laboratory studies, if needed.

Path Forward for Actinide Migration, June 1997 (Cont'd)

INFORMATION NEEDED	FISCAL YEAR	ACTION
Estimate erosion rates under current conditions and for proposed remediation options using the most current methodologies.	1997-1998	Use most current version of the Revised Universal Soil Loss Equation (RUSLE) model to estimate erosion rates. Work will begin in FY97 and is expected to continue into FY98.
Evaluate the groundwater U plume at the Solar Ponds and determine the predicted impact of the plume to surface water quality;	1997	Review existing data and summarize in the revised <i>Evaluation of Existing Data</i> .
	1998	Incorporate new chemical behavior data (speciation and Kd values) in modeling to predict impacts of the Solar Ponds U plume to surface water.
Evaluate the design of performance monitoring systems for surface water;	1997-1998	Review current surface water monitoring system, make recommendations for future environmental monitoring.
Estimate the potential remobilization of actinides in ponds to establish the significance of the pond sediments.	1997	Review data on the Walnut Creek Drainage Area and augment as needed.
	1997-1998	Collect sediment and water samples in the ponds; profile Pu and Am levels with depth; inventory quantities in ponds; determine the particle size; mineralogy; and chemical and physical behavior of Pu and Am in sediments.
Evaluate wind transport mechanisms at the time of Site closure and its potential contribution to the movement of actinides.	1997	Review and expand upon information available on wind erosion at the Site.

Path Forward for Actinide Migration, June 1997 (Cont'd)

INFORMATION NEEDED	FISCAL YEAR	ACTION
<p>Develop loading estimates for each drainage basin to meet surface water standards. Estimate remediation goals to meet estimated loading limits and compare to surface soil action levels.</p>	<p>1997 - 1998</p> <p>1998</p> <p>1998</p>	<p>Analyze existing air particulate collection filter from the 903 Pad area for Pu and Am.</p> <p>Model wind transport of actinides using new and existing data</p> <p>Use results of erosion rate modeling on soils adjacent to the Walnut and Woman Creek drainages to develop estimates of loading limits.</p>
<p>Summarize the refined Conceptual Model</p>	<p>1998-1999</p>	<p>Summarize the refined Conceptual Model in a report.</p>
<p>Finalize 903 Pad Interim Measure/Interim Remedial Action (IM/IRA) using information from the above actions.</p>	<p>1998-1999</p>	<p>Finalize the 903 Pad IM/IRA.</p>

**DEVELOPMENT OF CONCEPTUAL MODEL OF
POTENTIAL PATHWAYS FOR MIGRATION OF THE ACTINIDES
URANIUM, PLUTONIUM, AND AMERICIUM
AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE**

June 1997

1.0 INTRODUCTION

Conceptual models are developed to understand how a site "works" both chemically and physically. Conceptual models in environmental work are used to tie together the relationships between surface and subsurface geology, soil types, contaminant types, and distribution with air, groundwater, and surface water transport of contaminants. Development of a conceptual model based on existing data and understanding of the problem forces the identification of data deficiencies, where the problem is not known or not understood completely. Completion of the conceptual model will include filling in the data deficiencies and integrating the new data.

A conceptual model of actinide distribution and transport at the Rocky Flats Environmental Technology Site (RFETS) is being developed to better understand the relationship between soil types, actinide distribution, hydrogeology and the potential effects of the actinides on surface water quality and air quality. The conceptual model will be used to assist in evaluating and designing remedial actions.

The actinides of concern at RFETS are uranium, plutonium, and americium. For the purposes of this discussion, the term "actinides" refers to these three elements. Uranium contamination at RFETS was the result of different processes and is found in different areas than the plutonium and americium contamination at RFETS. Both uranium and plutonium were used in the manufacturing processes at RFETS. Americium at RFETS is the result of the decay of plutonium-241 to americium-241. The distribution and environmental behavior of plutonium and americium in RFETS soils are similar. Uranium, in contrast, has environmental characteristics and distribution that are distinct from plutonium and americium.

Transport of actinides at RFETS includes both chemical and physical mechanisms. Physical transport is the movement of the media that contains the actinides (i.e. soil or water). Once the transport mechanisms are understood, they can be quantified, and remedial design decisions can be made based on this understanding. Chemical transport is the transfer of actinides from one physical form to another, for example, the chemical transfer of actinides from being sorbed onto soil particles to being carried by water in solution as a dissolved form.

Potential transport pathways, data, and data evaluation needs for better understanding actinide migration from RFETS have been identified. These pathways are shown in Figure 1 and are generally broken down into discussion by environmental media:

- Soil erosion- The transport of surface soil particles by overland flow to surface water in the Walnut and Woman Creek Drainages will be evaluated. A data review and evaluation will provide the information needed to estimate how much material must be removed from sources, such as the 903 Pad and Industrial Area, to achieve residual

soil actinide concentrations that are protective of surface water quality. To determine this, the loading of sediments (and the related actinide) to each surface water drainage will be determined. Actinide migration to drainages will be estimated by conducting an inventory to determine the source areas and the quantities/nature of actinide contributions of soil erosional transport in each drainage. Rates of soil erosion will be determined so that post-cleanup levels of actinides will be protective of surface water quality.

- Surface water - An evaluation will determine if potential remobilization of actinides in the ponds and sediments by both physical erosion (mobilization of pond sediments to surface water) and chemical partitioning (from actinide-contaminated sediments to surface water in a dissolved phase) will impact surface water quality after final disposition of the ponds.
- Air pathway - The air monitoring performed at RFETS collects the fraction of actinides that is transportable by wind. The long-term transport of actinides by wind erosion will be assessed. Existing air monitoring data, association of plutonium and americium with surface soil grain size, and climate data will be evaluated and incorporated into the conceptual model. After evaluating existing data, a determination will be made regarding the need for additional data to better quantify air transport of actinides.
- Groundwater - Chemical and physical mechanisms contributing to actinide migration in groundwater to surface water under the environmental conditions encountered at RFETS will be assessed. The evaluation will also address groundwater transport and the soil action levels in relation to long-term protectiveness of surface water.

For the basic transport components of the conceptual model, actinides are present in the following, major environmental media (Figure 1):

- surface soils (0 to 15 cm);
- subsurface soils (15 cm to the top of the groundwater);
- the groundwater (saturated zone);
- surface waters (streams, ponds, , etc.) and
- surface water sediments.

Actinides in these media can be further broken down into two phases: a solid (particulate) phase and a dissolved phase. The particulate phase consists of the solid parts of a soil, sediments, and solids suspended in surface water and groundwater. Figure 1 illustrates the environmental media that contain actinides at RFETS, and the contaminant transport pathways

The rates of transport from the soils are represented by "R" in the figure. For example, the R_a , rate of transfer of actinides from soil to air will be evaluated. The subscripts designate the transfer process. Once the ranges of transport rates are known for the various mechanisms, the relative proportions of total actinide transport by each mechanism can be determined. The conceptual model will be refined to incorporate this information.

The unsaturated soils are divided into surface and subsurface soils because of the differences in actinide distribution and transport mechanisms that apply. Approximately 90% of the

plutonium and americium in soils at RFETS is within the top 12 cm (Krey, et al., 1976, Litaor et al., 1994). Overland transport is generally more important than chemical transport for actinide migration in surface soils (Marty et al, 1997). Transport mechanisms in the surface soil are physical erosion of actinides on soil particles by water and wind, the physical mixing of soils by burrowing animals, and downward transport in infiltrating water. These are shown as Ra, Re, and Ri in Figure 1.

The relationship between soil actinide concentrations, storm events and surface water quality needs to be better understood. If plutonium is strongly associated with soil particles Re will be the dominant transfer rate (i.e., plutonium and americium will enter the surface waters primarily through water-induced erosion of soil particles) and will settle out as pond and stream sediments. If the actinides are easily 'solubilized' then they will most likely enter the surface water through the dissolved phase.

The remaining sections of this report present the components of the conceptual model. Section 2.0 discusses erosional transport of actinides in surface soils. Section 3.0 discusses subsurface actinide transport in infiltrating water and in groundwater, and Section 4.0 discusses transport in surface water and sediments.

2.0 EROSIONAL TRANSPORT BY WIND AND WATER AT RFETS

This section discusses the data which must be evaluated to complete the erosional portion of the conceptual model of actinide transport.

The amount of actinides moved by soil erosion processes at RFETS depends on:

- spatial distribution of actinides;
- distribution of actinides in relationship to slopes and uplands;
- distribution of actinides with soil types;
- distribution of actinides with grain size;
- distribution of different sized particles within the soil (i.e., are the bigger rocks all in layers, or are they evenly distributed throughout the soil); and
- distribution and type of plant cover.

Information on the factors listed above are available from several sources at RFETS, including investigation and treatability study reports from the former Operable Units, ecology reports, and air monitoring reports. The existing data will be reviewed and utilized as discussed in the following sections.

2.1 Wind Erosional Transport of Actinides

In addition to the distribution of actinides in relationship to the various factors noted above, the transport of actinides by wind erosion is related to the climatic and topologic factors that control wind erosion of any soil. Climate factors include wind speed, direction, duration, timing of wind with seasons, and precipitation, both total amount, and when it occurs. Climate data have been gathered for a number of uses at RFETS and in the surrounding areas. These data will be incorporated into the conceptual model so that quantitative calculations of soil erosion can be performed for various remediation scenarios.

Paved areas and amount/type of vegetative cover over non-paved areas also affect wind erosion of soils. The types of vegetation at RFETS have been mapped, and will be combined with the spatial distribution of actinides and soil type. Current air monitoring at RFETS is designed to determine the amount of radionuclide emissions from paved, roofed, and unpaved areas. Monitoring is also designed to determine the radionuclide emissions from the 903 Pad and Lip area, the area with the most extensive surface soil actinide contamination at RFETS. This data indicates that the majority of actinide transport is from unpaved areas, specifically the 903 Pad and Lip Area. Data from the current and past air monitoring programs will be incorporated into the conceptual model to understand the main actinide air transport mechanisms.

Because the air monitoring system collects data on the actinides that are mobile in air, the actinide studies will include determining the chemical form (speciation) and particle size of the actinides collected on air monitors. This data and conceptual model of wind transport under different conditions will be used to assist in the design of remedial actions to lessen the effects of wind transport during remediation, and to lessen long-term wind transport following remediation.

2.2 Water Erosional Transport of Actinides in Soils

Erosion of soil by precipitation and related actinide transport is controlled by the distribution associations of the actinides noted above, and is also influenced by climatic and storm-specific factors. Surface soils are transported by water as overland or sheet flow. The amount of soils suspended in this flow is related to the intensity of the storm, the size and velocity of raindrops, and the condition of the soil before the storm, vegetative cover, and slope. Erosion of soil can be calculated approximately by use of the Revised Universal Soil Loss Equation (Renard and Foster, 1997). As part of the Conceptual Model of Actinide Transport, this equation will be applied to the soils at RFETS to determine the amount of actinide loading to each surface water drainage basin. Previous work on the analysis of soil transport at RFETS using the Universal Soil Loss Equation will be reviewed and updated, or incorporated into a larger-scale evaluation if appropriate.

The protectiveness of the Rocky Flats Cleanup Agreement soil action levels when transport to surface water is considered will be evaluated based on input to the various transport mechanisms developed for the conceptual model. Development of a thorough conceptual model will allow RFETS to determine the relationship between soil actinide activity levels and the effect of these actinide activities on surface water for various remediation scenarios.

3.0 SUBSURFACE SOIL AND GROUNDWATER ACTINIDE TRANSPORT AT RFETS

Much of the following discussion on groundwater and subsurface soil transport of actinides was derived from Honeyman and Santschi, 1997, Correspondence, *A conceptual model of Pu movement through RFETS soils*. Transport of actinides downward into subsurface soils occurs by several mechanisms, including dissolved transport with infiltrating water, as particles (including colloids) carried with infiltrating water, and by soil disturbance by burrowing animals and plant roots. These disturbances are known as bioturbation. Within subsurface soils, actinides may be transported downward towards the water table or laterally within the ground water in both solid and dissolved phases. Typically, a distinction is made between particle and

water (i.e., 'dissolved') forms of actinides through an operational definition: that portion of a soil/water system which is smaller than 0.45 mm is designated as 'dissolved'. This is based on the 0.45 um filter size most commonly used for collecting groundwater samples to be analyzed for dissolved contaminants.

Mathematical models for simulating the transport of chemical constituents through soils by water typically treat the transport as occurring primarily in the dissolved form. The distribution coefficient (K_d) values reflect the distribution of a chemical between a solid phase (e.g., soil or sediments) and the water phase in contact with the solid (e.g., groundwater or surface water). Most computer models used for simulating the transport of contaminants (including actinides) in the environment use K_d values as part of the "mobility" calculations in the model.

Typically, a dissolved element will interact to some extent with the solid (and usually immobile) phase. For a radioactive element, the K_d is simply the ratio of the activity¹ concentration of an element in the solid phase in pCi/kg (picocuries /kilogram) to the corresponding activity concentration in the 'dissolved' phase in pCi/L (picocuries /liter). As a result, the dimensions of K_d are L/kg (liters/kilogram). An element or compound with a low K_d will be easily dissolved and transported in water. Table salt for example, NaCl, has a low K_d . An element with a high K_d value will partition to the solid phase. Because transport through subsurface soils primarily occurs as the result of water movement (or transport in the dissolved phase), a high K_d element will not be very mobile in groundwater or surface water. An accurate knowledge of the particle/water partitioning of an element is important because the extent of partitioning regulates the mobility of an element by transport through the aqueous phase. The K_d relates to the transport velocity of an element to the velocity of the water. For example, a compound with a K_d of 0 L/kg will move at the same velocity as the water phase. A compound with a K_d of 1000 L/kg will move approximately at 1/1000th the velocity of the water.

One assumption often made when using K_d to calculate mobility is that a chemical equilibrium exists in the partitioning of an element between the solid and dissolved phases. Depending on the form of the dissolved plutonium this assumption may not be valid. For example, partitioning (i.e., the movement of plutonium from the dissolved phase to the solid phase or the reverse) may be very slow. Or, the 'dissolved' Pu may not be truly dissolved, but actually be in the form of micro-particles (i.e., colloids). For example, a $\text{PuO}_2(\text{s})$ particle (the '(s)' means 'solid') 0.1 mm in diameter, while operationally defined as 'dissolved' (i.e., it is smaller than the 0.45 mm limit of the common groundwater sampling filter), will behave quite differently than a truly dissolved form of Pu (e.g., PuO_2^+). In the case of $\text{PuO}_2(\text{s})$ colloids, the use of K_d values as part of a model for the simulation of Pu transport is inappropriate.

An illustration of the importance of the chemical form of an element (and the related K_d) is the response hypothesized by Liator (Liator, et. al., 1996 correspondence to RMRS) of plutonium to the May 1995 storms. Previous evidence suggests that, under normal weather conditions, dissolved plutonium transport through soils is relatively limited and plutonium transport is confined to macropore transport of low-solubility plutonium species (Liator, 1996 et. al., correspondence to RMRS). However, Liator hypothesized that prolonged soil flooding during May 1995 produced reducing (no oxygen) conditions which markedly increased the solubility of plutonium (i.e., K_d values possibly decreased by several orders of magnitude). The current

¹Activity is a measure of the number of nuclear transformations per time. A pCi (picocurie) is 10^{-12} Curies. One Curie corresponds to 2.2×10^{12} disintegrations per minute.

actinide studies will assess the effects of reducing conditions on plutonium mobility using RFETS soils and groundwater.

Groundwater transport of actinides at RFETS is expected to be especially important in evaluating the need for remediation of uranium-contaminated groundwater and the types of remedial actions to perform at the Solar Pond Area.

4.0 SURFACE WATER AND SEDIMENT ACTINIDE TRANSPORT

Actinides can enter surface waters by several different paths: 1) erosion of surface soils; and 2) from groundwater in the case of 'gaining' streams, and 3) re-suspension of sediments. The rate (R) of transfer of actinides from soil to the surface water depends on the magnitude of water flow and the amount of actinides in each media that is readily available to be transported by the moving water. Thus, the rate depends on the water velocity, and the distribution of the actinides between the solid and dissolved phases.

Monitoring of the ponds at RFETS has shown that actinides enter the ponds in higher concentrations than when they leave the ponds (RMRS, 1996). The actinides are adsorbed onto the pond sediments and settle out to the bottom of the ponds with the sediments. The ponds act as a repository for the actinides that are transported by water erosion of soil. The relationship between soil actinide activities, storm events, and surface water quality is not quantified.

When actinides are strongly associated with soil particles, (i.e., the K_d is large), erosion is the main transfer form and sediments will be the main receptors. Actinides will enter the surface waters and sediments primarily through water-induced erosion of soil particles. If the K_d for plutonium and americium is low, then these actinides will most likely enter surface water in the dissolved phase. Erosion is typically only important during high intensity storms. Lateral groundwater flow to the surface water may occur any time the surface water drainages are gaining streams. Groundwater flows into gaining streams and away from losing streams. The surface water drainages at RFETS are both gaining and losing. The drainages are generally gaining in their upper reaches, and losing streams in their lower reaches. This also varies seasonally, with the streams generally losing water in the drier seasons, and gaining water in the wetter seasons.

Actinides associated with solid phases can be transported to and in surface waters by re-suspension of actinide-contaminated sediments. The amount of sediments that can be carried in suspension depends on the stream velocity and the grain size of the sediments. The association of actinides with grain size in both surface soils (that can erode into streams) and in stream and pond sediments (that can be re-suspended) is important to the quantities of actinides that can be transported. When RFETS closes, the existing ponds will be transformed into a flow-through system. The actinide conceptual model will contribute to design of a flow-through system that maintains the quality and usability of the surface waters at RFETS even after site closure.

As part of the actinide studies, K_d values for actinides in RFETS soils and surface water sediments will be determined. These will be K_d values for the experimental conditions. Changes in chemistry of the water used in the experiments will cause changes in the K_d value.

For example, a K_d determined using distilled water on RFETS soils would be somewhat different from a K_d value determined using RFETS groundwater on RFETS soils. This is because the composition of the water affects the K_d . For this reason, the K_d determinations will be done using RFETS soils, groundwater, sediments, and surface water.

5.0 REFERENCES

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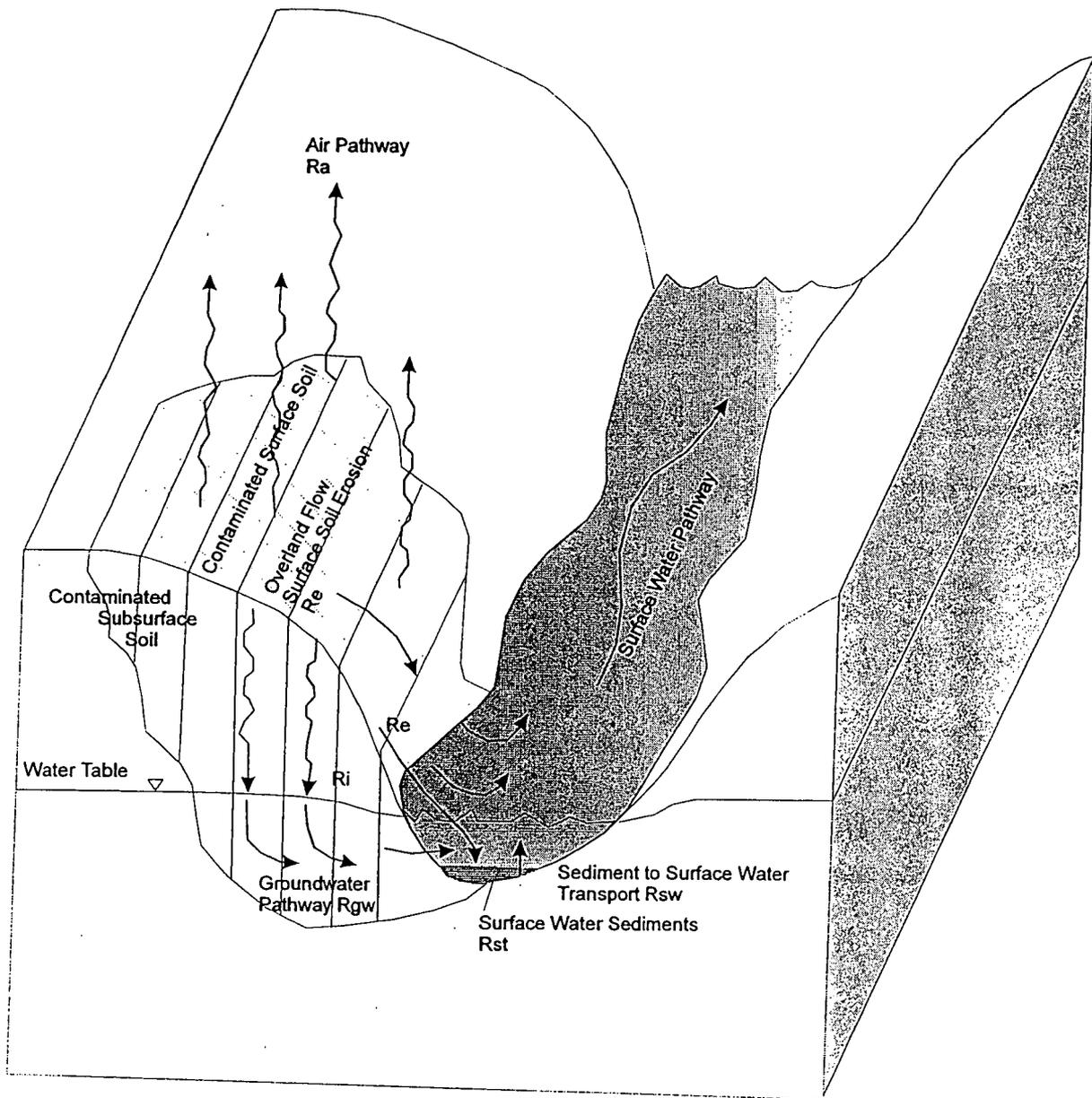


Figure 1 Preliminary conceptual model of potential actinide transport pathways

R = rate of actinide transport, subscripts indicate transport mechanism

Ra = air

Re = soil erosion

Rgw = groundwater

Ri = infiltration

Rsw = sediments to surface water

Rst = sediment transport

**Work Scope Document for 'Actinide Migration Studies at the
Rocky Flats Environmental Technology Site'**

Bruce D. Honeyman, Colorado School of Mines, and Peter H. Santschi,
Texas A&M University at Galveston

May 26, 1997

Document #: CSM-2-97

1. Objectives.

The scope of work for this fiscal year (May 1, 1997 to September 30, 1997) has two overall objectives: 1) provide a preliminary determination of the range in Pu phase speciation and soil distribution coefficient (K_d) values in 903 Pad area soils; 2) determine plutonium (Pu) inventories for ponds C-2, B-1 and B-5. This data, coupled with information about water flow during 'normal' rains and storm events, will provide a basis for further evaluation of the rate of radionuclide transfer to surface waters and the link between surface water quality and soil action levels (see Honeyman and Santschi, *A conceptual model of Pu movement through RFETS soils*). The overall objective is to provide information as a foundation for assessing whether additional soil cleanup goals must be established, or further institutional controls may be needed, for the protection of surface waters.

Sampling locations are shown on Figure 1.

The current contract provides the resources for approximately 80 analyses of Pu and 10 for uranium (U). These analyses will be distributed among the K_d and phase speciation determinations.

2. Tasks.

2.1. Task 1: Develop K_d s and speciation for Pu interaction with soils and sediments.

2.1.1. Objective:

The objective is to provide a baseline data set on Pu phase speciation and mobility in the Woman Creek drainage with a focus on the 903 Pad 'lip' area. This objective will be met through an evaluation of two types of analyses: 1) phase speciation; 2) K_d values. See correspondence to Christine Dayton, *A Conceptual model of Pu Movement through RFETS Soils*, May 26, 1997.

2.1.2. Justification:

Three Woman Creek drainage areas will be sampled: 1) 903 Pad 'lip' area soils; 2) South Interceptor Ditch (SID) sediments; 3) Pond C-2 sediments. 903 Pad 'lip' area soils are the primary source-term for Pu in the woman Creek basin. Sediments in the SID and Pond C-2 contain the 'mobile' fraction of Pu. The strategy is to compare Pu phase speciation in the three areas in support of model development for Pu transport away from the 903 Pad source areas.

2.1.3. Analytical Plan:

Two types of soil isolates (i.e., soil samples) will be removed from each of the six 'lip area' locations: 1) a 'box core', approximately 10 cm x 10 cm x 15 cm (depth): i.e., a surface soil sample; 2) a 1" x 1 m core. The box core will be the source of material for phase speciation analyses; 1" x 1 m core will be used for determinations of K_d with depth in the soil column.

Phase speciation: Litaor and Ibrahim (1996) describe preliminary evaluation of Pu soil phase speciation in the surface soils upgradient of seep SW 53 and conclude that most of the Pu is associated with organic carbon and sesquioxides (e.g., soil oxides composed containing iron and aluminum). The objective of this work is to evaluate Pu phase speciation in a series of locations in the 903 Pad 'lip' area (i.e., the sources areas) and, by comparing Pu phase speciation in SID sediments and Pond C-2 to that of the lip area, to deduce the mobile Pu fraction. Phase speciation is used to describe a series of selective chemical attacks on environmental media such as soils and sediments to semi-quantitatively evaluate the association of a target element with soil phases (e.g., Yong *et al.*, 1993, *Can. Geotech. J.*, 30, 834-847). Typically, the 'speciation' of the target element can be categorized as exchangeable, bound to carbonates, bound to iron (Fe) and manganese (Mn) oxides, bound to organic matter and a residual fraction typically considered to correspond to the mineral or rock matrix.

Distribution coefficients (K_d): Distribution coefficients, K_d s, are common model parameters for environmental mobility and soil action level calculations for target contaminant (see Honeyman and Santschi, *A conceptual model of Pu movement through RFETS soils*, Document #: CSM-3-97). A distribution coefficient reflects the net of all chemical and physical processes that result in the distribution of an element between the particle and solution phases. The variability in K_d values for a particular element reflects the range in environmental conditions at the site where the K_d values will be applied. As such, K_d values are 'conditional' on the characteristics of the system: K_d values simply reflect the distribution of a radionuclide between a solid phase and a water phase that is in contact with the solid. The evaluation of K_d values will follow standard analytical protocols (e.g., Honeyman, B.D. (1984). *Cation and Anion Adsorption at the Oxide/Solution Interface in Systems Containing Binary Mixtures of Adsorbents: An Investigation of the Concept of Adsorptive Additivity*. Department of Civil Engineering, Stanford University, Stanford, CA. 363 p.) The appropriate range in K_d values for model use at RFETS is not well constrained. Preliminary assessment of data for Pu in groundwater suggests that K_d values are likely to be on the order of several thousand $L\ kg^{-1}$. Current soil action level calculations use a 'site' K_d of $200\ L\ kg^{-1}$.

K_d values will be determined for soil isolates taken from the sampling sites using 903 Pad area seep water and ^{242}Pu as a yield tracer. Typically, 1 - 10 g of the soil is suspended in 10 - 100 ml of the target solution phase, the yield tracer is added and the fraction of the yield tracer remaining in solution is determined. The K_d value is the ratio of the particulate to solution-phase Pu, divided by the mass concentration of the suspended soil. These yield tracer experiments will be compared to the 'exchangeable fraction from the selective extraction (i.e., phase speciation) experiments.

The extent of Pu partitioning between particles and solution depends on time. This kinetic effect is manifest for both sorption (the transfer of Pu from the water phase to the particle phase) and desorption steps. Material from one box core will be used to evaluate sorption and desorption kinetics using standard procedures (e.g., Quigley, M.S., B.D. Honeyman and P.H. Santschi, 1996, Thorium sorption in the marine environment: equilibrium partitioning at the hematite/water interface, sorption/desorption kinetics and particle tracing. *Aquatic Geochemistry*, 1, 277-301).

2.1.4. Expected results: A preliminary Pu phase speciation and mobility 'map' for the 903 Pad 'lip' sampling areas. K_d and Pu phase speciation distribution across the lip area and with soil depth. Specific Pu soil activities (e.g., pCi g^{-1}) in SID and Pond C-2 sediments. An assessment of the mobile form of Pu. For example, if 20% of the Pu in lip area soils is associated with the soil organic fraction but 75% of the pond Pu is in the form of organic complexes then the likely mobile form of Pu is soil organics. This information is necessary for evaluating long-term surface water protection strategies.

2.2. Task 2: Analysis of mass loadings to RFETS Ponds.

2.2.1. Objective:

To use selected RFETS ponds as a time-integrator for basin discharge of Pu.

2.2.2. Justification:

Ponds C-2, B-1 and B-5 were selected as likely having sediments which will archive the historical runoff history of the basin. The episodic nature of storm events and constraints on the number and type of stream monitoring stations make a *per event* evaluation of Pu mobility within basins difficult. However, RFETS ponds can serve as sampling 'stations' to the extent that ponds function as efficient traps for particulate runoff from basin areas. Because essentially all of the mobile Pu is associated with particles, the inventories of Pu in the ponds (e.g., the total activity; Bq) provides a means of assessing the total and yearly Pu that is transported *to the ponds* as the consequence of storm events.

2.2.3. Analytical Plan:

See Sampling Procedures, below. One complete core from each pond for a time-series analysis of Pu loadings to the ponds; six additional core 'homogenates' for 1) inventories; 2) phase speciation. Inventories will be determined by total digestion of sediment homogenates coupled to an estimated of the pond sediment loadings. Sediment chronologies will be evaluated by gamma counting of ^{137}Cs in core slices. If the sediments retain their depositional structure, bomb fallout ^{137}Cs can be used to 'date' the depositional age of the Pu. The determination of the amount of Pu associated with a sediment of a certain age can provide information on the yearly transport of Pu to the ponds.

2.2.4. Expected results: An estimate of the yearly average accumulation of Pu in pond sediments determined from the total Pu inventory in the ponds divided by the pond age: χ Bq per basin per year. If pond sediments are not homogenized by pond processes, a yearly record of Pu loadings through correlation of Pu specific activity (Bq/g) and sediment age.

2.3. Task 3: K_d values for U(VI) interaction with solar pond soil isolates.

2.3.1. Objective:

To provide preliminary K_d data for U(VI) interaction with solar pond area soils.

2.3.2. Justification

Remediation strategies require knowledge of the mobility of U(VI) in the groundwaters influenced by the solar ponds. U(VI) is relatively mobile compared to Pu and Am as the