

DEVELOPMENT OF AN INDEPENDENT SCIENTIFIC ADVISORY COMMITTEE TO SUPPORT THE CLOSURE OF ROCKY FLATS

C. S. Dayton
Kaiser-Hill Company, L.L.C.

I. B. Paton
Wright Water Engineers, Inc.

ABSTRACT

The Rocky Flats Environmental Technology Site (RFETS) is a Department of Energy (DOE) facility near Denver, Colorado that was formerly used to produce nuclear weapons components. The site is currently undergoing environmental remediation and is scheduled to become a National Wildlife Refuge by 2006. Understanding the migration processes of the site's environmental contaminants, particularly actinides such as plutonium (Pu), americium (Am), and uranium (U), is necessary to develop a successful strategy for remediation and long-term stewardship of the site. In 1996, the Actinide Migration Evaluation (AME) advisory group was formed with personnel from universities, government agencies, and DOE National Laboratories, with expertise in subjects ranging from actinide chemistry to erosion processes, to document and facilitate better understanding of environmental actinide transport processes. The AME acts in an independent capacity, advising on site operations, integration, communications, and studies necessary to answer actinide-related questions related to the site's cleanup mission.

To provide a comprehensive overview of the mechanisms that impact actinide transport in the environment, the AME developed a site-specific conceptual model. To verify specific components of the conceptual model, an actinide migration pathway analysis was conducted, using measured field data and computer models, to quantify actinide transport via the air, surface water, groundwater, and biological pathways. Findings from this study indicate the air and surface water pathways are the dominant transport mechanisms at RFETS, particularly for Pu and Am, because of their low solubility and resulting transport in particulate form. Overall, the estimated amount of uranium activity transported off-site far exceeds the Pu and Am activity transported because of the abundant amount of natural uranium in the RFETS environment.

Projects supported by the AME group cover a broad range of topics. For example, at the 903 Pad remediation site, AME guidance was provided to identify the appropriate analytical method to determine the chemical species and oxidation state of plutonium (Pu) in soils. X-Ray Absorption Fine Structure (XAFS) analysis was employed to determine the species to be $\text{PuO}_2 \cdot x\text{H}_2\text{O}$, and X-Ray Absorption Near Edge Structure (XANES) spectroscopy was used to confirm the oxidation state is Pu(IV). These results provided verification that the Pu is highly insoluble, and support Radioactive Soil Action Levels that focus remediation efforts on the surface soils most apt to be transported by wind and water erosion processes. The XAFS and XANES results also helped to alleviate concerns about the potential for dissolved Pu to be transported in groundwater. Other AME-supported work in the 903 Pad area involved modeling of surface water erosion and actinide transport processes. The Water Erosion Prediction Project (WEPP) erosion model, HEC-6T sediment transport model, and soil actinide data were combined to develop predictions of actinide erosion and surface water actinide concentrations, for a range of storm events. Results from AME-supported investigations are frequently presented to stakeholders that include regulatory agencies, community representatives, and public interest organizations. These meetings help all parties involved with the RFETS closure to develop a better understanding of actinide transport mechanisms, and the implications for closure of the site.

INTRODUCTION

RFETS is a Department of Energy (DOE) facility that was formerly used to process and manufacture nuclear weapons components. RFETS is located approximately 16 miles northwest of Denver, Colorado, and has several neighboring communities. Production operations at the Site were discontinued in 1989. While the facility was in production mode, the primary actinides used were plutonium-239 (Pu), americium-241 (Am), and uranium (U). Historic operations caused contamination of the site with these radionuclides as well as with chlorinated hydrocarbons, including carbon tetrachloride, trichloroethene, tetrachloroethene, and others. Currently, the focus of work involves decommissioning and demolishing buildings and facilities, and remediating environmental contamination. Following closure, the site will be converted into a National Wildlife Refuge. This transition is scheduled to occur by 2006.

RFETS encompasses approximately 6,585 acres. Major structures are located within a centralized 385-acre Industrial Area, which in the late 1980s comprised over 440 structures used for manufacturing, chemical processing, laboratory, and support activities. A 6,200-acre grassland Buffer Zone surrounds the Industrial Area (see Fig. 1). Because of the close proximity of RFETS to a large metropolitan area, there is, expectedly, a large amount of public interest in the site's remediation and closure. Multiple stakeholder organizations are involved in monitoring the site's closure progress and remediation planning, including regulatory agencies, neighboring communities, and other public interest groups.

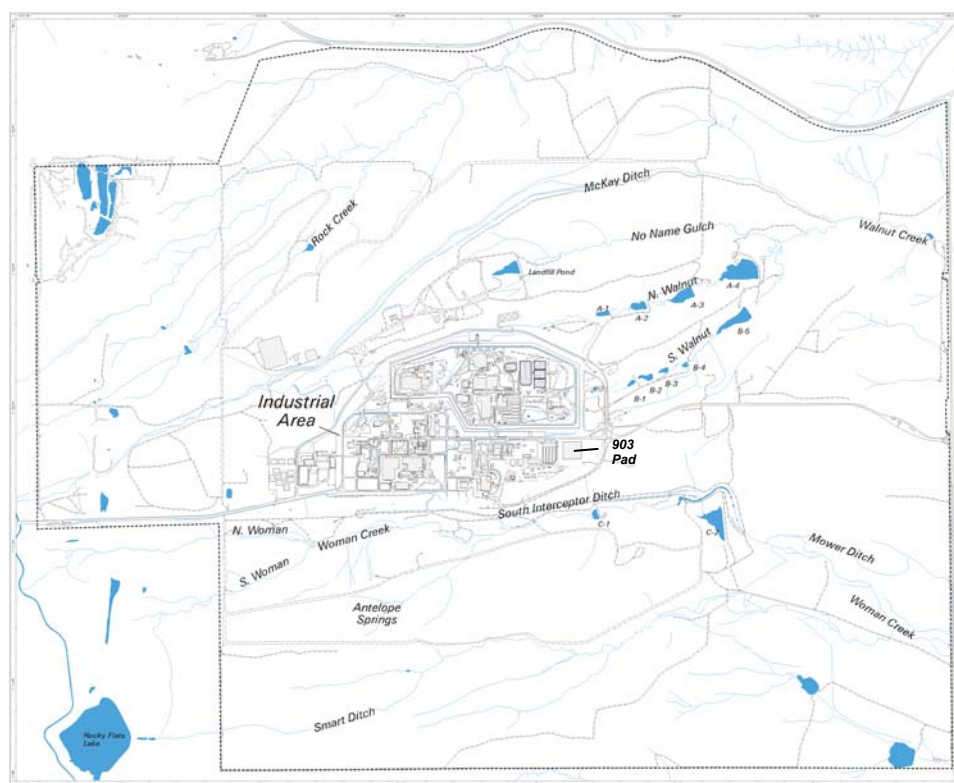


Fig. 1. The Rocky Flats environmental technology site includes a 385-acre industrial area surrounded by a 6,200-acre buffer zone.

Questions are continually raised regarding the proper approach for remediating RFETS as a result of the large scope and complexity of the site and its contaminants. Answers to these questions, with a well-founded technical basis, are required if the remediation strategy is to satisfy DOE, regulator, and community concerns. To address difficult questions related to actinide transport in the environment, DOE and Kaiser-Hill Company, LLC, the integrating contractor at RFETS, formed an advisory committee composed of members with expertise in a range of subjects. Scientists were brought together from the DOE National Laboratories, government agencies, and academia to provide ongoing guidance in fields such as actinide chemistry, geochemistry, erosion processes, colloidal transport, and microbiology.

HISTORY OF THE RFETS ACTINIDE MIGRATION EVALUATION

In 1995, wet spring conditions at RFETS caused questions to be raised by site personnel, as well as stakeholder groups, about the mobility of Pu in the environment and, in particular, the potential for Pu to be solubilized under reducing conditions. This hypothetical scenario raised concerns about the potential for Pu mobility in the environment to be greatly increased. When coupled with other questions regarding the mobility of different actinides at different locations, this event prompted DOE and Kaiser-Hill (the site integrating contractor) to establish in 1996 the Actinide Migration Evaluation (AME) advisory group.

The makeup of the advisory group is constantly evolving, as new members are added to provide expertise on particular subjects, and as active advisors complete their support when specific questions are resolved. Meetings of the AME group generally take place once per quarter, and often include a meeting with stakeholders.

Initial members of the AME were David Clark (actinide chemistry, Los Alamos National Laboratory [LANL]), David Janecky (actinide geochemistry, LANL), Bruce Honeyman (actinide geochemistry, Colorado School of Mines), and Peter Santschi (colloidal transport, Texas A&M University at Galveston). Other advisors who have served or who continue to support the AME include Sumner Barr (airborne transport processes, LANL [retired]), Gregory Choppin (actinide chemistry, Florida State University), Arokiasamy Francis (microbiology, Brookhaven National Laboratory), Annie Kersting (geochemistry and colloidal transport, Lawrence Livermore National Laboratory), Leonard Lane (erosion processes, U.S. Agricultural Research Service [retired]), and Kirk Nordstrom (hydrogeology, U.S. Geological Survey).

Specific goals identified by the AME were to answer the following questions in the order of importance shown:

- **Urgent**: What are the important actinide migration sources and migration processes that account for current increased levels of actinides in surface water?
- **Near-term**: What will be the impacts of actinide migration on planned remedial actions? To what level do sources need to be cleaned up to protect surface water from exceeding regulatory action levels for actinides?
- **Long-term**: How will actinide migration affect surface water quality after site closure? What soil action levels are necessary to be sufficiently protective of surface water over the long-term?
- **Long-Term**: What is the long-term off-site actinide migration, and will it impact downstream areas (e.g. accumulation)?

From its inception, AME members have acted as independent advisors on RFETS studies, versus working as principal investigators. The independent capacity of the AME was identified as crucial for fostering an environment in which the advisors provide unbiased guidance and review of scientific investigations. To clarify the advisor-client relationship, a Memorandum of Understanding was written for the AME group

members, DOE, and Kaiser-Hill, which spells out roles and responsibilities of the various team members. On occasion, if necessary, some advisors have switched positions and taken on the role of investigator.

A central principle of the AME, from its inception, has been to have ongoing interaction with the public. As actinide migration-related studies are performed and completed, meetings are held with representatives from the regulatory agencies, neighboring communities and citizens' groups to discuss study results and the implications for remedial actions at the RFETS. If warranted, additional personnel are brought in to provide expert opinions as necessary (e.g., chemists experienced with specialized analytical techniques). Discussion of issues occurs in the public forums, and has prompted additional research to be conducted to address unanswered questions.

Conceptual Model for Actinide Transport

In 1998, a conceptual model was developed by the AME to provide a comprehensive overview of the mechanisms that impact actinide transport in the environment (Kaiser-Hill, 1998). A conceptual model was developed for Pu and Am, because these actinides have similar dominant transport characteristics based on their similar, low solubilities and minor background concentrations in the environment. A separate conceptual model was developed for uranium, which has different dominant transport mechanisms because of its higher background concentrations and greater solubility. These conceptual model diagrams are displayed in Fig. 2. Since being developed, the conceptual model has been used to guide investigations to provide a better understanding of the different actinide transport pathways and their relative importance. Therefore, the understanding of actinide movement at RFETS (and the conceptual models) are continually being refined as new studies provide additional information to confirm or modify individual transport pathways.

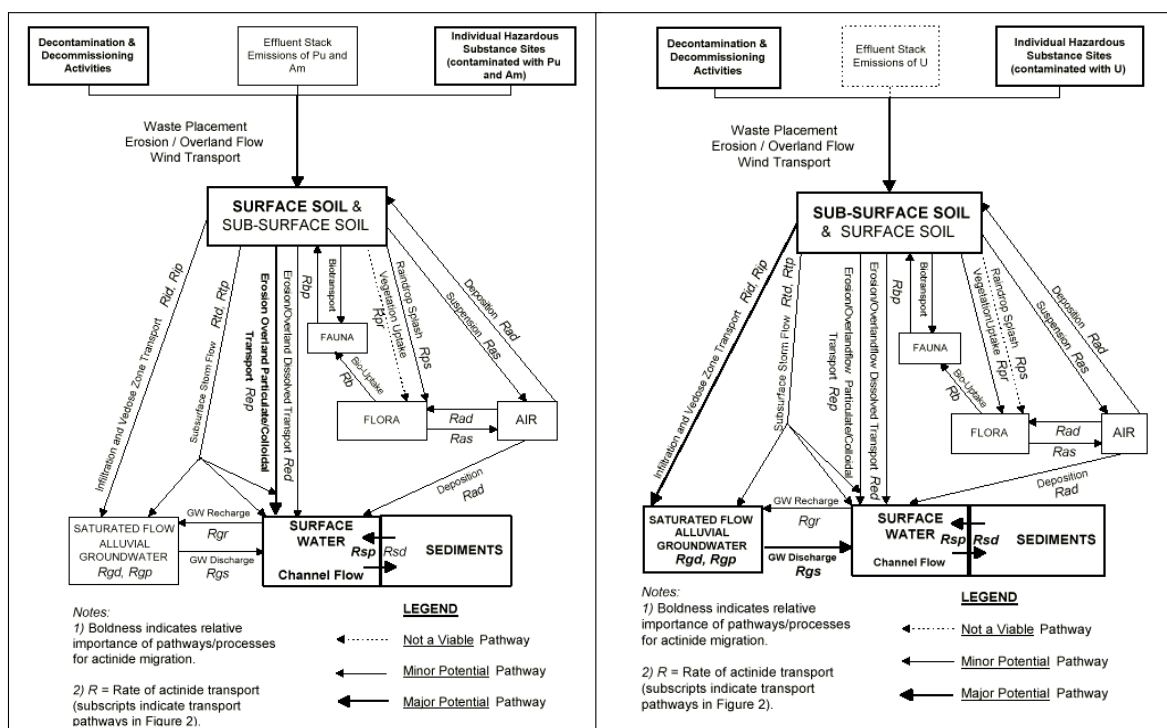


Fig. 2. Conceptual models for Pu and Am transport (left) and U transport (right) at RFETS. Bold text indicates those pathways that were thought to be the most important at the time the models were developed. The conceptual model evolved as greater understanding was gained.

Actinide Transport Pathway Analysis

An analysis of actinide transport pathways was conducted by the AME to verify major components of the conceptual model. This investigation involved developing quantified estimates of the Pu, Am, and U transported off-site annually by wind, surface water, groundwater, and biological transport mechanisms (Kaiser-Hill, 2002). Actinide transport estimates were developed using a combination of measured and modeled data. For example, estimates of surface water actinide loads transported off-site were developed using stream flow gaging data coupled with water quality data, collected by automated samplers that use a flow-weighted composite sampling protocol. Airborne actinide transport was estimated using wind data, in 15-minute time steps, coupled with air samples of actinides collected at the site boundary. Groundwater actinide transport was estimated using groundwater flux estimates, generated with the MIKE SHE integrated hydrologic flow model, in conjunction with groundwater quality data collected from shallow groundwater wells. Finally, biological transport of actinides was estimated for mule deer, the most abundant and mobile biological vector in contact with the soil, using a calculation based on their population, movement, and soil ingestion. The quantified analysis findings indicate, as anticipated, that the air and surface water pathways are the dominant transport mechanisms, particularly for Pu and Am, because of their low solubility and, consequently, transport in particulate form. Overall, the estimated amount of uranium activity transported off-site far exceeds the amount of Pu and Am activity transported because the abundant natural uranium is incorporated into the analysis for each of the pathways.

A summary of the pathway analysis results, with an order-of-magnitude comparison of the radioactivity transported by each pathway, for each isotope studied, is displayed in. These results support the RFETS closure regulatory strategy, as specified in the Rocky Flats Cleanup Agreement (RFCA), to focus remedial actions for actinides on surface soils, particularly for Pu and Am, because they are transported predominantly by erosion of contaminated surface soils. Findings from the pathway analysis were presented to the public in a workshop where each of the major pathway analyses was discussed, along with the actinide chemistry that dictates their behavior in the environment.

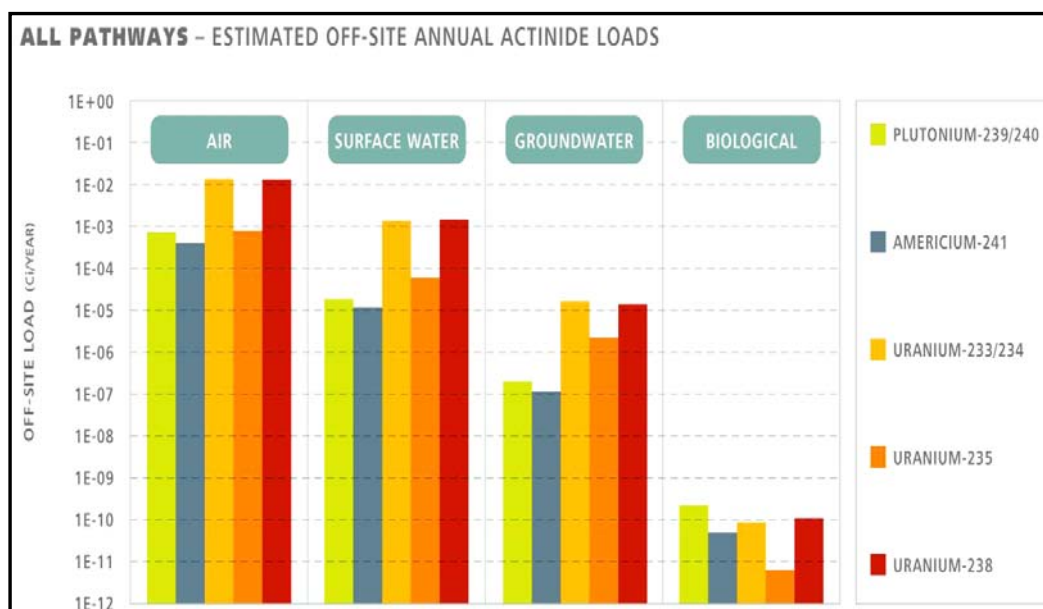


Fig. 3. Estimates of Pu, Am, and U activities transported off-site (Ci/year) annually at RFETS via different transport pathways.

CASE STUDY: THE 903 PAD

Background

The 903 Pad is located in the southeast corner of the Industrial Area (see Fig. 1) and was first established in July 1958 as a drum storage area. Drums stored at the 903 Pad contained hydraulic fluids and lathe coolant contaminated with radionuclides, including Pu and U. The drums also contained vacuum pump oils, trichloroethene (TCE), tetrachloroethane (PCE), silicone oils, and acetone still bottoms (DOE, 1995). In 1964, it was determined the drums were leaking and contaminating the soil beneath, and contamination was detected in the air samplers at the east fence, over a mile from the 903 Pad, following high winds (ChemRisk, 1992). A total of 5,237 steel drums were stored on the Pad, of which approximately 420 leaked to some degree (ChemRisk, 1992 and DOE, 1995), releasing approximately 86 grams (g) (5.3 Curies [Ci]) of Pu to the soil (DOE, 1995).

Removal of the drums at the 903 Pad occurred from early 1967 through June 1968 (ChemRisk, 1992). An asphalt cap, approximately 400 feet by 400 feet in size, was placed over the most contaminated area. During the drum removal and regrading activities, wind and water erosion caused the dispersion of Pu-contaminated soil. This soil erosion, primarily to the south and east of the 903 Pad, resulted in the contamination of the "Lip Area" beyond the 903 Pad area. Remediation of the 903 Pad was performed in 2002 and 2003, and involved excavation and shipment of soils to an off-site disposal facility. Remediation of the outlying Lip Area soils is planned for late 2003 and 2004.

Analysis of Pu Chemical Form and Oxidation State

Historically, it was suspected that the Pu associated with the 903 Pad and surrounding area was in the oxide form, PuO_2 , with very low solubility (Little and Whicker, 1978). However, definitive proof did not exist to verify its chemical form and oxidation state. At the suggestion of AME advisors, X-Ray Absorption Spectroscopy analyses were performed by LANL personnel using the Synchrotron Radiation Laboratory at the Stanford Linear Accelerator Center. Analysis with X-Ray Absorption Fine Structure (XAFS) spectroscopy was employed to determine the species to be $\text{PuO}_2 \cdot x\text{H}_2\text{O}$. X-Ray Absorption Near Edge Structure (XANES) analysis indicated the oxidation state is Pu(IV). Decades of study reveal a range of solubility centered around 1×10^{-11} M for PuO_2 in water at near-neutral pH (Knopp, et al., 1999). Therefore, the X-Ray Absorption Spectroscopy analyses provide verification that the Pu at the 903 Pad, the largest source of Pu in the environment at RFETS, is in a form that is nearly insoluble in environmental conditions. Consequently, these analyses helped to alleviate concerns about the potential for dissolved Pu from the 903 Pad area to be transported in groundwater. The very low solubility of the Pu also dictates that its transport will occur via particulate transport processes, such as by wind and water erosion of surficial soil to which the Pu is bound. Consequently, use of control measures to minimize soil erosion during remedial actions is of primary importance. This result is also consistent with long-term RFETS operations that use retention ponds to settle particulate matter in surface water in the major drainages of Woman and Walnut Creeks.

The XAFS and XANES results support the RFCA Radioactive Soil Action Levels (cleanup levels) that focus remediation efforts on surface soils, the environmental media most apt to be transported by wind and water erosion processes. Consequently, the RFCA-based surface soil remediation standard for Pu is stringent. Soils with 50 picoCuries per gram (50 pCi/g) or more of Pu activity, if detected at depths from zero to three feet, must be remediated. The soil standard for Am is structured in a similar manner as for Pu, with an emphasis on the top three feet of soil, since Am, like Pu, has very low solubility in the environment. If Pu or Am activity is detected at depths greater than three feet, a risk-based screening process is used to determine the remedial actions, if any, that are necessary.

Another question raised by stakeholders, and related to actinide chemistry at the 903 Pad, concerned the chlorinated solvents originally dispersed from the leaking 903 Pad drums, and their potential effect on Pu solubility. To address this question, rather than suggest that a full-scale investigation should be conducted, the AME advisors prepared a brief paper summarizing the issues. The paper described the chemical environment necessary to dissolve PuO_2 (laboratory conditions of extreme pH and high temperature), compared with the geochemical conditions created by the solvents in soil and groundwater. It was demonstrated that the chlorinated solvents in the RFETS environment would not alter the chemical conditions sufficiently to cause the Pu to become solubilized.

Watershed Erosion and Actinide Transport Modeling

Additional work supported by the AME that addressed the 903 Pad area involved modeling of surface water erosion and actinide transport processes. The Water Erosion Prediction Project (WEPP) erosion model, developed by the Agricultural Research Service, was calibrated for the 903 Pad and Lip Area watershed, which runs off into a diversion channel called the South Interceptor Ditch (see Fig. 1), that flows into retention pond C-2. A range of storm event sizes were simulated using WEPP, which provides the estimated runoff hydrograph for specific hillslope areas, and also provides an estimated mass of soil eroded, distinguishing by particle size. To estimate stream channel sediment erosion and deposition, output from the WEPP model is routed into HEC-6T. Model output from WEPP and HEC-6T are combined with soil actinide data (either Pu or Am), using Geographic Information Systems (GIS) software and a spreadsheet model, to develop predictions of surface water actinide concentrations in the watershed (Kaiser-Hill, 2000).

As part of the erosion modeling process, actinide mobility maps were developed that couple the predicted soil erosion (mass eroded/unit area) with soil actinide concentration data to generate a map of predicted actinide mobility for a specific storm event. An example of this type of map is provided in Fig. 4. Actinide mobility maps and model results of Pu and Am concentrations in surface water provide improved understanding of Pu and Am mobility as a result of surface water erosion processes. The model results provide a means to identify areas that are major sources of Pu delivered to surface water, and also to identify areas that will provide diminishing returns, in terms of benefit to water quality, if the area is remediated.

Accordingly, the model results make it apparent that the largest Pu and Am loads delivered to surface water do not necessarily originate from areas with the highest concentrations of Pu and Am in the soil. It is the combination of soil erodibility and soil actinide concentration that dictates the quantity of actinides delivered to surface water. For example, the area east of the 903 Pad alongside the East Access Road has generally the highest levels of Pu and Am in the soil of any portion of the Lip Area. However, this area is relatively flat, with slopes of approximately 1%. As a result of the moderate slopes, this area is less erodible than other, steeper parts of the watershed and therefore has less soil eroded, with a corresponding reduced amount of associated Pu and Am transport. This type of interpretation of model output is beneficial for prioritizing areas that should be considered for remedial action, if necessary.

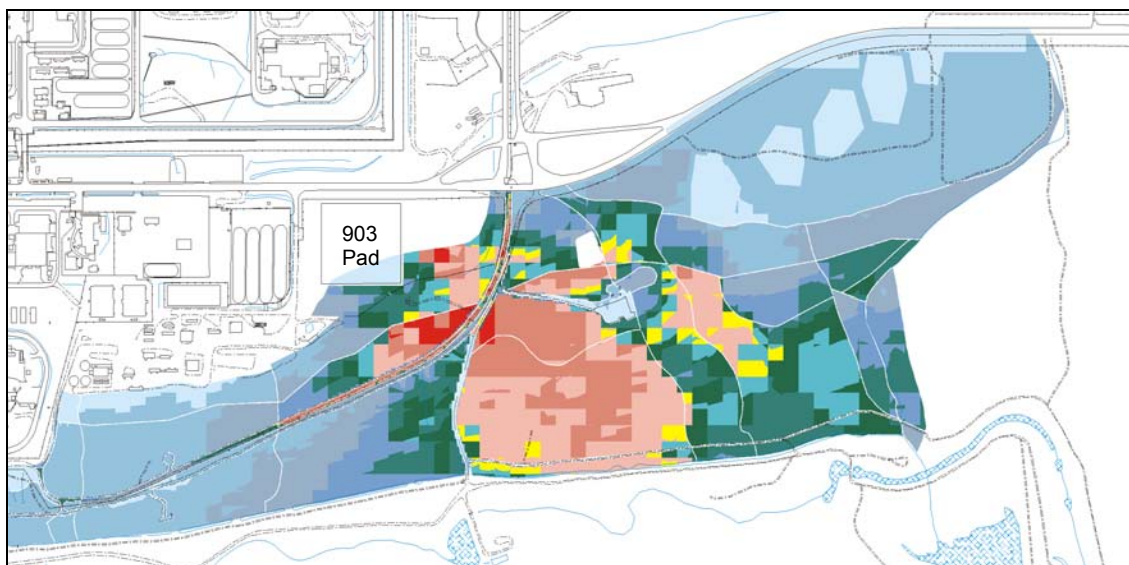


Fig. 4. Pu mobility map of the 100-year, 6-hour storm event (97.1 mm) for the 903 Pad and Lip Area watershed. Red indicates areas with highest Pu mobility; blue indicates areas with lowest Pu mobility.

SUMMARY AND CONCLUSIONS

Experience with the AME group at RFETS has shown that establishing an ongoing, external scientific advisory organization provides several benefits. First, formation of an advisory organization provides a mechanism that allows for specialized expertise to be readily available to address complex issues that on-site personnel may not be able or qualified to resolve on their own. Second, having an advisory organization composed of members who are not directly affiliated with the project site creates a more independent panel that is less prone to bias in their oversight and review of scientific investigations. The independence of the advisory group, combined with their scientific credentials, allows for development of their credibility with RFETS personnel and stakeholders.

The RFETS AME group has provided valuable guidance and review for a variety of investigations and modeling efforts. One location where AME input has been used to support site remediation is at the 903 Pad and adjacent Lip Area. X-Ray Absorption Structure analyses were used to determine that the Pu is in an oxidation state and chemical form with very low solubility (Pu(IV) as $\text{PuO}_2 \cdot x\text{H}_2\text{O}$). The low solubility was used as a basis for determining soil cleanup levels that focus on surface soil remediation, and as a basis for developing models to simulate wind and surface water erosion of insoluble Pu and Am in contaminated surface soils. These modeling tools were used to evaluate alternatives considered for the 903 Lip Area remediation.

As important as the AME group's oversight of investigations have been, of equal importance has been their ability to raise questions that otherwise may not have been considered. Continually pursuing answers to those questions has expanded the understanding of actinide migration at RFETS. In addition, keeping the stakeholders informed of study results has proved to be an essential component of the AME framework. Developing a better understanding of actinide transport mechanisms, and their implications for closure of the site, is beneficial for all parties involved.

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