

Rocky Flats Environmental Technology Site: Actinide Migration Evaluation (AME)

Meetings: January 31 to February 2, 2005

Advisory Group: Greg Choppin, David Clark, David Janecky, Leonard Lane

Summary and Recommendations for Path Forward

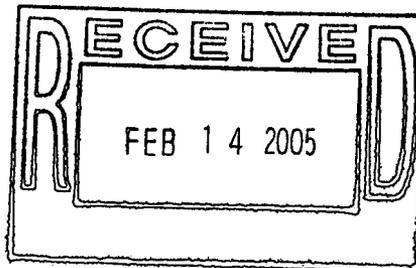
Contract completion is close, therefore, it continues to be very important to make the best decisions and track implementation to avoid re-doing work and raising questions as to the technical basis of the decisions and actions taken. Accomplishing these goals requires utilizing Site databases, modeling results, expertise, and documentation in plans and reports. The surface water monitoring projects continue to provide critical quantitative understanding of present controls and impacts. While erosion control efforts show expected response in contaminant transport, the Advisors recommend and encourage persistence, and even targeted enhancement. Surface water samples collected on the Site (in and above the ponds) may continue to have Pu and/or Am concentrations above regulatory limits, due to soil disturbance, remediation and demolition activities that create new disturbances and aggravate surface water response, erosion, and contaminant transport. Reconfiguration evaluation throughout the Site requires close attention at this time. These D&D, remediation, and other soil disturbance activities are critical in achieving closure and maintaining environmental protection. As contaminated areas are remediated to required standards and criteria, it is important to document the rationale and analytical basis. Transition to long-term stewardship (legacy management) and land configuration analyses are expected to be a focus of the next AME meeting(s).

Progress and Integration

The advisors identified two current D&D building sites where there are opportunities for enhancement in uniformity of attitude toward and the extent of in-place erosion control measures (e.g. one site had a detention reservoir and berm system to contain dust suppression water onsite while the other did not yet anticipate such approaches). We advise that increased integration and opportunities for exchange of best-management approaches be taken across all building D&D sites. We also recommend measuring the amount of dust suppression water applied to each area for mass balance evaluation.

We recognize that D&D, RSAL, and water quality standards range from nCi levels, to 50 pCi/g RSAL's, to the 0.15 pCi/L water quality limits. We recommend additional efforts to remind all personnel involved in D&D activities, soil remediation to RSAL, and water quality that all are responsible for protecting surface water quality through erosion control and other best management practices. The Advisors requested information on erosion control for the B-pond soil pile near Building 776, and were pleased with the information that the pile was contained within a bermed area and the surface was treated to minimize potential for spread of contamination.

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The advisors were impressed by the Site's response to the monitoring results, which revealed the presence of americium at SW093 and in Terminal Pond A-4. The extensive on-site knowledge of the overall water balance, drainage systems, and water flow paths allowed Kaiser-Hill personnel to rapidly identify the source of contamination, model it, and recommend swift action to both remediate the ponds, and to eliminate the pathway. The extensive background and understanding of actinide migration provided through the Pathway Analysis afforded the scientific understanding that the americium contamination was likely colloidal, and the Site's choice of co-precipitation and filtration to remove americium is based on this understanding. We were pleased with the overall integration between monitoring, modeling and treatment of the americium.

Results and Discussions

RFETS Project status

Building D&D continues at an impressive rate, with Buildings 371 and 776 the remaining major actinide processing facilities to be removed. Since the previous meeting, Buildings 771 and 707 D&D has been completed.

The latest revision of land configuration plans has been released. Three functional channels (FCs) for surface water flow across the Industrial Area have been defined and are being developed. In planned actions at the ponds, sediment removal is continuing from the upper B-series ponds on Walnut Creek. Notching of the Pond C-1 dam on Woman Creek is nearly complete, and the outlet works of Pond C-2 have been inspected.

The next meeting of the AME Advisors is scheduled for June 3-5, and listings of records retained by the Advisors have been requested by Chris Dayton (K-H). It is expected that non-duplicated records will be provided to Kaiser-Hill on paper copies (and electronic format, if available) for the October meeting of the Advisors.

Surface Water Quality – Current Status & Erosion Control

Ian Paton, WWE, presented a summary of recent results and activities on water quality in North Walnut Creek. This summary reviewed the activities for June 2004 through January 2005 in a timeline. This included a description of the events leading up to the identification of ^{241}Am contaminated drainage water from D&D activities at Building 771 as the source of elevated ^{241}Am concentrations in Pond A4.

George Squibb, URS, followed this with an overview and interpretation of surface water monitoring data from July through December, 2004 and demonstrated that the drainage areas contributing directly to gauging station SW093, and particularly, Building 771 as the source area for elevated ^{241}Am in Pond A-4. Load calculations for the area directly tributary to gauging station SW093, and not contributed by monitored sub-drainages, between July 1 and December 12, 2004, result in an estimate of 22.5 micrograms of ^{241}Am (associated with approximately 37.8 micrograms of $^{239,240}\text{Pu}$, and Pu/Am of less than 0.03 in Ci units). Evaluation of load contributions by sub-drainages is very consistent with identification of the ^{241}Am source within the footprint of Building 771. The total load of ^{241}Am is slightly larger than the estimated total inventory in Pond A-4 (0.6 pCi/L ^{241}Am in 24 million gallons gives approximately 18 micrograms ^{241}Am). Therefore, there may be significant ^{241}Am in contaminated sediment deposited (stored) in the wetlands below SW093 and above the A series ponds. The Advisors recommend that this wetland area be sampled to estimate the amount of ^{241}Am available for potential future transport to the A-series ponds.

For the same period, July through December, 2004 additional monitoring data indicated the major source area for $^{239,240}\text{Pu}$ upstream of gauging station SW093 was the drainage area above GS32, which monitored an integrated load of 178 micrograms of $^{239,240}\text{Pu}$ and an associated 0.8 micrograms of ^{241}Am (Pu/Am of approximately 4 in Ci units).

An overview of surface water quality data from Walnut Creek and the South Interceptor Ditch monitoring sites during water year 2004 to date showed that the additional erosion controls emplaced after June 2004 are apparently resulting in reduced $^{239,240}\text{Pu}$ and ^{241}Am concentrations.

Post-closure monitoring and surveillance

Scott Surovchak (USDOE/RFPO) and Sam Marutzky (SM Stoller, DOE/GJO) summarized progress on developing the Legacy Management transition, which includes a DOE subcontract to SM Stoller for management and operations. Counterparts between DOE EM and DOE LM have been identified as a result of meetings that were initiated in September 2003. Critical areas identified for transition include data records, communications and information technology (IT for technical application codes and databases). Points of contact have been identified in these areas. The biggest area requiring data and IT is environmental systems and characterization. This includes development of transition process and documentation of permits, and surface water and groundwater monitoring. Prior effort by DOE on legacy management (e.g. for the less complex Weldon Springs Site) is still ongoing, so this effort for the Rocky Flats Site is breaking new ground. Resource acquisition planning has identified substantial human resources, subcontract activities, and procedures that will be required. Much is already in existence, but will involve definition of management approach.

A draft Long-term Surveillance and Maintenance Plan has been developed, with release intended for 2006 due to explicit coupling with PCA and ROD. The continued emphasis by DOE management on perceptions of personnel issues as high risk components of Legacy Management, relative to environmental monitoring, maintenance and response requirements (particularly over the next few years) is an area of continuing concern to the AME Advisors. It is generally agreed that revegetation will need to continue as a Legacy Management activity. The first 3-5 years will be toughest for keeping ahead of erosion areas and identification of vulnerabilities in site stability. It must be expected to have people in the field to deal with those issues and not reinvent process/jobs and individuals. One of biggest challenges is to maintain the technical credibility and open communication mechanisms that have been developed.

Simultaneously, the CAB and RFCLOG will morph into a Local Stakeholder Organization (LSO). This is similar to organizations typical of CERCLA/RCRA sites. DOE LM plans on the commitment for annual meetings,

with quarterly meetings during at least first year and probably 2 years, primarily leading up to the second CERCLA 5-year review. In the event of actual exceedances, there are reporting requirements that are also written into the CAD/ROD.

Site Tour

The Advisory Group toured a number of D&D and monitoring activities on Site. Since the Site had received snow the previous night, the entire Site was muddy and wet, with standing water, flowing water and mud throughout all the D&D sites, and flowing water was observed in all the drainages. This was a good opportunity to re-emphasize the need to maintain vigilant control over contaminated soils, and the importance of erosion control in protecting surface water quality during and after D&D activities.

Chris Gilbreath (KH) toured the Advisors around the Building 371 D&D site and outlined the current approach for taking down the building. Terry Vaughn (KH) toured the Advisors around the Building 776/777 D&D site, and outlined the strategy for that building complex. The tour continued around the building 707, 771, 991, and 903 Lip Areas. We also observed the B pond remediation effort.

ER project update

Karen Weimelt (KH) provided a summary of the Environmental Remediation program and upcoming activities. Significant progress was reported in this area. In the East Firing Range, the excavation and reseeded has been completed. The remediation of B-1 Pond was almost completed, while that of Pond B-2 was about 50% finished and should be complete by the end of February. The preparation of the Portland cement to be used is 80% done. The sediment removal has begun for Pond B-3. The Pond C-1 dam notch project also is moving well to completion.

The remediation of the OPWL's was initiated in December 2003 and to date about a third of the OPWL has been removed from the open areas. OPWL remediation will continue as removal of buildings allow. About 30% of the OPWL's expected to be left in place as they can be closed in situ.

Remediation of other areas with VOC and PCB contamination (Oil Burn Pit #2) and of Pb (North Firing Range) are planned in the near future.

Progress was also reported in ER characterization at a number of sites. The work appears well directed, well connected to other teams across the site, and moving forward at a good pace.

Pond reconfiguration, management and remediation B-ponds

The report on the status of the B-ponds remediation by Mike Keating reflected the significant progress made. One factor that slowed progress was the

very high moisture level of the sediments (200-300%). However, the use of Portland cement allowed reduction to 20-25% with no significant associated fines. This has resulted in good progress in removal of contaminated sediments both in bags and by simple loading in trucks. Some of the soil will be used as bracing material in rail car shipments of Building 776 materials. The bagged soil will be directly shipped off site for disposal.

It was reported that excavation of the sediments in Pond B-1 was completed January 21, and would be complete in Pond B-2 by February 15, and in Pond B-3 by February 28. Successful completion by these times will meet planned deadlines.

Erosion controls program – description/status.

Three major sources of information were provided to describe the RFETS erosion control program: (1) The analyses and findings presented by Ian Paton, WWE, and George Squibb, URS, described earlier, (2) the Advisors field trip on Monday January 31, 2005, and (3) the presentation and handouts provided by Steve Nesta, K-H, on February 1, 2005.

We are very encouraged by RFETS' level of awareness and dedication to the importance of erosion control in protecting surface water quality (i.e. maintaining actinide concentrations below the 0.15 pCi/L standard) at the POE's and POC's. Of particular note are the documented management plans and operationally linked maps (i.e. "Rocky Flats Environmental Technology Site Erosion Control Management System, the GIS-based maps of current and pending areas for erosion control) that have resulted in a system actively utilized by management, field personnel and DOE. The related discussions during the AME meeting, showed strong evidence of implementation, and adaptive management, contributions to best practices.

However, during our field trip, we did visit two current D&D building sites where there were apparent differences in the uniformity of attitude toward and the extent of in-place erosion control measures. For example, one major site had a detention reservoir and berm system to collect and contain dust suppression water onsite while the other did not. We advise that more effort be taken to provide expertise to D&D project managers with the end goal of increasing integration and thus consistency across all building D&D sites. We also recommend measuring the amount of dust suppression water applied and recovered at each site for mass balance calculations and evaluation of water and contaminant migration during and following these operations.

We recognize that D&D, RSAL, and water quality standards range from nCi levels, to 50 pCi/g RSAL's, to the 0.15 pCi/L water quality limits. We recommend additional efforts to remind all personnel involved in D&D activities, soil remediation to RSAL, and water quality that all are responsible for protecting surface water quality through erosion control and other best management practices. The Advisors requested information on erosion control for the B-pond

soil pile near Building 776, and were pleased with the information that the pile was contained within a bermed area and the surface was treated.

A-Ponds water treatment and management plans.

Steve Nesta provided a report on the status of the A-ponds water treatment and management plans. As a result of routine monitoring of Terminal pond A-4 for discharge, americium contamination was identified at a level of approximately 0.6 pCi/L. This was confirmed by numerous subsequent analyses. Steve outlined a number of options that were considered for treatment of pond A-4 water, including conventional filtration, sorption, and co-precipitation (flocculation) followed by filtration. The extensive background and understanding of actinide migration provided through the Pathway Analysis afforded the scientific understanding that the americium contamination was likely colloidal. A discussion of americium geochemical behavior can be found in the appendix. The Site's choice of co-precipitation with calcium phosphate and filtration to remove americium is based on this understanding, and we are convinced that this treatment methodology should result in removal of the majority of colloidal material. We were pleased with the overall integration between monitoring, modeling and treatment of the americium contamination in pond A-4.

Now that this treatment plant has been set up on site, we feel that it would be prudent to maintain it until after the B776 and 371 D&D activities are complete, and that on-site monitoring shows no elevated actinide concentrations as a result of D&D activities. This would also protect against any possible future transport of any radionuclides trapped, or held up, in the wetlands between gauging station SW093 and the A-series ponds.

Documents Provided to Advisory Group

Agenda for meetings
Boulder Daily Camera 1/7/2005 – editorial: risks and realities, critics go too far
in criticism of Rocky Flats cleanup
Denver Post 1/21/2004 – slight taint among Flats deer
Rocky Flats Envision, v10, n18 Dec 15, 2004 – B707 reduced to rubble
Rocky Flats Envision, v11, n1 Jan 19, 2005 – 2004 in review: our annual year-
end roundup
Land configuration plan figure
North Walnut Creek water activity viewgraphs
South Walnut Creek data viewgraphs
ER projects update viewgraphs
B pond remediation update viewgraphs
Rocky Flats environmental technology site erosion control management
system, Rev 1, February 2005
A-4 Water Treatment calculation CALC-000-NA-000776Rev0

Documents and Information Requested for Advisory Group

Monitoring updates as available, particularly surface water, erosion, ponds, &
building D&D

Requests for Future Presentations and Information

Update on surface water quality monitoring data
Land configuration & hydrologic analysis – Status, analyses and path forward
Pond remediation and operations results
Update on transition to stewardship

Participants in AMS technical meetings

Name Organization

Chris Dayton	Kaiser-Hill
Greg Choppin	Florida State
David Clark	Los Alamos
David Janecky	Los Alamos National Laboratory
Leonard Lane	Tucson
Ian Paton	Wright Water Engineers
George Squibb	URS
Bob Nininger	Kaiser-Hill
Sam Marutzky	SM Stoller/GJO
Scott Surovchak	DOE LM
Melvin Madril	SM Stoller/GJO
John Stover	DOE/RFPO
Chris Gilbreath	Kaiser-Hill
Terry Vaughn	Kaiser-Hill
Karen Weimelt	Kaiser-Hill
Mike Keating	Kaiser-Hill
Steve Nesta	Kaiser-Hill
David Shelton	Kaiser-Hill

Future Meetings

June 5-8, 2005
October 2-5, 2005

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Appendix**Americium Geochemical Behavior at RFETS
Actinide Migration Evaluation – Advisory Group**

February 12, 2005

Background. Extensive research has been conducted in the U.S. and internationally on the environmental behavior of actinide elements in a very diverse set of environments over the past 30-40 years. This has provided a rather good base of understanding of the major types of species and their transport mechanisms in soils and natural waters. A dominant, and often controlling feature of plutonium (Pu) and americium (Am) geochemistry is their low solubility in natural waters, and their strong tendency to adsorb to soil and mineral surfaces. In many cases, Pu and Am adhere to the geological matrix, and remain immobile in the environment. There are also a number of field studies documenting that small concentrations of low-solubility radionuclides,^[1-4] such as Pu and Am can be transported in surface or groundwater through the association with naturally occurring particulates whose small size (1 nm – 1 μ m).^[5,6] These small particles remain suspended and are therefore transported in natural aquatic systems. These geochemical behaviors are related to the hydrolysis and solubility of Pu and Am.

Hydrolysis and Solubility Characteristics. The hydrolysis and solubility of Am(III) has been studied by a number of authors, and a critical review of the chemical thermodynamic data of Am has been published by the Nuclear Energy Agency (NEA) in 1995,^[7] with an update in 2003.^[8] The study of Am(III) hydrolysis is extremely complicated due to the propensity of the Am(III) hydroxide products to both adsorb to surfaces and precipitate from solution. In controlled laboratory experiments, as the pH is increased towards neutral and basic conditions, the solubility of Am(III) decreases, and should be controlled by formation of crystalline Am(OH)₃ solid. These observations are reflected in the experimental solubility curve for Am(III) shown in Figure 1, which represents integration of data from multiple studies. The unusual units of $\log[Am]_{tot} - \log k_{sp}$ were used by the NEA reviewers to compare data from multiple authors under different solution conditions. The solid line in Figure 1 represents the solubility calculated with the NEA recommended equilibrium constants, and the dotted curves show the uncertainties ($\pm 3\sigma$) associated with the recommended value.

In aquatic environments, the carbonate anion is the only naturally occurring inorganic ion with sufficient complexing strength to compete with hydrolysis reactions. The calculated solubility for the Am(III) hydroxide-carbonate system under low ionic strength and carefully controlled laboratory conditions (25°C, fixed partial pressure of CO₂) is shown in Figure 2. Note that at very low carbonate concentrations (extremely low pCO_2), the calculated solubility curve is essentially identical to that shown in Figure 1, with a corresponding solubility of ca. 10^{-10} mol/L at pH 11, and 10^{-7} mol/L at pH 7. This curve is shown with a bold

line in Figure 2. As the carbonate concentration is increased, the calculated Am(III) solubility curve exhibits a minimum value as pH increases and then

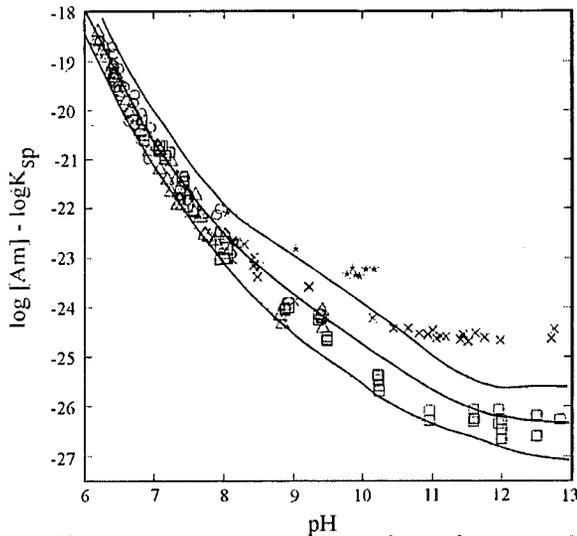


Figure 1. Experimental and calculated Am(III) solubility as a function of pH.^[6]

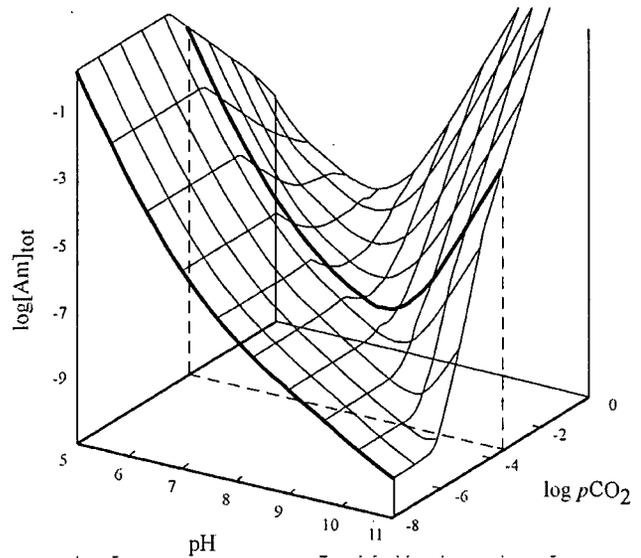


Figure 2. Calculated solubility diagram for the Am(III) hydroxide-carbonate system.^[6]

increases as carbonate complexes form. Under natural atmospheric conditions, the partial pressure of CO_2 corresponds to a value of 0.0003 atmospheres ($\log p\text{CO}_2 = -3.5$). The calculated Am(III) solubility curve for this $p\text{CO}_2$ concentration is also highlighted with a bold line in Figure 2. From this calculated solubility curve, one can see that under high pH conditions in equilibrium with atmospheric CO_2 , the solubility of Am(III) is expected to decrease to a minimum of 10^{-7} mol/L near pH 8.5, and then increase with increasing pH.

In the field, one rarely finds Am concentrations in natural waters that approach the calculated concentrations shown in Figure 2 because of the propensity for hydrolyzed Am to adsorb to mineral and soil surfaces in the environment. For example, the calculated solubility of Am based on laboratory experiments is orders of magnitude higher than any observed Am concentration in RFETS waters. The high specific activity of ^{241}Am (3.24 Ci/g) is such that a solubility-limiting concentration of 10^{-10} mol/L corresponds to an ^{241}Am activity of 78 nCi/L. Thus the solubility limits for Am are tens of thousands of times greater than measured values found at RFETS. Site-specific data indicate that Pu and Am in RFETS ponded waters have an extremely low solubility, with concentrations in the femtomolar (10^{-15} mol/L) range, similar to global fallout. Unlike the carefully controlled laboratory system, the tendency to adsorb to natural minerals and colloids dominates the geochemistry of Am.^[3, 4, 9-12]

Colloid Characteristics. Colloids are small, naturally occurring particulates that are ubiquitous in groundwater, and composed of inorganic minerals or organic species. These natural colloids originate from the mechanical weathering of rocks, plants and soil.^[13] In surface and shallow ground waters, colloids of organic matter are fairly common, while deep waters in fractured rock is generally dominated by colloids of inorganic materials.^[14-16] Colloid-facilitated transport of radionuclides can occur if those radionuclides strongly associate with the colloids, and the colloids themselves minimally interact with the stationary phase. The radionuclides that are most likely to be transported by colloids are of extremely low solubility; and this makes Pu and Am likely candidates for colloid-facilitated transport. When Pu or Am associate with natural inorganic and organic colloids they form an associated complex commonly referred to as pseudocolloids.^[9-12] In addition to association with inorganic and organic colloids, Pu(IV) and Am(III) ions are prone to undergo polymerization reactions under environmental conditions to form colloid-sized polymers, and these are often referred to as intrinsic colloids.

The Role of Colloids at RFETS. Several field studies have been undertaken at RFETS to assess the ability of colloids and particulates to transport Pu and Am in the surface and shallow groundwater. Surface water samples were collected from storm runoff and pond discharge between 1998 and 2000.^[3] The collected water contained low levels of Pu and Am. Results showed that greater than 90 percent of the Pu and Am were detected in the particulate ($\geq 0.45 \mu\text{m}$; 40-90%) and colloidal (c.a. 2 nm or 3 kDa - $0.45 \mu\text{m}$; 10-60%) fractions of the groundwater. Controlled laboratory studies of soil resuspension, which simulated storm and erosion events, confirmed most of the Pu in the $0.45 \mu\text{m}$ filter-passing phase was in a colloidal form ($\geq 80\%$).^[3] This state-of-the-art study showed that the low levels of Pu and Am in surface water at RFETS are associated with colloids and particulates, and not soluble forms of the actinide elements.

In an earlier field study conducted in 1984, shallow groundwater from a single well was filtered, and analyzed for radionuclides.^[17] Low levels of Pu were associated with the particulate and colloidal fractions. Colloid concentrations were low ($< 1 \text{ mg/L}$) and consisted predominantly of clays. This study documents that Pu is associated with the colloidal fraction of the groundwater; yet, the low concentration of colloids observed limits the ability of colloids to transport significant quantities of Pu or Am. The concentrations and corresponding colloid loads that are found at RFETS are low, and therefore colloids do not represent a significant source for transport of low-solubility radionuclides at RFETS.

Implications for Americium Behavior at RFETS. Field studies at RFETS have demonstrated that particulate- and colloid-facilitated transport of low-solubility radionuclides, such as Pu and Am, is the dominant mechanism for transport in the surface water and shallow groundwater pathways. The low concentrations of colloids detected in shallow groundwater wells, limits the

amount of Pu and Am that can be transported in this pathway. Studies carried out at RFETS have significantly improved our understanding of the *process* by which Pu and Am are transported, and give confidence to the recent pathway analysis that concludes that wind and surface water erosion are the dominant actinide migration pathways at RFETS.^[18] Unlike other geologic environments, deep groundwater at RFETS is effectively isolated from shallow and surface water, preventing a pathway for deep vertical transport of Pu and Am.

Pu and Am migration in the environment can occur because small amounts of these very low-solubility actinides can associate with particles or colloids (pseudocolloids) or are themselves colloid sized polymers (intrinsic colloids). Pseudocolloids are present in nearly all waters and are formed as a result of the weathering of rocks, soil and plant material. The Pathway Analysis Report^[18] shows that the two dominant pathways for Pu and Am transport are by air and surface water pathways. Shallow groundwater and biological transport were found to be only minor migration pathways. At RFETS, sedimentation and resuspension of colloids and particulates in both the surface and shallow groundwater represent the dominant processes for low levels of Pu and Am migration. Since colloids are a part of the overall particulate spectrum, the migration of Pu and Am as colloids at RFETS has been quantified in the Pathway Analysis Report.^[18]

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