ACTINIDE RESEARCH QUARTERLY

Rocky Flats: *From* *Weapons* *to* *Wildlife*

CONTENTS

Task completed a year ahead of schedule and under budget

Scientific understanding
aids *Rocky Flats* cleanup

This article was contributed by David L. Clark (PMT-DO), David R. Janecky (ENVP-AQE), Leonard J. Lane (EES-ACED and USDA-Agricultural Research Service, retired), and Gregory R. Choppin (Florida State University).

Background on the Site's contamination

The Rocky Flats Environmental Technology Site (RFETS) was a DOE environmental cleanup site located about 16 miles northwest of downtown Denver. Today more than 2.5 million people live within a 50-mile radius of the Site, and 300,000 of those live in the Rocky Flats watershed. From 1952 to 1989, the Rocky Flats Plant made components for the nation's nuclear weapons arsenal using various radioactive and hazardous materials, including plutonium and uranium, toxic metals such as beryllium, and hazardous chemicals such as cleaning solvents and degreasers.

The key component produced at Rocky Flats was the plutonium pit, commonly referred to as the "trigger." The pit provided energy to fuel the explosion of a nuclear weapon. In 1989, the FBI and the Environmental Protection Agency (EPA) abruptly halted nuclear production work to investigate environmental and safety concerns, and the Site was added to the Superfund list later that year. In 1993, the Secretary of Energy announced the end of the Rocky Flats nuclear production mission. Nearly 40 years of nuclear weapons production left behind a legacy of contaminated facilities, soils, surface water, and groundwater. Accordingly, RFETS was designated as an EPA Superfund cleanup site.

THE DENVER POST
Voice of the Rocky Mountain Empire
**** 50 cents
June 7, 1989

Federal Agents Raid Rocky Flats
By Tom Graph, Peter G. Chronis, Mark Obmascik, Alan Gottlieb
Denver Post Staff
Copyright 1989, The Denver Post Corp.

From 1952 to 1989, the primary mission of the Rocky Flats Plant was making components for nuclear weapons. A raid in 1989 by the FBI and the EPA for alleged environmental violations resulted in an abrupt halt to nuclear material production. Large quantities of plutonium and other hazardous materials were left in various stages of processing and storage, as shown in the top photo of the Building 707 storage area. The environmental cleanup was originally estimated to cost more than $37 billion and take more than 70 years to accomplish.

The sudden shutdown left large quantities of plutonium and other hazardous substances in various stages of processing and storage. Because plutonium is dangerous to human health, even in minute quantities, the cleanup of plutonium-contaminated materials is complex, tedious, labor intensive, and slow. In March 1995, DOE estimated the cleanup for Rocky Flats would cost in excess of $37 billion and take 70 years to complete. By 1996 DOE and Kaiser-Hill had initiated efforts that eventually resulted in a credible plan to accelerate the closure of Rocky Flats by December 31, 2006, at

Rocky Flats CLEANUP

Actinide Migration Evaluation team members gather at the Site in October 2005. From left to right are Gregory Choppin (Florida State University), David Clark (Los Alamos), Ian Paton (Wright Water Engineers), David Janecky (Los Alamos), Leonard Lane (U.S. Department of Agriculture, retired), Robert Nininger (Kaiser-Hill, LLC), and Christine Dayton (Kaiser-Hill, LLC).

a contracted cost of $7 billion. After a troubled start, Kaiser-Hill completed the task nearly a year ahead of schedule.

Many around the U.S. Nuclear Weapons Complex are analyzing the factors that led to the turnaround. Without question, contributing factors included the incentive-laden contract, strong support and stable funding from Congress, high-level DOE support that mobilized the entire complex to assist in the cleanup, technological and operational innovation, and scientific understanding. Much has been made of the contractor's fee of more than $500 million, but less has been said about the role that scientific understanding played in guiding key cleanup decisions and facilitating good project management.

The drive to understand the science behind plutonium contamination gained momentum in 1995 when intense rainfall and wet spring conditions raised concerns among Site personnel and stakeholder groups about the potential for increased plutonium mobility and offsite transport. There was a hypothesis that the plutonium was soluble in surface and groundwater, which would account for increased plutonium concentrations at onsite monitoring locations.

Modeling efforts and Site data at the time predicted no or very limited movement of plutonium. The prediction of no plutonium mobility coupled with the observation of plutonium transport at surface-water monitoring stations led to public mistrust and lack of confidence. When coupled with other questions regarding the mobility of different actinide elements (uranium, plutonium, and americium) at different Site locations, DOE and Kaiser-Hill established the Actinide Migration Evaluation (AME) advisory group in 1995 to provide advice and technical expertise on issues of actinide behavior and mobility in the air, surface water, groundwater, and soil.

Actinide Migration Evaluation projects

The Actinide Migration Evaluation (AME) projects were commissioned at Rocky Flats in 1995 to address how actinide elements move in the environment. Initially, AME advisors were recruited to evaluate and provide guidance on environmental conditions (including actinide chemistry, geochemistry, migration, and erosion) at RFETS. The charter was rapidly expanded to include recommendations of paths forward for long-term protection of surface-water quality as the primary technical and regulatory measure of remedial action quality.

Over the 10-year history, the group was led by Christine S. Dayton (formerly with Kaiser-Hill), and the following served as advisors: Sumner J. Barr (Los Alamos, retired), Gregory R. Choppin (Florida State University), David L. Clark (Los Alamos), Arokiasamy J. Francis (Brookhaven National Laboratory), Bruce D. Honeyman (Colorado School of Mines), David R. Janecky (Los Alamos), Annie B. Kersting (Lawrence Livermore National Laboratory), Leonard J. Lane (USDA Agricultural Research Service, retired), D. Kirk Nordstrom (U.S. Geological Survey), and Peter H. Santschi (Texas A&M University, Galveston).

The following overriding technical questions and priorities were identified:
- Urgent: What are the important actinide migration sources and migration processes that account for surface-water quality standard exceedances?
- 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 action levels for actinides?
- Long-term: How will actinide migration affect surface-water quality, airsheds, and impact on downstream areas (e.g., what soil-action levels will be sufficiently protective of surface water)?

A central principle of AME, from its inception, was to have ongoing interaction with the public. As actinide-migration-related studies were performed and completed, meetings were held with representatives from the regulatory agencies, neighboring communities, and citizens' groups to discuss study results and the implications for remedial actions at the Site. When warranted, additional personnel were brought in to provide technical expertise as necessary (for example, chemists experienced with specialized analytical techniques). Discussion of issues occurred in the public forums and prompted additional research to be conducted to address unanswered questions.

Through a combination of expert judgment supported by state-of-the-art scientific measurements, it was shown that under environmental conditions at Rocky Flats, plutonium and americium form insoluble oxides and colloids that adhere to small soil and mineral particles. These particles can migrate in the Rocky Flats environment by wind and surface-water resuspension and sedimentation processes.

The scientific understanding showed that soluble transport models were not appropriate and led to the development and application of erosion and sediment transport models. The scientific understanding developed through these integrated studies provided the basis for the negotiation of plutonium and americium cleanup levels selected by the Rocky Flats Cleanup Agreement (RFCA) parties of 50 picocuries per gram (pCi/g) of plutonium in surface soils. A curie is a measure of the amount of radiation emitted by a radionuclide. It is the quantity of a radionuclide that undergoes 37 billion disintegrations every second.

Did it save a lot of taxpayer dollars? That's more difficult to determine. What everyone agrees on is that it provided clarity and focus on the real issues surrounding plutonium and americium in the RFETS environment, and that allowed for good project management to move forward on Site cleanup. It helped all parties focus remediation efforts on surficial contamination and transport pathways that posed the greatest risk to human health and the environment. It helped guide selection of surface-specific removal technologies, and future land-configuration strategies.

Thus, scientific understanding, through advanced measurement techniques, was developed into science-based communication and decision-making for Kaiser-Hill and DOE that helped focus Site-directed efforts, aided the DOE in its effort to close RFETS in December 2005—one year ahead of schedule, and most certainly helped shave decades and billions of dollars off initial cleanup estimates.

A 1995 aerial photo of RFETS shows the Industrial Area and the surrounding, controlled open space.

Rocky Flats CLEANUP

DETAILS OF THE ROCKY FLATS SITE

Rocky Flats was similar to a small city. It comprised more than 800 structures on a 385-acre industrial area surrounded by nearly 6,000 acres of controlled open space. The open space serves as a buffer between Rocky Flats and the nearby growing communities and is home to many species of animals and plants. In addition to the industrial facilities, Rocky Flats had its own fire department, medical offices, cafeterias, garage, gas pumps, steam plant, and water- and sewage-treatment plants.

Water at RFETS and the surrounding area is distributed among surface water, shallow groundwater, and deep groundwater. Surface-water flow across RFETS is primarily from west to east, with three major drainages: North and South Walnut Creeks and Woman Creek in the Industrial Area. Walnut Creek flowed into the Great Western Reservoir about a mile and a half from the plant. In 1989, Broomfield built a diversion ditch around the reservoir to prevent surface drainage and runoff from Rocky Flats from entering the reservoir via Walnut Creek. In 1997, the city secured a new drinking-water supply; Great Western Reservoir is no longer used as a drinking-water source. Woman Creek drains the south portion of the site and before 1996 flowed into Standley Lake, which was a source of irrigation water for the area and supplies water for Westminster, Thornton, and Northglenn. In 1996, a reservoir was constructed upstream from Standley Lake to capture Woman Creek runoff and divert it away from Standley Lake.

Several series of detention ponds were constructed to manage plant wastes and surface-water runoff, including the A- and B-series Ponds along North and South Walnut Creeks and the C-series Ponds along Woman Creek. Past discharge of low-level contaminated wastes to both the A- and B-series Ponds resulted in the accumulation of plutonium and americium in pond sediments. Shallow groundwater refers to water within the alluvium and weathered bedrock geologic units and is found to a depth of 30 meters. Surface water and shallow groundwater are inextricably linked. Water from stream channels infiltrates downward, recharging the shallow groundwater, and the shallow groundwater recharges the stream and channels, depending on the time of year.

Beneath the alluvium is a highly impermeable bedrock layer that inhibits vertical flow. As a result, shallow groundwater flows laterally where it either discharges as baseflow into the streams or as hillslope springs and seeps. Approximately 200 to 300 meters below the surface lies the Fox Hills Sandstone, where the deep regional groundwater flows. Because of the intervening bedrock, this regional groundwater aquifer is hydraulically isolated from the Rocky Flats surface water and shallow groundwater and actinide contaminants.

The climate is temperate and semiarid, characteristic of Colorado's Front Range. The average annual precipitation is approximately 14.5 inches, with about half occurring as rain and half as snow, and falls primarily as snow from late October through early April and as

A deer grazes at the edge of the B-4 Pond. One of the goals of remediation efforts at Rocky Flats was to maintain the wetlands and wildlife habitat. Special care had to be taken not to disturb the habitat of the "threatened" Preble's meadow jumping mouse (inset), a short-grass prairie species that lives near the creeks and streams in the buffer zone around the Site.

rain during the remaining months. Evapotranspiration averages more than 15.8 inches per year, creating a water deficit in most years. Prior to the removal of the Industrial Area, much of the runoff feeding the Site's drainages occurred rapidly, originating from the mainly impervious Industrial Area surfaces.

Winds at RFETS are predominantly from the northwest toward the southeast. This wind pattern reflects the influence of local terrain combined with prevailing winds from west to east. The RFETS is noted for the periodic occurrence of strong, gusty winds (more than 100 miles per hour) that were known to shatter the windshields of vehicles parked on the Site. These winds were and are an important factor in the resuspension of soil and actinides. Indeed, air monitoring and subsequent calculations of the actinide loads showed that air transport was a dominant actinide migration pathway.

The Rocky Flats Site map showing the industrial area, and the Walnut Creek and Woman Creek drainage basins. The inset shows the location of the Great Western Reservoir and Standley Lake in relation to the Industrial Area.

Rocky Flats CLEANUP

The nature of actinide contaminants

Production operations conducted at Rocky Flats generated a huge volume of wastes contaminated with radionuclides and other hazardous substances. The majority of wastes were shipped off-site for disposal, but many areas at Rocky Flats had contaminated soil and water due to the improper disposal of contaminated materials, ruptured or leaking pipes, fires and explosions, or faulty storage units. Because many of the activities at the plant involved plutonium, much of the contamination was associated with this element. Based on historical data, there were five plutonium accidents and incidents that resulted in the majority of accidental releases of plutonium to the environment. The majority of these drew considerable public concern and media attention.

These releases included a plutonium worker's worst nightmare: major plutonium glovebox fires in 1957, 1965, and 1969. The 1969 incident occurred when spontaneous combustion of plutonium initiated a fire in Building 776. The fire was drawn through the glovebox line by the glovebox ventilation systems, spreading through several hundred interconnected gloveboxes. Had these fires not been contained within the buildings, they would have threatened contamination of a broad area surrounding Rocky Flats. These incidents brought significant media attention to the Site.

By far the largest source of actinide release to the environment came from improper storage (1958 to 1967) in an open field of about 5,240 55- and 30-gallon drums containing spent machine cutting oil, which was contaminated with plutonium and uranium. It was discovered as early as 1959 that some of these drums were leaking, but the drums were not removed until 1967. Based on material balance around the drums, it is estimated that 5,000 gallons containing approximately 86 grams (5.3 curies) of plutonium were released into the soil. In 1969 some of the contaminated soil was removed and the remaining soil covered with an asphalt cap known as the 903 Pad. During this initial remediation it is believed that winds distributed substantial amounts of contamination to the east and southeast.

Between 1991 and 1999, nearly 2,500 surface-soil samples were collected and analyzed across the Site to assess the level and extent of actinide contamination. A variety of geostatistical analysis tools were used, including variograms (a tool that describes the spatial or the temporal correlation of observations) and kriging (a regression technique to approximate or interpolate data that apply known values in one sample to produce an unbiased estimate of values in another). These are commonly used approaches when small-scale concentration distribution is heterogeneous and sample data exist in a large spatial area, such as at RFETS.

Estimated spatial concentrations of plutonium-239 and -240 in surface soil at RFETS were calculated by kriging analysis. Plutonium and americium generally exhibit the same spatial distribution in surface soils, with wide variations in

In 1969 spontaneous combustion in a glovebox in Building 776 started a fire that spread through the filter plenum.

It is estimated that 5,000 gallons of oil containing 86 grams of plutonium leaked into the soil from these drums improperly stored in an open field. After the drums and some of the contaminated soil were removed, the area was covered with an asphalt cap known as the 903 Pad.

To keep down dust during demolition, water was sprayed on Building 771, dubbed by the media as "the most contaminated building in America." When demolition of the building was begun in July 2004, Secretary of Energy Spencer Abraham issued a press release that read in part: "This represents a historic milestone in closing Rocky Flats and the most significant cleanup accomplishment to date in the DOE Complex. The demolition of one of the most contaminated buildings in the country, once thought impossible, demonstrates the nation's commitment to accelerated cleanup and closure of Rocky Flats."

Rocky Flats CLEANUP

activities occurring throughout the Site. The highest concentrations were found at the 903 Pad and areas to the east of the pad, and display a wind-driven dispersal pattern to the east of the primary source area (the 903 Pad). The plutonium and americium radioactivity in RFETS soils is highly heterogeneous, often consisting of "hot particles." Approximately 90 percent of the plutonium and americium inventory was in the top 12 centimeters of the soil.

In addition to contamination produced during manufacturing operations at RFETS, the closure project was required to deal with contamination that resulted from remediation and decontamination and decommissioning activities. Concentrations of plutonium and americium ranged from fractions of picocuries in streams and pond waters, to nanocuries in soils and sediments, to curies in contaminated building materials.

Because it was not feasible to sample surface soil at every location, a geostatistical modeling technique known as kriging was applied to the plutonium surface-soil data for plutonium-239 and -240 to estimate concentrations in surface soil. The "hot spot" of plutonium-239 and -240 concentrations in excess of 1,000 pCi/g at the 903 Pad is shown in red. A clear plume of plutonium-239 and -240 contamination that tracks roughly with the prevailing winds from northwest to southeast is evident from the data. This figure represents conditions at Rocky Flats prior to soil remediation.

Plutonium-239/-240 Activity in Surface Soils
Kriging Analysis (pCi/g)

- Result < = 0.01
- 0.01 < Result < = 0.05
- 0.05 < Result < = 0.1
- 0.1 < Result < = 0.5
- 0.5 < Result < = 1.0
- 1.0 < Result < = 5.0
- 5.0 < Result < = 10.0
- 10.0 < Result < = 100.0
- 100.0 < Result < = 1000.0
- Result > 1000.0
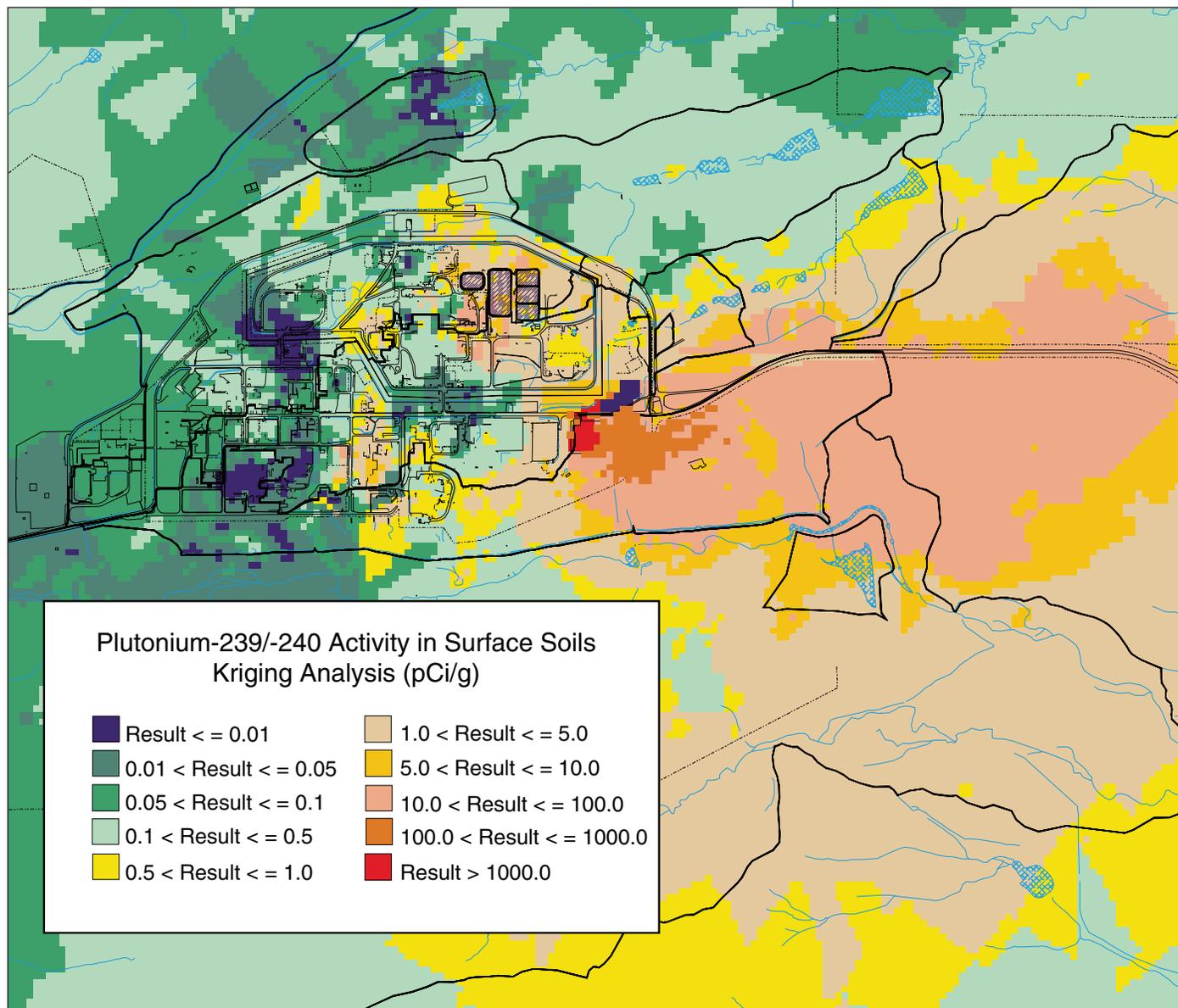
Rocky Flats CLEANUP

Steven Conradson of Los Alamos inspects the low-temperature cryostat and sample positioner at the Stanford Synchrotron Radiation Laboratory. Researchers used X-ray absorption spectroscopy to gather information about the chemical form of plutonium in RFETS soils and concretes.

A comparison of synchrotron-based XANES spectra for plutonium in environmental samples collected from 903 Pad soil and smoke exposed concrete from building fires. The spectra labeled Pu(III), Pu(IV), Pu(V), and Pu(VI) are oxidation-state standards. The spectra labeled RFETS soil and RFETS concrete are environmental samples. From the XANES it is clear that the RFETS environmental samples are nearly identical to the Pu(IV) standard. The inset shows the second derivative, which is another way of comparing and analyzing XANES spectra, and is commonly used to determine the energy of the absorption edge.

THE CASE FOR PARTICLE TRANSPORT

Expectations based on actinide chemistry

Migration of actinides in the environment takes place within the context of physical and chemical processes. Chemical reactions, particularly redox reactions within soil and ponds, were and are often hypothesized to explain actinide mobility. At one extreme, the actinide(s) may react chemically along with the surrounding materials, to create soluble (dissolved) and mobile components. At the other extreme, the actinide(s) might remain unchanged at the molecular and atomic scale while the associated materials react to create mobile and immobile components.

The great contrast between actinide solubilities—plutonium and americium have very low solubilities, whereas uranium has relatively higher solubility—drove consideration of colloidal and particulate transport processes and required careful evaluation of evidence that could distinguish solubility and redox process results. For example, actinide chemists have recognized the preferred stability of plutonium in the form of plutonium dioxide (PuO_2) and as colloidal-sized materials, but detailed knowledge of reactivity and mobility of such materials in the environment is limited at concentrations of picocuries per liter (pCi/L) in waters and picocuries per gram (pCi/g) to nanocuries per gram (nCi/g) in soils.

Extensive field observations and research have been conducted internationally on the environmental behavior of actinide elements in diverse sets of environments over the past 30 to 40 years. This research has provided a good base for understanding the major types of species and their transport mechanisms in soils and natural waters.

In natural waters plutonium solubility is limited by the formation of amorphous $Pu(OH)_4$ or polycrystalline PuO_2, which provides an upper limit on the amount of dissolved (i.e., ionic/molecular) plutonium that can be present. PuO_2 has a laboratory-measured solubility range of 10^{-10}-10^{-13} mole per liter (mol/L) and is limited by the formation of amorphous $Pu(OH)_4$.

Because of the very low solubility and the tendency of Pu(IV) compounds to adhere to organic and mineral particles, the primary path of plutonium transport is usually migration of colloidal particles. Indeed, when concentrations above fallout levels of plutonium have been investigated in detail, they have been linked to colloids and particulates. The problem with plutonium is that it is a very emotional issue with concerned citizen and stakeholder groups. There
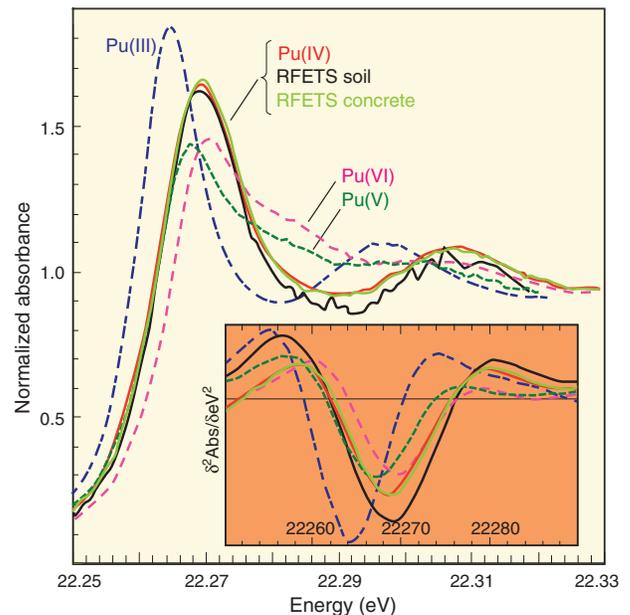
was a clear need to provide the stakeholders with a scientific understanding of actinide transport mechanisms to move forward with planning a strategy to close the Site.

Synchrotron radiation studies

Although it was suspected that plutonium contamination in the RFETS environment was in the oxide form, likely PuO_2, with very low solubility, definitive proof did not exist to verify its chemical form and oxidation state.

Los Alamos researchers, led by Steven Conradson, used X-ray absorption spectroscopy using the Stanford Synchrotron Radiation Laboratory (SSRL) to extract information about the chemical form (or speciation) of plutonium in RFETS soils and concretes. This technique is not well suited for the extremely dilute samples that were typical of the RFETS environment but were successfully applied by selective use of higher-concentration samples, autoradiographic identification of plutonium-containing sample components, and careful tuning of the spectroscopy data-collection systems at SSRL.

X-ray absorption near edge structure (XANES) analysis was used to determine that the oxidation state of plutonium in soils and concretes was Pu(IV). Comparison of XANES spectra of plutonium standards in oxidation states III, IV, V, and VI with spectra obtained from 903 Pad soil and smoke-exposed concrete from buildings 707, 771, and 776 indicated that the spectral features exhibited by the 903 Pad soils and the concrete are nearly identical in energy, intensity, and structure to the spectral features of the Pu(IV) standard.

Further analysis using extended X-ray absorption fine structure (EXAFS) spectroscopy was employed to unambiguously determine the chemical form of plutonium to be the relatively insoluble hydrous oxide, $PuO_2 \cdot xH_2O$. Similar studies were employed on soil and contaminated concrete samples from around the site. The EXAFS data can be fit to extract the number of near-neighbor atoms at different bond distances to determine information about the local chemical structure of plutonium in the sample.

The Fourier transform of the EXAFS data gives a distinct peak for each interatomic distance in the sample. For PuO_2, the Fourier transform and fit both show peaks for shells of Pu-O at 2.33 angstroms (Å), and Pu-Pu at 3.86 Å. This Fourier transform is unique for PuO_2. The EXAFS Fourier transforms of the RFETS soils and concrete samples are nearly identical with that found for PuO_2, with one small exception. In addition to interatomic distances of 2.33 and 3.86 Å, the RFETS soil and concrete data show some small peaks at intermediate distances between 2.3 and 3.0 Å, which are consistent with additional Pu-OH or Pu-OH_2 interactions that would be expected for hydrated PuO_2 exposed to water in the environment. These data were verified experimentally by examining the EXAFS of a number of laboratory-prepared samples of hydrated $PuO_2 \cdot xH_2O$.

One final question of interest to the scientific and local communities was whether the observed EXAFS could be consistent with the recently reported PuO_{2+x}. The EXAFS of $PuO_{2.2}$ was clearly different from that of PuO_2 and hydrated PuO_2. In particular, the $PuO_{2.2}$ shows an additional Pu-O peak at the very short distance of 1.84 Å that is not present in PuO_2 or the RFETS samples. These X-ray absorption spectroscopy (XAS) studies demonstrated that the largest source of plutonium in the environment at RFETS was in an oxide form.
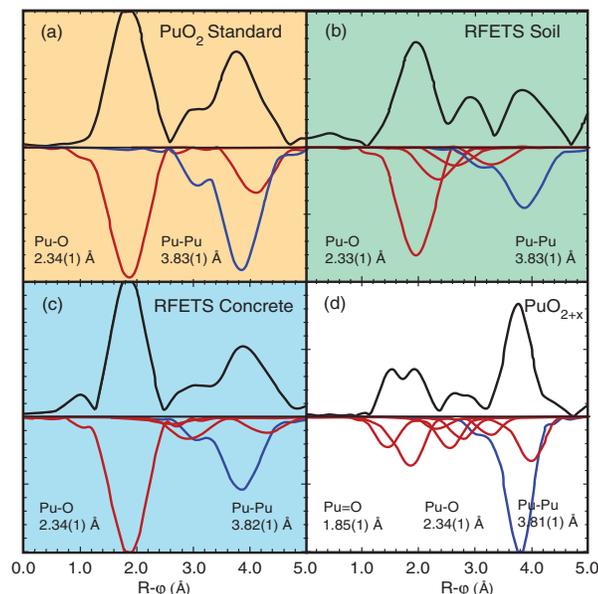
Fourier transform (positive amplitude) and fit (negative amplitude) of synchrotron-based EXAFS data for plutonium in (a) laboratory-produced PuO_2, (b) contaminated soil, (c) contaminated concrete, and (d) the recently reported PuO_{2+x}. The components of the fit, shown beneath the spectrum with negative amplitudes, correspond to individual shells of atoms. In the spectrum of PuO_2 (a), the first peak (red) corresponds to a shell of eight near-neighbor oxygen atoms at 2.34 Å, followed by a second shell of 12 plutonium atoms at 3.83 Å, and a third shell of 24 oxygen atoms (red) at 4.47 Å. Similar features are found in the first of the corresponding RFETS soil, RFETS concrete, and PuO_{2+x} samples.

Rocky Flats CLEANUP

(a)
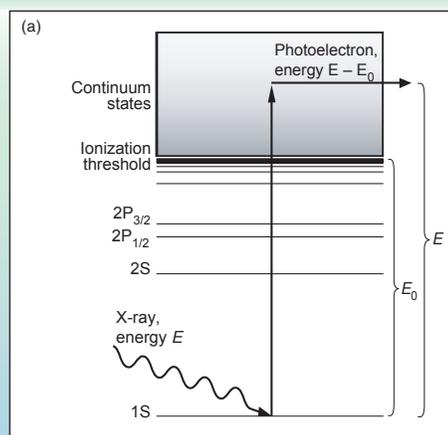
A schematic energy-level diagram of an absorbing atom shows the atom's first few core-electron orbitals: $1S$, $2S$, $2P_{1/2}$, $2P_{3/2}$. The ionization potential energy is denoted by E_0. With the absorption of a photon of energy E, the electron undergoes a transition to an unbound state in the continuum and, by conservation of energy, acquires a kinetic energy $(E-E_0)$.

(b)
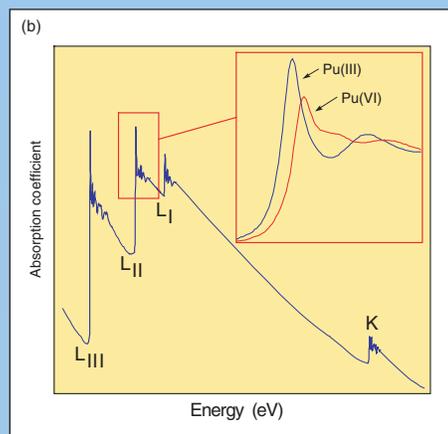
The abrupt jumps in this simplified absorption spectrum are the absorption edges, which correspond to the excitation of an electron from a specific orbital. The absorption edge due to excitation of the $1S$ electron is called the K edge, while excitations from the less strongly bound $2S$, $2P_{1/2}$, and $2P_{3/2}$ electrons are called the L_P L_{IP} and L_{III} edges, respectively. The inset shows how variations in the near-edge structure within the first 20 to 30 eV of the edge change with oxidation state of the absorbing atom.

X-ray absorption spectroscopy

X-ray absorption spectroscopy (XAS) is one of the most powerful tools available for mapping local atomic structure. In this technique, a sample is probed with X-rays that are tuned to the energy of a core electron shell (1s, 2s, 2p, etc.) in the element of study. The number of absorbed X-rays is monitored as a function of energy.

A plot of the X-ray absorption as a function of energy shows a decrease in absorption with increasing energy; the presence of a sharp rise at certain energies referred to as absorption edges; and if taken with sufficient accuracy, small oscillations that are the result of the local environment's influence on the target element's basic absorption probability (a). From the spectrum, one can extract the oxidation state of the absorbing element, the distances between the absorber and its near-neighbor atoms, and the number and type of near-neighbor atoms—all parameters that determine local structure. By selecting different X-ray energies, one can obtain this information for any element in the sample.

In the example of the absorption spectrum in the X-ray region (b), the step-like rises occur where the X-ray energy has come into resonance with a core electron shell of one of the elements in the sample (plutonium in this example), exciting the electron into the continuum (a). Because of the shape of the spectral feature (b), the data are referred to as an absorption edge. For the most part the edges are widely separated, and the target element is selected simply by scanning over an appropriate energy range. Following the edge, the absorbance decreases monotonically with increasing X-ray energy as the X-ray penetration depth becomes larger.

Fine structure is observed when the spectrum is expanded past a specific edge (b). The X-ray absorption near-edge structure (XANES) region occurs as peaks and shoulders over a 20-to-30-eV-wide region immediately past the edge onset. The fine structure on the high-energy side of the edge that damps out over several hundred eV is termed extended X-ray absorption fine structure (EXAFS) (c). This fine structure in both the XANES and the EXAFS regions is well understood and enables XAS to be applied to the determination of chemical speciation and local structure.

X-ray absorption near-edge spectroscopy

The X-ray absorption near-edge structure (XANES) can be used to determine the oxidation state of the target (X-ray absorbing) element in solution or in the solid state. The energy at which an absorption edge appears depends on the ionization potential of the ion. This ionization potential increases with the ion's valence state, so in general the absorption shifts to higher energy with increasing oxidation state. This known effect has been observed in plutonium and can therefore be used to determine the oxidation of plutonium in soils and concrete samples from RFETS.

An example of the differences in XANES spectra of plutonium is shown in the inset to (b), where XANES of Pu(III) and Pu(VI) are compared. There are distinct differences in the energy of the rising absorption edge, the intensity of the peak (sometimes referred to as the "white line"), and the structure in the absorption features at the higher energies beyond the absorption peak. All of these differences in the XANES spectra are used to identify the oxidation state of plutonium in RFETS samples.

For plutonium studies, researchers prefer to use the L_{III} X-ray absorption edge (which appears near 18,060 eV) because it has the highest absorption intensity (c). However, in RFETS environmental samples there were other elements present in the samples that exhibited X-ray absorption in this same region, and therefore interfered with the plutonium measurements. As a result, the plutonium X-ray absorption studies of RFETS samples were performed instead at the L_{II} X-ray absorption edge, which appears near 22,270 eV. This absorption edge has a lower absorption intensity than the L_{III} edge, making the RFETS samples very hard to study using X-ray absorption spectroscopy.

Rocky Flats CLEANUP

Extended X-ray absorption fine structure

Beyond the edge region, fine structure occurs as a series of oscillations superimposed upon what would be the smooth absorbance of the isolated atom. The origin of this fine structure arises from constructive and destructive interference between the outgoing photoelectron wave and the portions of this wave backscattered off of neighboring atoms as illustrated in (d). The modulation of the interference condition with the change in X-ray energy results in oscillatory fine structure contributed by each neighboring atom and depends on the electron wavelength and the distance to the backscattering atoms.

The modulation in the extended X-ray absorption fine structure (EXAFS) is described by a single-scattering formulation that contains several structurally significant metrical parameters. These parameters (noninclusive) include the number of neighbor atoms of the same atomic number at the same distance from the absorbing atom (i.e., a shell of neighbor atoms), a Z-dependent, per-atom backscattering amplitude function for that shell, the pair-wise Debye-Waller factor, and a phase-shift characteristic of the particular absorber-scatterer pair.

Mathematically, the scattering contributions can be summed over all of the shells and result in the composite EXAFS spectrum. These metrical parameters are extracted from the data via nonlinear least-squares-curve fits. A model consisting of a number of neighboring shells of atoms (supplemented by multiple scattering paths when necessary) is first devised. The EXAFS is the sum of the individual waves from these shells, calculated from the single-scattering equation. The source of the requisite phases and amplitudes for the fitting has evolved from spectra of structurally analogous standard compounds to very accurate ab initio calculations for clusters of atoms that can be quite close to the final structure.

Phase shifts and amplitudes are unique to the different elements, with enough difference with increasing Z to allow the type of element to be identified to ± 3-4 in principle. The structural parameters, e.g., R, N, and sometimes σ, are allowed to float until the least-squares difference between the data and the fit are minimized. Additional chemical information such as relationships between various parameters or shells or a permissible range for a parameter are introduced as constraints.

The view of EXAFS as a superposition of sine waves dictates Fourier analysis, converting from $\chi(k)$ to $\chi(R)$ by Fourier transformation (e). Since this converts each wave into a peak, $\chi(R)$ is related to the population-weighted average radial structure function around the absorber and the modulus does suggest a pair distribution around the absorber. For this reason, the Fourier transform representation ($\chi(R)$), and usually just the modulus, is most often used in figures despite most of the analysis actually occurring in k space ($\chi(k)$). The data are frequently shown as weighted spectra ($\chi^3(k)$) to (over)emphasize scattering contributions at longer distances.
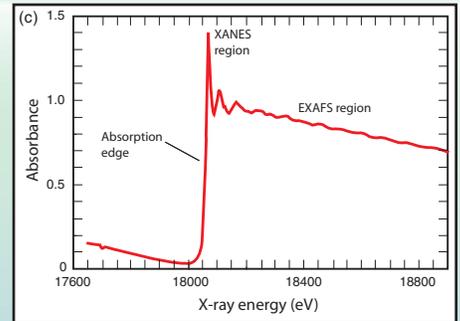
An X-ray absorption spectrum over a 1,000-eV region shows the rising absorption edge, the X-ray absorption near-edge structure (XANES) that occurs within the first 20 to 30 eV of the edge, and the EXAFS oscillations that occur past the edge.
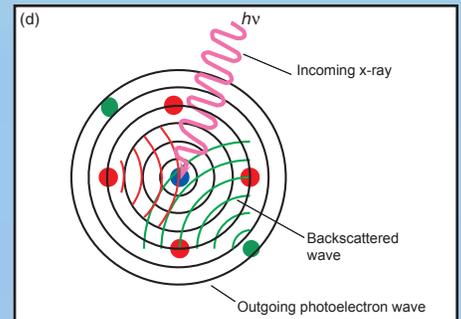
When X-ray energy is absorbed by a central atom (blue), a photoelectron wave is propagated outward. This outgoing wave is backscattered off neighboring atoms (red and green) and interferes with itself at the origin. Because of the ratio of the absorber-neighbor distance to the photoelectron wavelength, it is interfering constructively with respect to the first shell (red) and destructively with the second shell (green).

Right: EXAFS Fourier transform (black line) and fit (blue line) of the first three shells of PuO₂. The individual shells that make up the fit are shown with negative amplitudes and correspond to eight oxygen atoms at 2.34 Å, 10 plutonium atoms at 3.83 Å, and 23 oxygen atoms at 4.53 Å. The experimental data can be compared with expectations based on the crystal structure shown in (f).

Far right: The cubic crystal structure of PuO₂ can be viewed from the perspective of the central plutonium atom (black) to understand the results of EXAFS studies. The central plutonium atom is surrounded by a shell of eight near-neighbor oxygen atoms (red) with a Pu-O distance of 2.33 Å. Next comes a shell of 12 neighboring plutonium atoms (grey) at 3.81 Å, followed by a shell of 24 oxygen atoms (pink) at 4.47 Å. These three shells of atoms correspond to the three shells shown in the EXAFS Fourier transform spectrum (e).
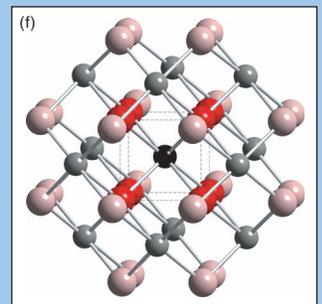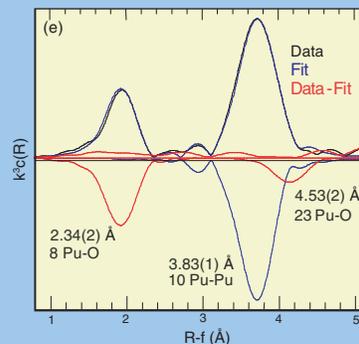
Rocky Flats CLEANUP
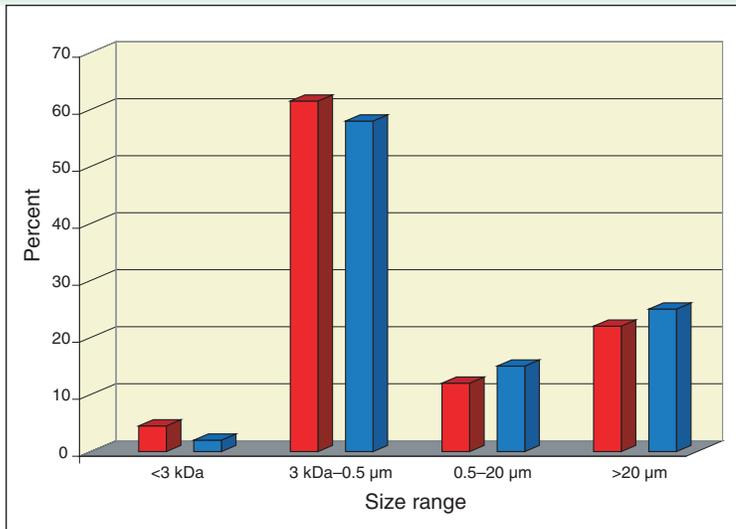
This data plot of tangential-flow ultrafiltration results is from field studies and experiments by Peter Santschi and coworkers at Texas A&M University, Galveston. Ultrafiltration demonstrated that the majority of material (more than 90 percent in this example) that passed a 0.5-μm filter was not dissolved, but colloidal, as it could be filtered out using smaller, 3-kDa ultrafilters. The blue bars represent normal discharge and the red bars represent data from a storm event, recorded in April and May 2000, respectively.

Ultrafiltration of field samples

Based on the geochemical characteristics discussed above, one might conclude that insoluble oxides of plutonium and americium would be trapped by the geological matrix and remain immobile in the environment. However, there have been a growing number of field studies documenting movement of low concentrations of low-solubility radionuclides in surface water and groundwater.

Small concentrations of low-solubility radionuclides such as plutonium and americium can be transported in surface water or groundwater as small-sized particulates, which range from 1 nanometer (nm) or one-billionth of a meter to 1 micrometer (μm) or one-millionth of a meter. These small-sized particles remain suspended and therefore transported in natural aquatic systems. At RFETS, the monitoring and historical observations were that plutonium and americium could migrate in small quantities that would settle out in the A-, B-, and C-series Ponds. Thus colloids were likely to play a role in plutonium and americium mobility at RFETS.

From 1998 until 2001, Peter Santschi and coworkers of Texas A&M University, Galveston, used field studies and laboratory experiments to examine actinide (plutonium-239 and -240 and americium-241) concentrations and phase speciation in the surface environment at RFETS. Since the environmental forms of actinides present in the surface waters were in the 10^{-3}-10^{-1} pCi/L concentration range, filtration, and tangential-flow ultrafiltration were the only methods suitable for the separation and analysis.

Measurements of total plutonium-239 and -240 and americium-241 concentrations in storm runoff and pond discharge samples, collected during spring and summer from 1998 to 2000, demonstrated that most of the plutonium-239 and -240 and americium-241 transported from contaminated soils to streams occurred in the particulate (≥0.45 μm) and colloidal (approximately 2 nm or 3 kilodaltons [kDa] to 0.45 μm) phases. In general, most of the plutonium and americium in RFETS water was found in the particulate phase (>20 μm and 0.5-20 μm) with most of the material that passed a 0.5-μm filter being colloidal, i.e., it was filtered out by 100- or 3-kDa ultrafilters, with 20 percent or less of 0.5-μm filter-passing plutonium and americium passing a 3-kDa ultrafilter. Based on graphite furnace atomic absorption spectroscopy, transmission electron microscope, and energy-dispersive X-ray microprobe images, colloids in these size ranges were primarily composed of clay and organic-matter.

Plutonium concentrations above the action level (≥0.15 pCi/L) at RFETS historically occurred predominantly during storm runoffs in early spring and summer at monitoring locations either within or adjacent to the Industrial Area. During these events, elevated concentrations of colloidal plutonium, accompanying those in the particle phase, were observed. Controlled laboratory investigations of soil resuspension, which simulated storm and erosion events, confirmed that most of the plutonium in the 0.45-μm filter-passing phase was in the colloidal phase (≥80 percent) and that remobilization of colloid-bound plutonium during soil-erosion events can be greatly enhanced by humic and fulvic acids present in these soils.

However, the fraction of remobilized colloidal plutonium, at most 1 percent in these soil-resuspension experiments, was much smaller than what was observed during storm runoff or pond discharge, where the fraction of colloidal plutonium passing a 0.5-μm

filter was as much as 66 percent. This showed that suspended matter and colloids were likely derived less from eroding soils directly but more likely from sediment resuspension of streambeds and banks.

Isoelectric focusing experiments of radiolabeled colloidal matter extracted from RFETS soils revealed that colloidal plutonium is in the tetravalent state and was mostly associated with a negatively charged organic colloid with molecular weight of 10 to 15 kDa, rather than with the more abundant inorganic (iron oxide and clay) colloids. Santschi's evidence strongly argues against the presence of mobile colloidal microparticles consisting mainly of PuO_2 but suggests that it is imbedded in, or attached to, a matrix of organic matter and some iron and with organic matter determining the surface charge and migration behavior of the 10-to-15-kDa colloid.

These studies were of great importance in that they provided evidence that the low levels of plutonium and americium in surface water at RFETS are transported by the colloidal and particulate fraction and not the dissolved fraction of the water. These findings were essential for the understanding of actinide transport pathways, the confirmation that groundwater is not a major pathway for plutonium and americium transport, and the development of actinide transport models.

Tangential-flow ultrafiltration

In tangential-flow ultrafiltration (also called cross-flow ultra-filtration), a fluid is pumped tangentially along the surface of a membrane. An applied pressure drives a portion of the fluid through the membrane (permeate) to the filtrate side. Particulates and macromolecules that are too large to pass through the membrane pores are retained on the upstream side (retentate).

The tangential flow of sample solution across the membrane surface helps slow the buildup of molecules and particles that can clog the pores or form a cake layer that reduces the permeate flow, i.e., "fouling" the membrane. This feature of tangential-flow ultrafiltration makes it an ideal process for fine-particle separations.

In ultrafiltration, a broad range of membranes with different size cutoff ranges can be employed to affect size separation, ranging from approximately 1 micrometer (µm) to 0.003 µm or approximately 1,000 kilodaltons (kDa) to 1 kDa on a molecular-weight scale. The membrane systems can be fabricated in a variety of mechanical configurations. Flat-sheet membranes can be used in plate-and-frame or spiral-wound configurations. Hollow fibers of membrane material packed into a module are another common equipment design. Polymers are the most common porous materials used for ultrafiltration membranes, but some inorganic materials such as alumina are also used.
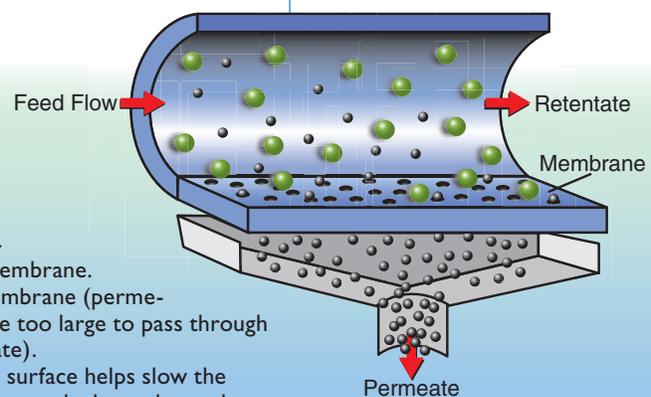
In tangential-flow ultrafiltration, a sample solution is pumped tangentially along the surface of a membrane. An applied pressure serves to force a portion of the fluid through the membrane (permeate) while the remainder (retentate) is recirculated back to the feed reservoir. The cross-flow prevents buildup of molecules at the surface that can cause fouling. A membrane-pore-size rating, typically given as a micron value, indicates that particles larger than the rating will be retained by the membrane, making it an ideal process for fine-sized-based separations.

Rocky Flats CLEANUP

Plutonium and americium transport modeling

With the enhanced understanding of plutonium and americium particle transport processes and exclusion of soluble transport, it was clear that contaminant transport models based on soluble forms of plutonium were not applicable or defensible. Unfortunately, before the formation of the AME, risk assessment was based on the assumption of soluble species transport. Therefore, alternate contaminant transport modeling approaches were sought that focused on spatially distributed contaminants in soils and sediments. Part of the capability required was the ability to predict contaminant transport under existing conditions and for a range of possible future site remediation and management scenarios.

The state-of-the-art model selected for simulating hillslope erosion processes at the RFETS was the Water Erosion Prediction Project (WEPP) model. It is a process-oriented computer model that incorporates improvements in erosion-prediction technology based on erosion mechanics, soil physics, plant science, hydrology, infiltration theory, and stochastic weather generation. The WEPP model estimates the spatial and temporal distributions of soil erosion and sediment deposition from overland flow on hillslopes, and the erosion, sediment transport, and deposition in small channels and impoundments. It also accounts for enrichment of transported sediment in finer (or smaller) particles and is thus well suited for contaminant transport calculations.

The WEPP erosion model was calibrated using data from large rainfall simulations on RFETS soil plots that measured runoff and sediment yield during simulated storm events. The frequency of storm events was derived from the weather database at Fort Collins, Colo., which spans more than 100 years. A range of storm-event sizes was then simulated using the WEPP model, which provided the estimated runoff hydrograph

Plutonium mobility map of a 100-year, 6-hour storm event for the 903 Pad and lip area watershed. Red indicates areas with the highest plutonium mobility; blue indicates areas with the lowest plutonium mobility. The models show that up to 99 percent of plutonium input to surface water in the channel (South Interceptor Ditch) is from hillslope erosion. After the water, sediment, and plutonium are delivered to the channel, the HEC-6T model was used to route water, sediment, and plutonium downstream. The arrow indicates the general direction of sediment transport toward the South Interceptor Ditch. Model results are based on conditions prior to the completion of remediation activities.

903 Pad

South Interceptor Ditch
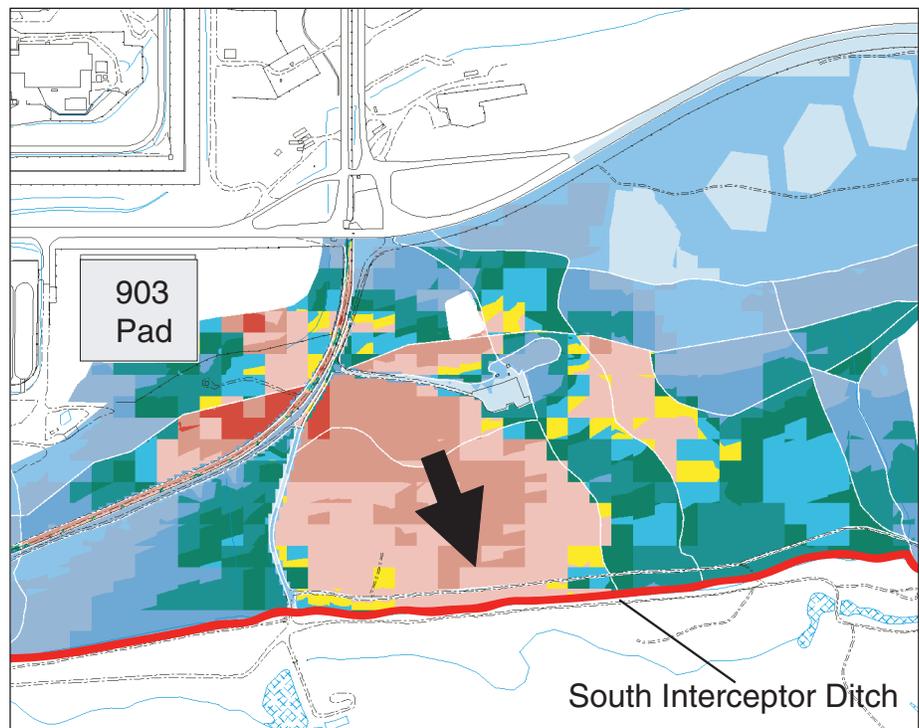
for specific hillslope areas and also provided an estimated mass of soil eroded, distinguished by particle size.

To estimate stream-channel sediment erosion and deposition, output from the WEPP model was routed into a modification of the U.S. Army's Hydrologic Engineering Center (HEC) sediment transport model HEC-6T. The HEC-6T model allows for up to 100 tributary inflows to the main channel, which was crucial for modeling the RFETS watersheds. Model output from WEPP and HEC-6T were combined with soil actinide data (either plutonium or americium), using Geographic Information Systems software, to develop predictions of surface-water actinide concentrations in the watershed.

Under the drastically reconfigured site conditions (with buildings and pavement removed), historical monitoring data are limited in their usefulness to predict future runoff, sediment, and contaminant yields. Therefore, RFETS adopted a methodology of using historical data to calibrate simulation models and then using the simulation models to predict future runoff, sediment, and contaminant yields. The models were also used to design and evaluate temporary and smaller-scale erosion control and remediation actions because they can predict the consequences of changing land management or configuration.

As part of the erosion-modeling process, the predicted soil erosion (mass eroded/unit area) was coupled with soil actinide concentration data to generate a map of predicted actinide mobility for a specific storm event. Actinide mobility maps and model results of plutonium and americium concentrations in surface water provided improved understanding of plutonium and americium mobility as a result of surface-water erosion processes.

The model results made it apparent that the largest plutonium and americium loads delivered to surface water do not necessarily originate from areas with the highest concentrations of plutonium and americium 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 plutonium and americium in the soil of any portion of the Lip Area. However, this area is relatively flat, with slopes of approximately 1 percent. 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 plutonium and americium transport. These modeling tools were used to evaluate alternatives considered for the 903 Lip Area remediation, and general future land configuration scenarios for the Site.

The drastically reconfigured site conditions shown in these aerial photos taken in 1995 (top) and after buildings and pavement were removed 10 years later (bottom) limited the usefulness of historical monitoring data to predict future runoff, sediment, and contaminant yields. To compensate for this, RFETS used historical data to calibrate simulations models and then used to models to predict actinide mobility

Rocky Flats CLEANUP

IMPACT OF SCIENTIFIC UNDERSTANDING ON SITE OPERATIONS

The reconfigured Walnut Creek basin functional channel shows the heavily armored channel and extensive erosion-control measures on stream banks. The measures included straw bales, wattles, and crimping; silt fences; mats; hydro-mulch and Flexterra™; and rip-rap (rock) lining of drainage channels. The inset shows a remedi-ated area of the 903 Pad covered with coconut matting. The biodegradable matting prevents wind and water erosion and allows native vegetation time to take hold.

This temporary water-detention pond was constructed to collect and store dust-suppression water used during the decontamination and decommissioning of Building 371. The pond was used to ensure that plutonium and americium contaminants did not enter the streambeds dur-ing remediation of the Site. This is an example of the extraordinary efforts employed by Site personnel to control erosion—and hence actinide transport—during decontamination and decom-missioning operations at the Site.

The scientific understanding developed through the integrated studies provided clarity and focus on the real issues surrounding plutonium and americium migration in the RFETS environment. Once Kaiser-Hill, DOE, EPA, Colorado, and the concerned citizens' groups reached an understanding of the technical issues surrounding plutonium and americium migration at the Site, then these groups were able to reach long-sought-after agreements on how to proceed with cleanup.

The common understanding that plutonium and americium were predominantly in particulate and colloidal forms led to the recognition that environmental migration occurs through sedimentation and resuspension of small particles by action of wind and surface water at the Site. This knowledge helped all parties focus remediation efforts on surface contamination and wind and surface-water transport pathways that posed the greatest risk to human health and the environment. It also helped guide selection of surface-specific removal technologies and future land-configuration strategies.

In recognition of the new understanding, Site operators responded with a major shift in emphasis to erosion and the need to control it. The most poignant illustration of this shift was a Management Directive (NRT-011-04) from Kaiser-Hill President Nancy Tuor, which discussed the importance of erosion control in all Site activities.

The recognized need for erosion controls "close in space and close in time" helped prevent movement of contaminants during Site remediation activities and reduced the transport of plutonium and americium to the Site's stream channels and ultimately off-site. The additional protection provided by soil-erosion control measures allowed Site remediation to proceed rapidly and thus meet or exceed the project deadlines.

In 1996, the Rocky Flats Cleanup Agreement radionuclide soil action level for plutonium cleanup was 651 pCi/g, and was based on dose. In 2002, armed with improved understanding of plutonium behavior, the DOE, Colorado Department of Public Health, and EPA released reports that formed the basis for a new surface soil-action level of 50 pCi/g that was based on risk and resulted from unprecedented community involvement.

Because plutonium contamination was generally confined to surface soils, the greatest risk to public health was from dispersal due to action of wind and surface-water erosion processes. This new risk-based agreement focused on removal of surface-soil contamination at a

more aggressive 50 pCi/g standard to three feet below the surface, with the tradeoff that contaminated soil below three feet could remain in place at higher concentrations.

The Site developed a Storm Water Pollution Prevention Plan that encouraged minimum soil disturbance, which resulted in control and minimization of erosion and sedimentation and minimization of runoff across the Site. Each project was reviewed for impacts to surface water with a specifically designed control system. Erosion-control measures included straw bales, wattles, and crimping; silt fences; mats; hydromulch and Flexterra™; and rip-rap (rock) lining of drainage channels. In addition, some new wetland areas were created. Several of these methods have expected useful lifetimes of a few months to a few years, and will require regular maintenance until the location is stabilized and vegetation well established.

In actual decontamination, demolition, and remediation work, the Site employed a combination of tents, comprehensive dust- and erosion-control measures, and general environmental protection during cleanup activities. As a result, surface-water and air monitoring stations at the Site boundary showed little change in actinide migration as a result of the Site cleanup activities.

Conclusions

Superfund sites such as RFETS are environmental problems of national significance. As such, we in the scientific community hope that our best science is brought to bear on decision making to improve its technical basis and to make it more transparent, repeatable, and thus, scientifically defensible. This came about at RFETS because of several factors, including the willingness of the integrating contractor to seek outside scientific advice and guidance; the acceptance through time, down to the project level, of the value of scientific advice in avoiding pitfalls and improving operations; and the gradual acceptance of the independence and veracity of the AME scientific advisors by the stakeholders. This willingness and acceptance allowed DOE, the integrating contractor, the regulators, and other stakeholders to focus on specified goals and objectives.

Making the case for particle-transport mechanisms, rather than aqueous sorption-desorption processes, as the cause of plutonium and americium mobility established a successful scientific basis for the dominance of physical transport processes by wind and water. The scientific basis was successful because it was in agreement with general theory on insolubility of PuO_2 in oxidation state IV; results of ultrafiltration analyses of field water and sediment samples; and XAFS analyses of soil, sediment, and concrete samples. It was also in general agreement with on-site monitoring data.

This understanding in turn allowed Site contractors to rapidly move to adopt soil-erosion and sediment-transport models as the means of predicting plutonium and americium transport, which led to design and application of sitewide soil erosion control technology to help control downstream concentrations of plutonium and americium in streamflow.

Finally, good scientific understanding in the public interest helped bring clarity and focus to real issues of actinide migration at RFETS. This in turn helped to develop a more defined scope with a clearer endpoint that allowed the most extensive cleanup in the history of Superfund legislation to finish one year ahead of schedule, ultimately resulting in billions of dollars in taxpayer savings and removing a $600-million-plus annual liability from the DOE budget forever.

Cleanup excavation of the 903 Pad took 13 months, during which approximately 32,000 tons of contaminated soil and asphalt were removed. Two 90-by-100-foot tents were erected over the site; sampling, excavation, waste loading, and backfill were conducted inside the tents to prevent wind dispersal of contaminants during remediation.

Rocky Flats CLEANUP

Two views of Rocky Flats' Central Avenue.

Rocky Flats National Wildlife Refuge

The Rocky Flats National Wildlife Refuge Act of 2001 began a process to finalize the reclamation of the land and secure its natural state into the future. In April 2005 a Comprehensive Conservation Plan was published in the Federal Register; it outlined habitat restoration plans for the next 15 years. The refuge is scheduled to partially open next year and be in full operation about 2012. It will have hiking trails, interpretive signs, and limited hunting. Approximately 1,000 acres in the center of the site will be maintained by DOE for long-term surveillance and maintenance while the remaining 5,200 acres will be transferred to the Department of the Interior.

The Xeric Tallgrass Prairie is a rare grassland type believed to be a remnant from the ice age and is perhaps the largest contiguous remnant of this grassland type in North America. This area is identified by species such as big bluestem, little bluestem, prairie dropseed, Indian grass, and switchgrass. Because of the prairie's proximity to the mountains, the species composition is influenced by mountain muhly, Porter's aster, blazing star, scurfpea, and Canada bluegrass.

The expansive prairie at Rocky Flats consists of grasslands, wetlands, and shrublands. The Xeric Tallgrass Prairie is a rare grassland type believed to be a remnant from the ice age and is perhaps the largest contiguous remnant of this grassland type in North America.

The Site:
- 6,266 acres within 50,000 acres of publicly owned open space.

Habitat zones:
- tallgrass prairie;
- mixed prairie grasslands (mixed-mesic grassland, xeric needle and thread grassland, reclaimed mixed grassland); and
- riparian corridors and wetlands (marsh, wet meadows, upland shrublands).

The wildlife:
- Preble's meadow jumping mouse, Black-tailed Jack Rabbit, Black-tailed Prairie Dog, Mule Deer, Whitetail Deer;
- Painted Turtle, Prairie Rattlesnake;
- Red-tailed Hawk, Northern Harrier, Peregrine Falcon, Western Meadowlark, Killdeer, Warbler; and
- Aragos Skipper (rare butterfly).

Acknowledgments

Additional reading

Actinide Migration Evaluation Pathway Analysis Summary, Kaiser-Hill, Rocky Flats Environmental Technology Site, Report ER-108, 2002.

S.D. Conradson, B.D. Begg, D.L. Clark, C. den Auwer, M. Ding, P.K. Dorhout, F.J. Espinosa-Faller, P.L. Gordon, R.G. Haire, N.J. Hess, R.F. Hess, D.W. Keogh, L.A. Morales, M.P. Neu, P. Paviet-Hartmann, W. Runde, C.D. Tait, D.K. Veirs, and P. M. Villella, *Journal of the American Chemical Society,* **126**(41), 13443-13458, 2004.

P.H. Santschi, K.A. Roberts, and L. Guo, *Environmental Science and Technology,* **36**(17), 3711-3719, 2002.

ARQ WINS TWO AWARDS

Actinide Research Quarterly has won two awards in a Society for Technical Communication (STC) Southwest Regional Competition. The magazine won an Award of Excellence for technical art and an Award of Merit for technical publications.

The awards were given to three consecutive issues considered as a whole: "Actinide Oxides" (2nd Quarter 2004), "Actinide Oxides II" (3rd Quarter 2004), and "10 Years" (1st Quarter 2005). Over the past several years ARQ has received five Awards of Excellence and two Awards of Merit from STC.

Illustrator/designer Susan Carlson and editor Meredith Coonley received the award for technical art. Contributing scientific advisor Mark Paffett, Carlson, and Coonley received the award for technical publications. ARQ's scientific advisors are David L. Clark and Gordon Jarvinen of the Seaborg Institute.

STC's 15,000 members make it the largest professional society in the world "dedicated to the advancement of the theory and practice of technical communications," according to its website.

Announcing publication of the third edition of

THE CHEMISTRY OF THE ACTINIDE AND TRANSACTINIDE ELEMENTS

A team of international experts, including a substantial number from Los Alamos, has coauthored the third edition of a classic text, *The Chemistry of the Actinide and Transactinide Elements*. Edited by Lester Morss, Norman Edelstein, and Jean Fuger, with Joseph Katz as honorary editor, the book will be released this summer by Springer Publishers. The five-volume set is critically acclaimed as the most authoritative and comprehensive compilation to date of the chemical properties of the actinide and transactinide elements and is anticipated to be the definitive work on actinides for the next 25 years.

The first edition of *The Chemistry of the Actinide Elements,* edited by Joseph Katz and Glenn Seaborg, was published in 1957 before the discovery of nobelium, lawrencium, and the transactinide elements. The second edition, edited by Katz, Seaborg, and Morss, was published in 1986 by Chapman and Hall, London and New York.

The third edition, containing 31 chapters, will include a contemporary and definitive compilation of the chemical properties of the elements from actinium (atomic number 89) to hassium (atomic number 108). Also included are authoritative review chapters on specialized topics such as thermodynamics, electronic theory, spectroscopy, magnetic properties organoactinide chemistry, coordination chemistry, solution chemistry separations science and technology, environmental science, analysis, and future-element predictions.

Los Alamos authors and the chapters to which they contributed include David L. Clark, Siegfried Hecker, Gordon Jarvinen, and Mary Neu (Plutonium); Wolfgang Runde (Americium); Robert Penneman and P. Gary Eller, both retired (Curium); David Hobart (Berkelium); P. Jeffrey Hay (Theoretical Studies of the Electronic Structure of Compounds of the Actinide Elements); A.J. Arko, retired, and John Joyce (The Metallic State of the Actinides); Carol Burns (Organoactinide Chemistry: Synthesis and Characterization and Organoactinide Chemistry: Reactivity in Catalytic Processes); and Jerry Stakebake (Handling, Storage, and Disposition of Uranium and Plutonium).

For the complete table of contents and author listing, see the web site at *http://www.springeronline.com*.

About the Quarterly

Actinide Research Quarterly
highlights recent achievements
and ongoing programs of the
Plutonium Manufacturing and
Technology (PMT) Division.
We welcome your suggestions
and contributions. ARQ can
be read online at

www.lanl.gov/arq
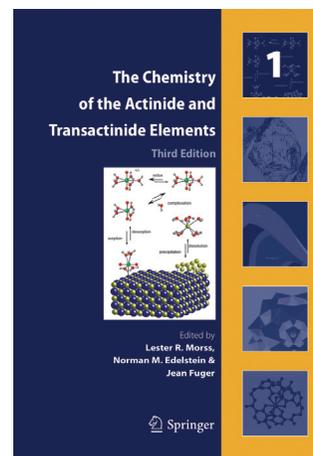
Address correspondence to
Actinide Research Quarterly
c/o Meredith Coonley
Mail Stop E500
Los Alamos National Laboratory
Los Alamos, NM 87545

If you have questions, comments, suggestions,
or contributions, please contact the
ARQ staff at arq@lanl.gov.

Phone (505) 667-0392
Fax (505) 665-7895

ARQ Staff