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## ACTINIDE MIGRATION EVALUATION PATHWAY ANALYSIS SUMMARY REPORT



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**'Actinides** are those **14 elements** with atomic numbers 90 to 103 that follow the element actinium in the **Periodic Table of Elements.'**

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# INTRODUCTION AND RFETS HISTORY

**THE ACTINIDE ELEMENTS**

90	91	92	93	94	95	96	97	98	99	100	101	102	103
Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
232.0	231.0	238.0	237.0	244.0	243.0	247.0	247.0	251.0	252.0	257.0	258.0	259.0	261.0

**INTRODUCTION** The Rocky Flats Environmental Technology Site (RFETS or Site), located near Denver, Colo., and owned by the United States Department of Energy (DOE), was formerly a manufacturing facility in the nation's Nuclear Weapons Complex. The Site is currently undergoing cleanup, closure and conversion to a National Wildlife Refuge. An important question was identified early in the closure planning – how do radioactive elements move in the environment?

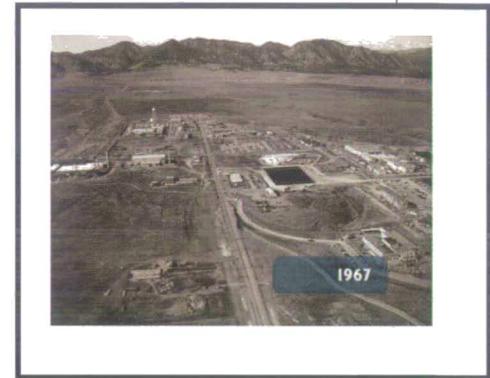
The Actinide Migration Evaluation (AME) Program was initiated in 1996 to address this question. Specifically, the AME focuses on issues of actinide behavior and

mobility in surface water, groundwater, air, soil and biota at RFETS. For the purposes of this study, an actinide refers to the radioactive element uranium (U), plutonium (Pu) or americium (Am).

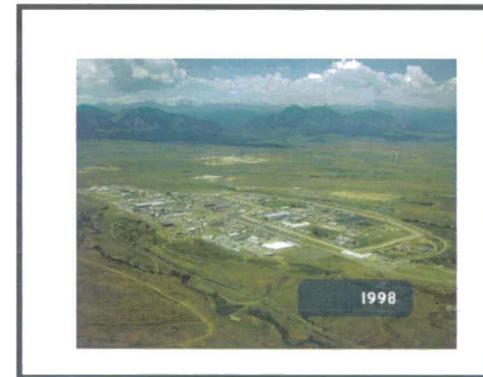
To address issues of actinide migration, the AME Program has brought together personnel with a broad range of relevant expertise in technical investigations, project management and external advisory roles. This effort, funded by DOE, involves identification of research investigations and approaches that can be used to solve short- and long-term issues related to actinide migration at the Site. Knowledge garnered through the AME Program is being used to characterize current RFETS environmental conditions and to recommend a path forward for long-term protection of surface water quality during closure and long-term stewardship of the Site.

## WHAT ARE ACTINIDES?

Actinides are those 14 elements with atomic numbers 90 to 103 that follow the element actinium in the Periodic Table of Elements. Actinides are among the heaviest known elements and all are radioactive. Only thorium and uranium can be found naturally in abundance. Plutonium and americium are man-made. Actinides of concern at RFETS addressed in this report are uranium (atomic number 92), plutonium (atomic number 94) and americium (atomic number 95).



In the early 1950s, Rocky Flats was built as part of the nation's Nuclear Weapons Complex. In 1989, following decades of expansion, production operations were halted. Current cleanup efforts are scheduled for completion by 2006. The Site will then become a National Wildlife Refuge.





**Effective cleanup of the Site requires a thorough understanding of how actinides move in the environment.**

Throughout the AME Program, there has been extensive public discussion and participation in the scientific process and review of findings. Discussion of actinide migration technical issues with stakeholders, regulators, administrators and staff has been valuable as a means of focusing efforts on critical questions.

Data presented in this Report show that air and surface water are the major transport pathways for all actinides. This is particularly true for plutonium and americium, which are largely insoluble and are transported when wind and water erosion move the soil and sediment particles to which the plutonium and americium are bound. Groundwater is a significant pathway for uranium, which is more soluble than plutonium or americium. The biological pathway is a minor transport mechanism for all actinides.

This Summary Report is a condensed review of the study's major topics and findings. Detailed discussions, calculations and literature references to support subjects discussed in this document are included in the companion Technical Appendix.

**PURPOSE** The purpose of the AME Pathway Analysis Report is to provide a summary of the quantitative analyses that have been performed to examine the many processes that impact movement of actinides in the environment at RFETS. Evaluation of alternatives for remediating actinide contamination at RFETS must consider migration and mobility along all available environmental pathways. The ultimate objective of the pathway study is to compare and quantitatively rank the various pathways in terms of total actinide loads transported off site for a given time period. Major transport pathways addressed in this study include: air, surface water, groundwater and biota.

This study is limited to quantifying actinide movement and does not assess actinide-related human health impacts. However, references to pertinent risk-based health standards are made to provide perspective.

**SITE HISTORY** RFETS is located 16 miles northwest of downtown Denver. It was built as a production plant to manufacture triggers for nuclear weapons and purify plutonium recovered from retired weapons. These operations involved fabricating components out of plutonium, enriched and depleted uranium, beryllium and stainless steel. Nearly 40 years of weapons production left a legacy of radiological waste at the Site, including contaminated facilities, process waste lines and buried wastes. Plutonium dispersal from fires in production buildings and leakage of waste oil stored outdoors caused contamination of the immediate environment.

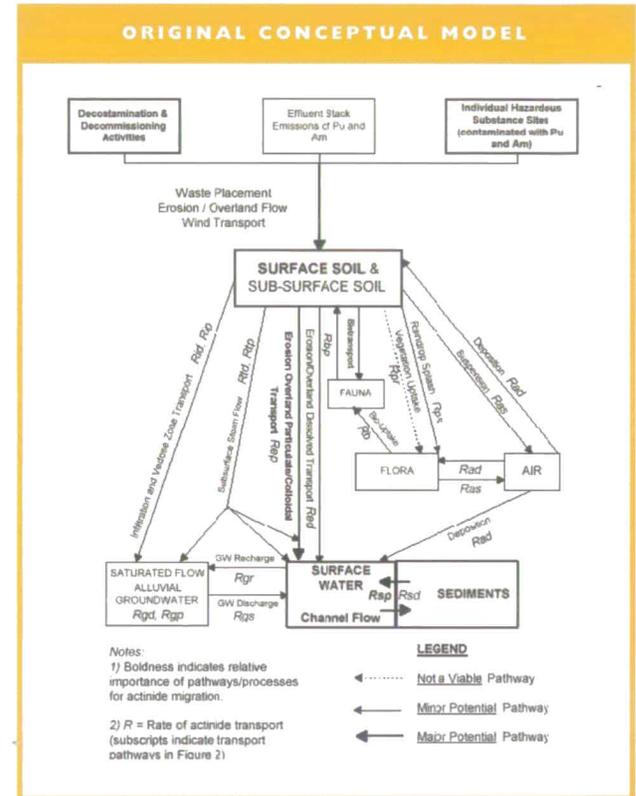
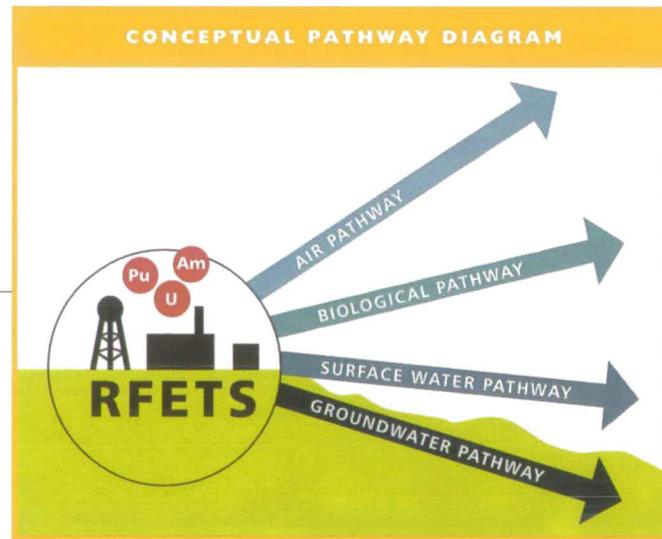
**CLOSURE AND CLEANUP** In 1992, the Site mission changed from production to one of closure and cleanup of the 385-acre Industrial Area and the surrounding 6,165-acre Buffer Zone. Today, RFETS is in the process of deactivating, decontaminating, decommissioning and demolishing all of the weapons production facilities and support buildings in the Industrial Area. The objective of the final closure phase is remediation of the environmental legacy of nuclear weapons production and transition to long-term stewardship as a National Wildlife Refuge.

# CONCEPTUAL MODEL

**CONCEPTUAL MODEL** In 1998, a document entitled "Conceptual Model for Actinide Migration Studies at the Rocky Flats Environmental Technology Site" was developed as an initial effort to provide a qualitative description of the relationships among potential actinide sources and transport pathways at RFETS (Kaiser-Hill, 1998).

The transport of actinide elements in the environment involves complex chemical and physical processes. These processes depend on the type and source of the actinide as well as the influence of the surrounding environmental media. To facilitate understanding of the potential routes for actinide transport in the RFETS environment, schematic models of actinide transport pathways were developed. One conceptual model was developed specifically for plutonium and americium, because they have similar geochemical and transport properties. A separate model was developed for uranium because of its different properties. These models formed the basis for quantitative analyses described in the Pathway Analysis Report. Development of the Pathway Analysis Report used both existing data from the literature and site-specific analyses. Field, laboratory and modeling studies were conducted to provide quantitative estimates of actinide migration.

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## INITIAL CONCEPTUAL MODEL DIAGRAM

This chart was the first effort by the AME group to diagram how plutonium and americium move in the environment at RFETS. It was a familiar tool at public meetings and has evolved into the chart on the following page.

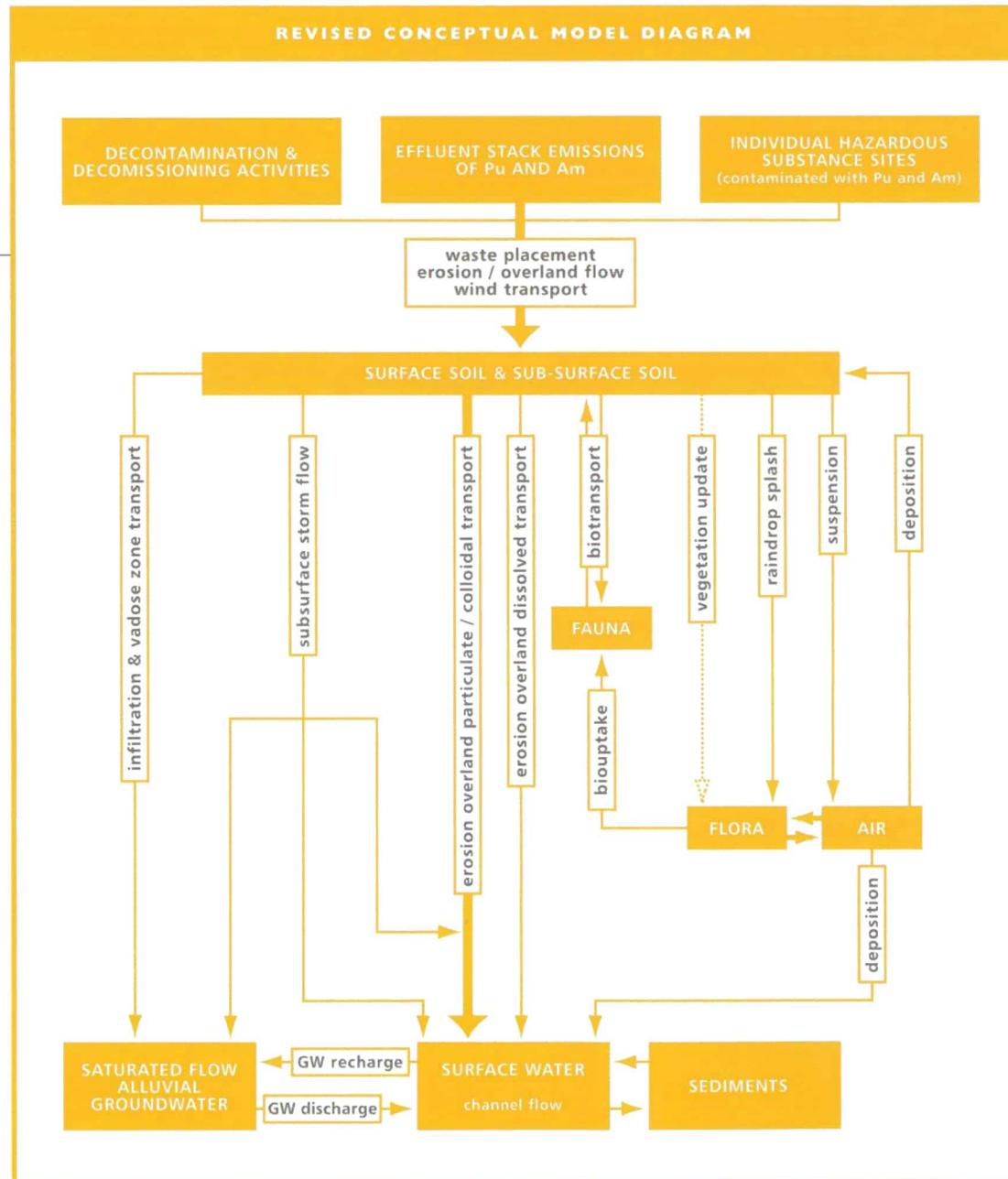
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**CONCEPTUAL MODEL LEGEND**

- not a viable pathway
- minor potential pathway
- major potential pathway

**ACTINIDE MIGRATION CONCEPTUAL MODEL**

This flowchart, developed from the conceptual model, is a qualitative diagram of potential plutonium and americium movement pathways at RFETS. The Pathway Analysis Report quantifies potential pathways to determine their relative importance in RFETS actinide migration. Since the geochemical behavior of uranium is different from that of plutonium and americium, a separate conceptual model flowchart developed for uranium is in the Technical Appendix.



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# ACTINIDE SOURCES



These drums leaked contaminated waste oil in the 1960s. The 903 Pad area is the Site's primary known source of plutonium and americium in the environment and is scheduled for cleanup in 2002.

## LEAKING DRUMS RELEASED CONTAMINATION

A major release of plutonium to the environment occurred when plutonium-contaminated waste oil leaked from approximately 3,750 drums stored outside from 1958 to 1968. Although the drums were removed after leakage was detected, plutonium-contaminated soil was dispersed into the air during remediation activities and deposited east of the drum storage area. In 1969, the area was covered with gravel fill and an asphalt layer to prevent further wind dispersal. The remaining contamination in this area, known as the 903 Pad, continues to be one of the major sources of plutonium and americium contamination at the Site. Further remediation will remove the source material and reduce airborne transport of plutonium and americium.

**ACTINIDES IN THE ENVIRONMENT** Actinide elements occur in the environment at RFETS as both "background" material and as material released during operations at the Site. For plutonium and americium, background concentrations exist because of global fallout from historic atmospheric nuclear testing.

With uranium, background quantities occur naturally in the soil and underlying geologic material. A significant amount of naturally occurring uranium exists at RFETS as well as in the surrounding vicinity, as evidenced by the presence of the Schwartzwalder uranium ore mine within 16 kilometers (10 miles) of the Site. Differentiation between natural and man-made uranium contributions can be accomplished by examining characteristic differences in the mixtures of uranium isotopes. Such isotopic analyses have detected low levels of man-made uranium in shallow groundwater at locations somewhat removed from contaminant sources. However, in general, beyond the immediate vicinity of man-made uranium sources, the observed uranium concentrations are difficult to distinguish from natural background uranium.

**SPATIAL DISTRIBUTION** Plutonium and americium generally exhibit the same spatial distribution in surface soils, with wide variations in activities occurring throughout the Site. The highest concentrations are found at the 903 Pad and areas to the east of the Pad. Nearly all the plutonium and americium in RFETS soils is confined to the top 20 centimeters (8 inches) of soil and approximately 90 percent is located in the top 12 centimeters (5 inches) (Webb, et al., 1993; Litaor, et al., 1994).

## BACKGROUND LEVELS OF ACTINIDES

**Plutonium and Americium – Global Fallout from Nuclear Tests** There were 541 acknowledged atmospheric nuclear tests conducted around the world, primarily from 1945 through 1963, prior to the Limited Test Ban Treaty. These tests resulted in the global dispersal of approximately 4,000 kilograms (360,000 curies) of plutonium and 95 kilograms of americium. Most of this fallout was distributed across the temperate regions of the Northern Hemisphere, resulting in background plutonium levels that generally range from approximately 0.003 to 0.03 picocuries per gram (pCi/gram) of surface soil. The background plutonium level found in Front Range soils is approximately 0.04 pCi/gram.

**Uranium – Naturally Occurring in the Earth's Crust** Uranium is found naturally in the earth's crust with an approximate average concentration of 1.6 pCi/gram. This amount varies depending on local geology, with natural uranium activity in Colorado soils ranging from approximately 0.5 to 3.0 pCi/g. Three isotopes compose natural uranium. The percent occurrences by mass are: uranium-238 (99.275%), uranium-235 (0.719%) and uranium-234 (0.0057%). Each of these isotopes has different amounts of activity per unit mass, which explains why the activity in soil emitted from uranium-234 approximately equals the activity from uranium-238, even though there is much less uranium-234 by mass (see "Radioactivity per Unit Mass," Page 8).

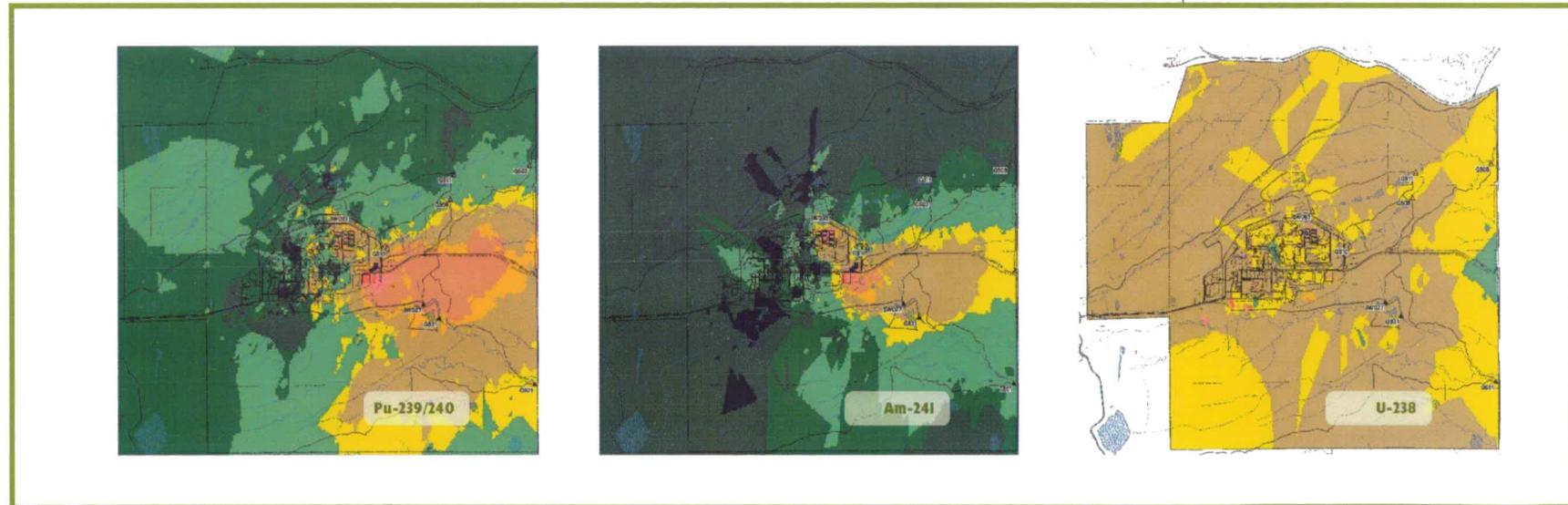
Uranium does not have the same spatial distribution observed for plutonium and americium in surface soils. Uranium is observed at varying levels of natural background activity across the Site, which complicates identifying uranium from man-made, versus natural, sources.

**DATA GAPS** The "Historical Release Report" identifies 215 total locations that are potentially contaminated by actinides. Acceptable data, as defined in the Technical Appendix, exist for surface or sub-surface soil contamination for plutonium, americium and uranium at 95 locations. Additional sampling is needed to more fully characterize actinide contamination at RFETS.

#### STATISTICAL METHODS USED WITH SOIL DATA

Although an extensive program exists to sample RFETS surface soils for actinides, it is not feasible to collect soil samples from every location at the Site. Therefore, to estimate actinide concentrations in soil at locations that have not been sampled, it is necessary to use data from adjacent locations that have been sampled. Various computerized estimation techniques have been developed for this purpose.

A geostatistical technique known as kriging was applied to the plutonium, americium and uranium surface soil sample data at RFETS to estimate concentrations of these actinides in the surface soil and generate the maps shown below.

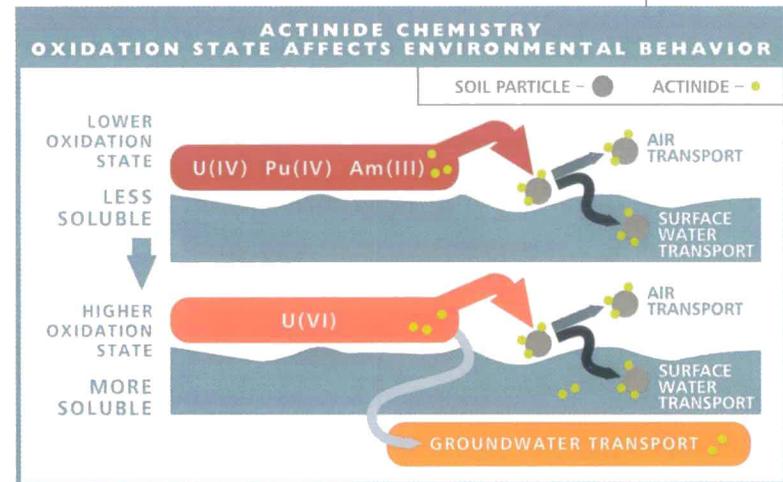


**ACTINIDE SURFACE SOIL MAPS** Surface soil data for plutonium (left) and americium (center) display a similar pattern of wind-driven dispersal to the east of the primary source area – the 903 Pad. In contrast, uranium (right) exists at natural background levels across most of the Site except for small areas of higher activity located near contamination sources. In these maps of kriged data, red indicates highest contamination activity and green indicates areas with lowest activity. Larger versions of these maps are in the Technical Appendix.

# ENVIRONMENTAL CHEMISTRY Pu, Am AND U

**TRANSPORT** Scientific literature and RFETS-specific studies indicate that the chemical and physical characteristics of plutonium, americium and uranium control how they are transported and where they eventually reside in the environment.

**OXIDATION STATES** The oxidation state of an actinide is determined by the number of electrons lost when the actinide combines with oxygen. The oxidation state is a function of the unique chemical characteristics of each actinide element as well as the geochemical conditions in the surrounding soil and water. In environmental conditions, plutonium and americium tend to exist in low oxidation states III (Am) and IV (Pu) that are relatively insoluble. In contrast, uranium is stable in both oxidation states IV and VI, with VI dominant in surface and near-surface oxidizing conditions. Because U (VI) forms compounds of greater solubility than Pu (IV) or Am (III), uranium exhibits a greater tendency to exist in chemical forms that are more soluble than plutonium or americium.



## COLLOIDS AND ACTINIDE TRANSPORT

Colloids are naturally occurring particles, defined as ranging in size from 0.1 to 0.001 micrometers. Colloids are found in nearly all surface water and groundwater and are formed as a result of the weathering of rocks, soils and decomposing plant materials. Due to their small size, colloids can remain suspended and are readily transported with groundwater. Suspended colloids are of interest as a transport mechanism for contaminants that strongly attach to mineral or organic surfaces, such as plutonium and americium (i.e., contaminants that do not readily dissolve in groundwater). The hydrology, water chemistry and geology of the surrounding environment influence the importance of colloids in facilitating transport of insoluble contaminants. Though colloid-facilitated transport of actinides has been observed at the Nevada Test Site, it is important to recognize that plutonium there was deposited during an underground nuclear test in fractured volcanic rock below the groundwater table. Geologic conditions at RFETS are significantly different than at the Nevada Test Site, but colloidal transport of actinides is a mechanism that still warrants consideration in the RFETS pathway analysis.

**PLUTONIUM AND AMERICIUM GEOCHEMISTRY** Because of the extremely low solubilities of plutonium and americium, these elements are predominantly associated with solids. They are either strongly sorbed, or attached, to soil and sediment particles or precipitated as oxides and hydroxides. The concentrations found in solution under the oxidizing environmental conditions common at RFETS are very low, around  $1 \times 10^{-15}$  moles/liter (also represented herein as  $1E-15$  moles/liter). Evidence indicates that reducing conditions which may exist in the treatment ponds or in landfill locations do not influence plutonium solubility at RFETS.

Studies performed to date and measurements at RFETS indicate that groundwater transport of plutonium and americium should be very low. Measured plutonium and americium concentrations in shallow groundwater below the Industrial Area range from the analytical detection limit (about 0.02 picocuries/liter [pCi/L]) to about 0.1 pCi/L. At present, it is not clear whether detections of plutonium and americium in shallow groundwater arise from surface contamination carried downward by well-drilling activities, from contamination during sampling and analysis, from sub-surface transport of actinide-bearing colloids or from a combination of these processes. These possibilities are currently being studied with a series of wells drilled and sampled under conditions that minimize the possibility of extraneous contamination.

Surface soil (0 to 15 centimeters [0 to 6 inches] below original grade), in contrast to the low levels observed in groundwater, has plutonium activities that range between 0 to 152,000 picocuries/gram [pCi/g]. Measurements of plutonium and americium movement show that the mobility of these actinides is largely controlled by erosion of surface soil by wind and water.

Since the data amassed indicate that plutonium and americium are present as insoluble forms and migration occurs via colloidal and particulate transport, contaminant transport modeling calculations must take these facts into account. Contaminant transport models that assume soluble forms and the existence of equilibrium conditions between soil and solution phases of plutonium and americium are of limited value for assessing the risk of exposure at RFETS. For plutonium and americium, models based on particulate transport processes are more appropriate and have been developed for use at the Site.

**URANIUM GEOCHEMISTRY** In contrast to plutonium and americium, uranium is most stable in the oxidation states IV and VI, with VI dominating in surface and near-surface oxidizing conditions. Because U (VI) forms compounds of much greater solubility than those formed by Pu (IV) or Am (III), uranium exhibits a greater tendency to exist in dissolved forms. Uranium is predominantly transported as dissolved chemical species, although transport can also occur in particulate form. Models used to estimate uranium transport must account for these processes and, accordingly, might suitably include a solubility and sorption-controlled mobility component.

### MEASURING RADIOACTIVITY

**What is a curie?** The curie (Ci) is a unit of measure for radioactivity. The nuclei of the heaviest elements in the periodic table are unstable and emit radiation when their nuclei break up. An element that emits radiation is called radioactive and the emission process is often referred to as radioactive decay. The Ci was established as a unit of measure based on the radioactivity emitted by 1 gram of radium-226. The Ci is defined as  $3.7 \times 10^{10}$  nuclear decays per second. The activity emitted by a gram of an isotope of a radioactive element may vary greatly from the activity emitted by a gram of a different element or a different isotope and is related to its rate of radioactive decay (the half-life). Therefore, it is more meaningful to use a measure of radioactivity like the Ci, versus using mass or volume units, when discussing actinides and their radioactivity.

**What is a picocurie?** A picocurie (pCi) is one trillionth of a Ci ( $1 \times 10^{-12}$  Ci). For studying actinides in the environment at RFETS, the Ci is often too large a unit of radioactivity in the same way that a fraction of a mile would be an awkward way to describe the thickness of a human hair. Therefore, activity in the environment at RFETS is frequently presented in units of pCi.

### RADIOACTIVITY PER UNIT MASS

Specific activity is used to quantify the amount of radioactivity emitted per unit of mass. The specific activity for each isotope of a given element is related to its radioactive half-life. The half-life is the time it takes for half of the atoms to decay. Specific activities for isotopes of interest are listed below. Note how the amount of activity per unit mass can vary by several orders of magnitude from one actinide isotope to another.

RADIONUCLIDE	HALF-LIFE (years)	SPECIFIC ACTIVITY (Ci/gram)
americium-241	$4.32 \times 10^2$	$3.53 \times 10^0$
plutonium-239	$2.42 \times 10^4$	$8.48 \times 10^{-2}$
plutonium-240	$6.57 \times 10^3$	$3.10 \times 10^{-2}$
uranium-234	$2.47 \times 10^5$	$6.25 \times 10^{-3}$
uranium-235	$7.04 \times 10^8$	$2.14 \times 10^{-6}$
uranium-236	$2.34 \times 10^7$	$8.85 \times 10^{-6}$
uranium-238	$4.51 \times 10^9$	$3.33 \times 10^{-7}$

An example of the importance of specific activity is demonstrated by examining the natural occurrence of uranium. Three uranium isotopes are found naturally in the environment. By mass, uranium-238 accounts for nearly all (99.275 %) of the naturally-occurring uranium, while uranium-235 (0.719 %) and uranium-234 (0.0057 %) account for the remaining mass. However, in terms of radioactivity, the amount of activity emitted from naturally-occurring uranium-234 and uranium-238 is roughly equal, despite the overwhelming abundance of uranium-238 atoms in a given sample.

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# AIR PATHWAY

**INTRODUCTION** Transport of actinides through the air at RFETS occurs largely by wind erosion of actinide-containing particulate matter from soil and vegetation surfaces. RFETS-specific research suggests that dust-laden vegetation is the primary source for resuspended airborne plutonium under most conditions (Langer, 1991). Resuspension of actinides directly from soil surfaces is thought to be a lesser source except during high wind events or after soil has been disturbed and made more erosion-prone. Building stack and vent emissions are, to a much lesser extent, also sources of airborne actinides, though these sources will be eliminated as buildings are removed.

Overall, the general direction of airborne actinide transport at the Site follows the prevailing winds, from the north and west to the south and east. More importantly, Site data show that higher wind speeds occur almost exclusively from the northwest quadrant. This is significant because the amounts of soil resuspended are much higher during high-wind events than during periods with lower winds. Higher winds are also more effective at transporting particles further downwind from source areas before being redeposited.

Although the first few minutes of high winds may result in significant airborne particle transport, the emission rate decreases rapidly with time as the available inventory of erosion-prone particles is depleted. Sustained windy periods do not result in significantly greater emissions until the inventory is replenished by deposition or by other factors that increase soil erosion potential, such as freeze/thaw cycles, wet/dry cycles, rangeland fires, animal activities, rainsplash effects or other processes that disturb the soil. Following disturbances, erosion protection is restored by crusting of the soil, regrowth of vegetation and regeneration of a litter layer.

**METHODOLOGY FOR QUANTIFYING ACTINIDE TRANSPORT** Two different methods were used to quantify actinide transport via the air pathway. The first method is more closely linked to measured site data. It uses airborne average actinide concentration data from 1997 through 1999, collected at site perimeter monitoring stations, coupled with on-site wind data.

The second method involves a wind erosion emission estimation method and dispersion / deposition model developed for the Site. Off-site airborne transport was calculated for plutonium and americium as the difference between annual wind erosion emissions from the Site and deposition of actinides back onto the Site. Though this approach does not account for possible contributions from project or building emissions, wind erosion of actinides from soil and vegetation has been determined to represent the majority of air emissions from the Site during recent years.



Data collected from air-monitoring stations like this one, near the 903 Pad, are used to quantify actinide movement by the wind. Air is a major transport pathway.

Although the first method is a more "data-driven" estimation approach, it has uncertainty associated with wind speed data and airborne actinide data collected in different time steps, 15-minute and monthly intervals, respectively. The dispersion modeling approach, though not tied as closely to measured air actinide concentrations, provides the advantage that hypothetical off-normal events can also be investigated. Results from both methods, for normal conditions, provide a range of results for estimated annual quantities of airborne actinides transported off site.

**CHART 1**

**AIRBORNE ACTINIDE CONCENTRATIONS – MEDIAN MEASURED ACTIVITIES AT SITE PERIMETER COMPARED WITH REGIONAL BACKGROUND ACTIVITIES**

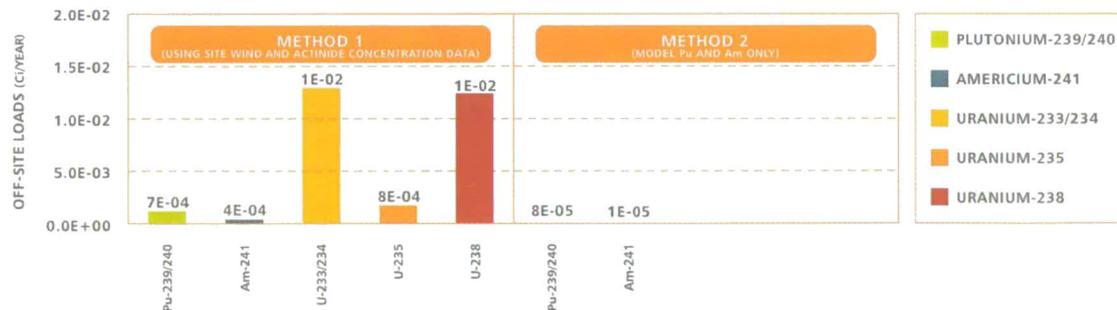


**ACTINIDE CONCENTRATIONS IN AIR**

Chart 1 presents airborne actinide concentrations measured at the RFETS boundary. Units of measurement are pCi per cubic meter of air. Regional background activities are provided for plutonium and americium for comparison. Background concentrations of airborne plutonium and americium exist, as discussed earlier, because they were globally dispersed from historic weapons testing. Resuspension by the wind of the residual plutonium and americium causes a background level of these actinides in the air. Airborne uranium measured at the Site is similar to background because of its natural abundance in the soil. In Chart 1, the concentration presented for each actinide is the median of annual average concentrations measured at the RFETS perimeter monitoring locations from 1997 through 1999.

**CHART 2**

**AIRBORNE ACTINIDES – TOTAL AVERAGE ANNUAL ACTIVITY TRANSPORTED OFF SITE – RESULTS FOR TWO ESTIMATION METHODS**



**TOTAL ACTIVITY IN AIR**

Estimated annual off-site airborne actinide loads are shown in Chart 2. Results are presented for two modeling methods described previously.

## AIR PATHWAY

**DISCUSSION: AIRBORNE PLUTONIUM AND AMERICIUM** Model estimates for average annual off-site transport of plutonium range from  $8 \times 10^{-5}$  Ci to  $7 \times 10^{-4}$  Ci and for americium range from  $1 \times 10^{-5}$  Ci to  $4 \times 10^{-4}$  Ci. For both plutonium and americium, the estimation method based on measured Site wind and airborne actinide concentration data yielded higher predicted off-site transport than the model estimation method. The primary source of plutonium and americium in airborne loads at RFETS is from contaminated surface soil, or soil on vegetation surfaces, in the area near and east of the 903 Pad. Additional minor sources are building stack and vent emissions as well as background plutonium and americium in surface soil from global atmospheric nuclear fallout that gets resuspended by the wind.

Modeling results are consistent with the observed pattern of plutonium and americium surface soil contamination, originating in the 903 Pad area and migrating eastward as a result of prevailing winds from the west and northwest. Reconstruction of events associated with the 903 Pad contamination in the late 1960s suggests that much of the contamination was likely dispersed during a few high-wind events that followed closely after the contaminated soil had been disturbed by grading or weed control efforts (Meyer et al., 1996). Such activities can break up the surface crust, crush aggregated soil particles and remove vegetative cover, thereby renewing and increasing the reservoir of particles available for erosion. The resulting dispersion and deposition pattern indicates that substantial quantities of material can be moved through the air pathway by the sporadic events.

**DISCUSSION: AIRBORNE URANIUM** Naturally occurring uranium from the soil is the major component of airborne uranium leaving the Site. Based on the relative concentrations of uranium-233/234 and uranium-238, data from the sampling network confirm that almost all airborne uranium is naturally occurring. For comparison, the concentration of airborne uranium-233/234 activity measured at site boundary monitors ranges from 10 to 60 times more than the activity measured for airborne plutonium.

**DISCUSSION: EXTREME EVENTS** As a hypothetical extreme event, a model simulation was performed to study the effect on airborne actinide transport following a rangeland fire occurring on approximately 40 hectares (100 acres) in a plutonium-contaminated area near the 903 Pad. Modeling results indicate that average airborne plutonium concentrations would increase an estimated 5- to 13-fold in the vicinity of the burned area in the first year following a fire. Such an increase in concentrations would lead to greater off-site transport until the vegetation recovered and soil loss from wind erosion returned to pre-fire levels. The actual increase in actinide transport following a fire would depend on the size of the burned area, the intensity of the fire and the actinide concentrations in the area burned. Other extreme conditions, such as soil disturbance by heavy equipment, can increase airborne particulate emissions by nearly a factor of 20 (EPA, 1995).

# AIRBORNE ACTINIDE CONCENTRATIONS

## AIRBORNE ACTINIDES

The air-monitoring location with the highest total average actinide concentration had a level equal to approximately 1.4 percent of the 10 millirem standard governing airborne radionuclide concentrations leaving DOE facilities. Results are based on data collected from 1997 through 1999.

S-201	
Pu-239	1.07E-06
Am-241	5.46E-07
U-233/234	2.51E-05
U-235	1.20E-06
U-238	2.25E-05
0.7% of standard	

S-254 *	
Pu-239	4.35E-07
Am-241	4.39E-07
U-233/234	2.32E-05
U-235	1.65E-06
U-238	2.90E-05
0.7% of standard	

S-134	
Pu-239	7.10E-07
Am-241	3.31E-07
U-233/234	1.77E-05
U-235	9.65E-07
U-238	1.61E-05
0.5% of standard	

S-201 *	
Pu-239	9.03E-07
Am-241	7.64E-07
U-233/234	3.60E-05
U-235	2.31E-06
U-238	3.59E-05
1.1% of standard	

S-136	
Pu-239	7.90E-07
Am-241	4.66E-07
U-233/234	1.82E-05
U-235	9.84E-07
U-238	1.63E-05
0.5% of standard	

S-132	
Pu-239	1.13E-06
Am-241	3.29E-07
U-233/234	4.37E-05
U-235	2.09E-06
U-238	4.38E-05
1.2% of standard	

S-131	
Pu-239	9.88E-07
Am-241	9.91E-07
U-233/234	3.03E-05
U-235	1.45E-06
U-238	2.94E-05
0.9% of standard	

S-137	
Pu-239	1.38E-06
Am-241	2.06E-07
U-233/234	2.72E-05
U-235	6.57E-07
U-238	2.25E-05
0.7% of standard	

S-138	
Pu-239	1.81E-06
Am-241	4.07E-07
U-233/234	2.00E-05
U-235	1.23E-06
U-238	1.92E-05
0.6% of standard	

S-207	
Pu-239	9.70E-07
Am-241	3.64E-08
U-233/234	7.65E-05
U-235	1.18E-06
U-238	2.57E-06
0.7% of standard	

S-209	
Pu-239	5.29E-07
Am-241	1.67E-07
U-233/234	2.45E-05
U-235	1.13E-06
U-238	2.38E-05
0.7% of standard	

S-142	
Pu-239	3.95E-07
Am-241	2.88E-07
U-233/234	2.16E-05
U-235	1.43E-06
U-238	2.05E-05
0.6% of standard	

S-141	
Pu-239	9.48E-07
Am-241	1.11E-07
U-233/234	2.09E-05
U-235	1.11E-06
U-238	2.06E-05
0.6% of standard	

S-139	
Pu-239	8.88E-07
Am-241	5.73E-07
U-233/234	2.87E-05
U-235	1.31E-06
U-238	2.74E-05
0.8% of standard	



STATION S-140		
Pu-239	7.92E-07	pCi/m <sup>3</sup>
Am-241	3.70E-07	pCi/m <sup>3</sup>
U-233/234	4.82E-05	pCi/m <sup>3</sup>
U-235	2.36E-06	pCi/m <sup>3</sup>
U-238	4.81E-05	pCi/m <sup>3</sup>
1.4% of standard		

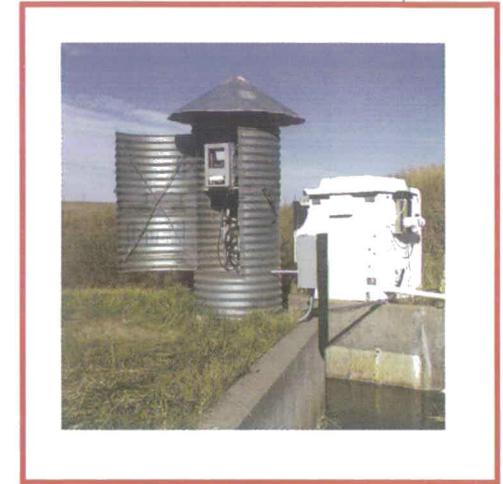
# SURFACE WATER PATHWAY

**INTRODUCTION** Actinides are transported in surface water by two main processes, depending on the actinide's solubility. First, insoluble actinides, such as plutonium, americium or uranium in lower oxidation states, sorb to soil or sediment particles that are eroded by water. The particles thereby transport the attached actinides. The second transport process involves actinides in solution, primarily uranium in the VI oxidation state, that move in surface water. Plutonium and americium are essentially insoluble and are not transported as dissolved species in significant quantities.

Surface water at RFETS flows generally from west to east, with three major drainages traversing the Site (see map at back of report, Page v). Walnut Creek drains the northern portion of the Site, including the majority of the Industrial Area, which runs off to the A- and B-series detention ponds. Woman Creek drains the southern portion of the Site, including southern Industrial Area runoff after it is diverted by the South Interceptor Ditch into Pond C-2. The third major drainage, Rock Creek, does not receive runoff from the Industrial Area or other contaminated areas. This pathway study focuses on the Walnut and Woman Creek drainage basins.

**METHODOLOGY FOR QUANTIFYING ACTINIDE TRANSPORT** The amount of actinide material, or load, transported in surface water past a specific location is a function of both the volume of water that flows past the location and the actinide concentration in the water. This surface water actinide load is calculated using data from automated monitoring stations that continuously measure water flow and periodically collect samples using a "flow-weighted" sampling protocol. This means sample volumes are collected in equal proportion to the volume of water passing the station. Multiple samples are collected and combined, resulting in an accumulated composite sample. The sample is representative of the actinide concentration for an entire volume of water passing the monitoring station. Annual surface water actinide loads were quantified in this study at eight site monitoring locations, using data from water years 1997 through 1999.

In addition to using measured data to quantify surface water actinide loads, models were developed to estimate impacts to surface water from pathways for which measured data is unavailable. Estimates of plutonium and uranium-238 inputs and outputs to surface water were made for: 1) deposition of airborne actinides to surface water, using a Gaussian plume model; 2) hillslope erosion and runoff of actinides to surface water, using the Watershed Erosion Prediction Project (WEPP) model coupled with actinide soil data; and 3) inflow and outflow of actinides to surface water from shallow alluvial sub-surface water, using water balance calculations coupled with monitoring-well data. These mass balance analyses were conducted on three study areas: the Walnut Creek detention ponds, Walnut Creek between the ponds and the site boundary and the South Interceptor Ditch drainage basin.



Surface water is monitored throughout the Site at automated stations. When the water flow rate of the water increases, this unit is programmed to increase the number of samples it collects.

**CHART 3**

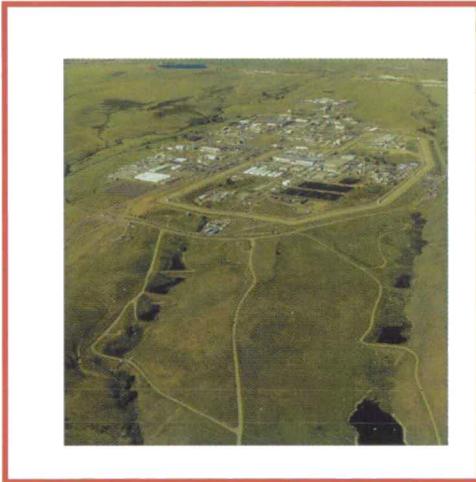
**SURFACE WATER ACTINIDE CONCENTRATIONS – WALNUT AND WOMAN CREEKS COMPARED WITH REGIONAL BACKGROUND CONCENTRATIONS IN SURFACE WATER**



NOTE: BOUNDARY URANIUM CONCENTRATIONS ESTIMATED USING VOLUME-WEIGHTED DATA FROM UPSTREAM STATIONS

**SURFACE WATER CONCENTRATIONS**

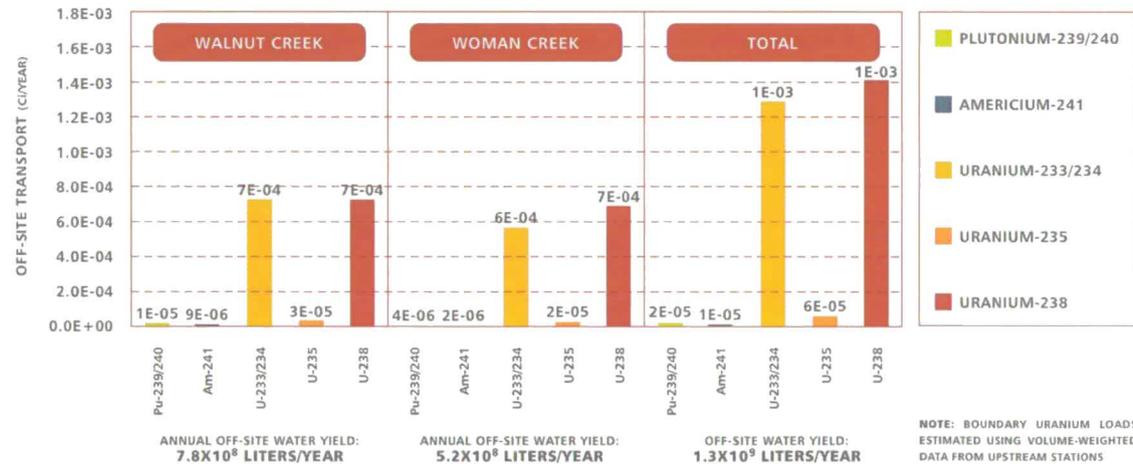
Average surface water actinide concentrations in Walnut and Woman Creeks at the Site's eastern boundary are presented in Chart 3. Concentrations were calculated using a volume-weighted average based on samples and flow data collected from water years 1997 through 1999. Site measurements are compared with background concentrations of actinides measured in Front Range regional surface water that is not impacted by RFETS.



The actively managed detention ponds on South Walnut Creek (left) and North Walnut Creek (right) settle out 80 to 90 percent of the plutonium and americium loads carried into them from runoff.

**CHART 4**

**SURFACE WATER ACTINIDE LOADS – ESTIMATED OFF-SITE TRANSPORT**



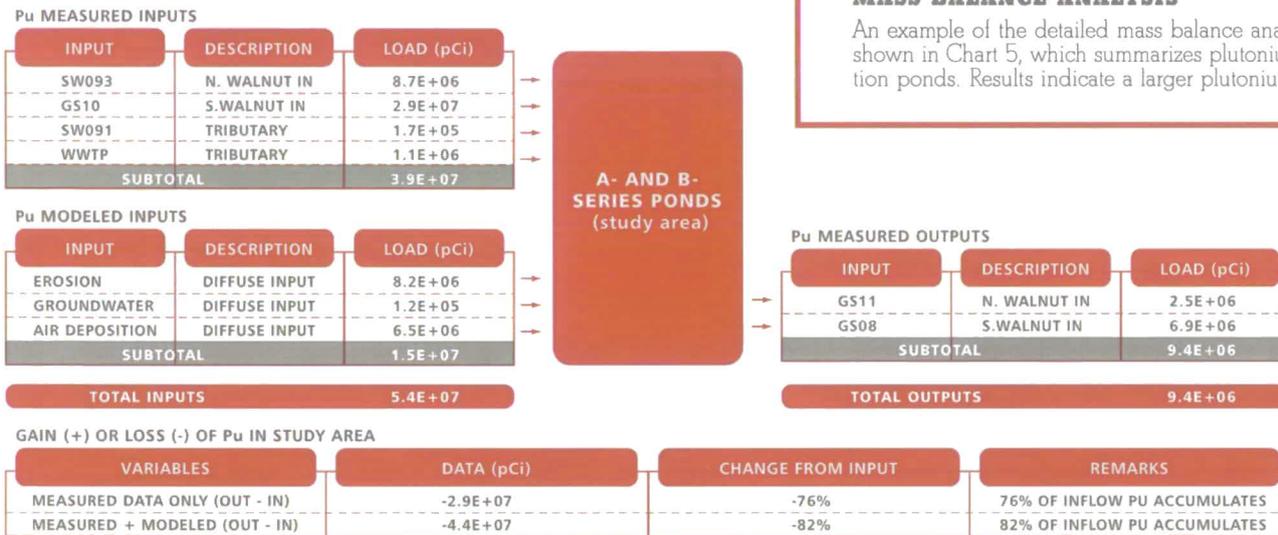
NOTE: BOUNDARY URANIUM LOADS ESTIMATED USING VOLUME-WEIGHTED DATA FROM UPSTREAM STATIONS

**SURFACE WATER LOADS**

Off-site actinide loads in the Walnut and Woman Creek drainage basins, as well as the total load of both basins combined, are summarized in Chart 4. The measured average annual volume of surface water flowing off site is displayed for each basin at the bottom of the chart.

# SURFACE WATER PATHWAY

**CHART 5**



## MASS BALANCE ANALYSIS

An example of the detailed mass balance analyses performed on three surface water study areas is shown in Chart 5, which summarizes plutonium input and output loads to the Walnut Creek detention ponds. Results indicate a larger plutonium load flowing into the ponds than flowing out. This accumulation of plutonium in the ponds is attributed to particle settling which removes plutonium from the water column. Contributions of modeled input loads, such as deposition of airborne plutonium to surface water, are also quantified. A similar analysis for uranium-238 was done in the same study area. Those results are tabulated in the Technical Appendix. Other study areas analyzed in the same manner are the South Interceptor Ditch drainage basin and the section of Walnut Creek between the terminal ponds and the site boundary.

**DISCUSSION: PLUTONIUM AND AMERICIUM IN SURFACE WATER** The South Interceptor Ditch drainage basin, which includes hillslopes near the 903 Pad, has the highest levels of surface soil plutonium contamination at the Site. This basin is characterized by well-vegetated slopes and has only 14 percent impervious surface coverage. In contrast, the highly-developed central Industrial Area drainage basin is covered by approximately 47 percent impervious surfaces. Therefore, the South Interceptor Ditch basin has more water infiltration and less runoff per unit area than the central Industrial Area. Less runoff equates to less soil erosion and less actinide transport. As a result, despite having higher plutonium activities in the soil, the surface water plutonium load discharged per square meter of the South Interceptor Ditch basin (3.8 pCi/m<sup>2</sup>/year) is roughly one-tenth of that measured in the central Industrial Area runoff.

Average concentrations of plutonium in surface water vary by a factor of nearly 40 at monitoring stations across the Site. Average plutonium concentrations measured in surface water range from 0.191 pCi/L, for central Industrial Area runoff monitored at station GS10, to 0.005 pCi/L for Woman Creek at station GS01 located near Indiana Street.

The actively managed detention ponds on North and South Walnut Creeks settle out particles and, as a result, remove roughly 80 percent to 90 percent of the plutonium and americium that flows into the ponds. The fraction of plutonium that doesn't settle is at least partially explained by site research which indicates approximately 10 percent of the plutonium and americium in runoff from the central Industrial Area, at station GS10, is attached to sub-micrometer-sized colloid particles (Santschi, 2000). The colloids are not likely to settle in the ponds. An additional important observation regarding

plutonium transport involves the lower section of Walnut Creek, between the terminal detention ponds and the site boundary, where the average annual plutonium load measured at the downstream end is approximately 30 percent greater than the plutonium load measured at the upstream end. Site investigations suggest the plutonium source in this area is diffuse, low-level legacy contamination in watershed soils and channel sediments (RMRS, 1998).

**DISCUSSION: URANIUM IN SURFACE WATER** Concentrations of uranium, in contrast to plutonium and americium, are relatively uniform in surface water across the Site. As a result, uranium loads in each basin are largely a function of each basin's water yield. Quantifying the fractions of natural versus man-made uranium in surface water requires that samples be analyzed using a high-resolution analytical technique, such as inductively coupled plasma/mass spectrometry (ICP/MS). This type of analysis is planned to permit more accurate detection of man-made uranium in site surface water. Although surface water flowing from RFETS is not utilized for drinking water supplies, comparison with the drinking water standard for uranium provides perspective on water quality. Total uranium concentrations at RFETS Point of Evaluation and Point of Compliance monitoring stations from water years 1997 through 1999 averaged roughly one-tenth of the 30 microgram per liter Maximum Contaminant Level for drinking water.

**DISCUSSION: AIR-TO-SURFACE WATER PATHWAY** Model estimates were generated to characterize the air-to-surface water pathway for plutonium and uranium-238. These analytes also serve as analogs for the transport behavior of americium and other uranium isotopes. Model estimates indicate the air-to-surface water pathway provides a relatively minor load, less than 1 percent of the total input to surface water, for all actinides and for all areas of the Site, with one exception. For the Walnut Creek detention ponds, model results indicate approximately 12 percent of the total input load is from airborne deposition to surface water. The increased fraction from airborne deposition in this location is a function of the large surface area of the ponds and the close proximity of the 903 Pad, a large surface soil plutonium source.

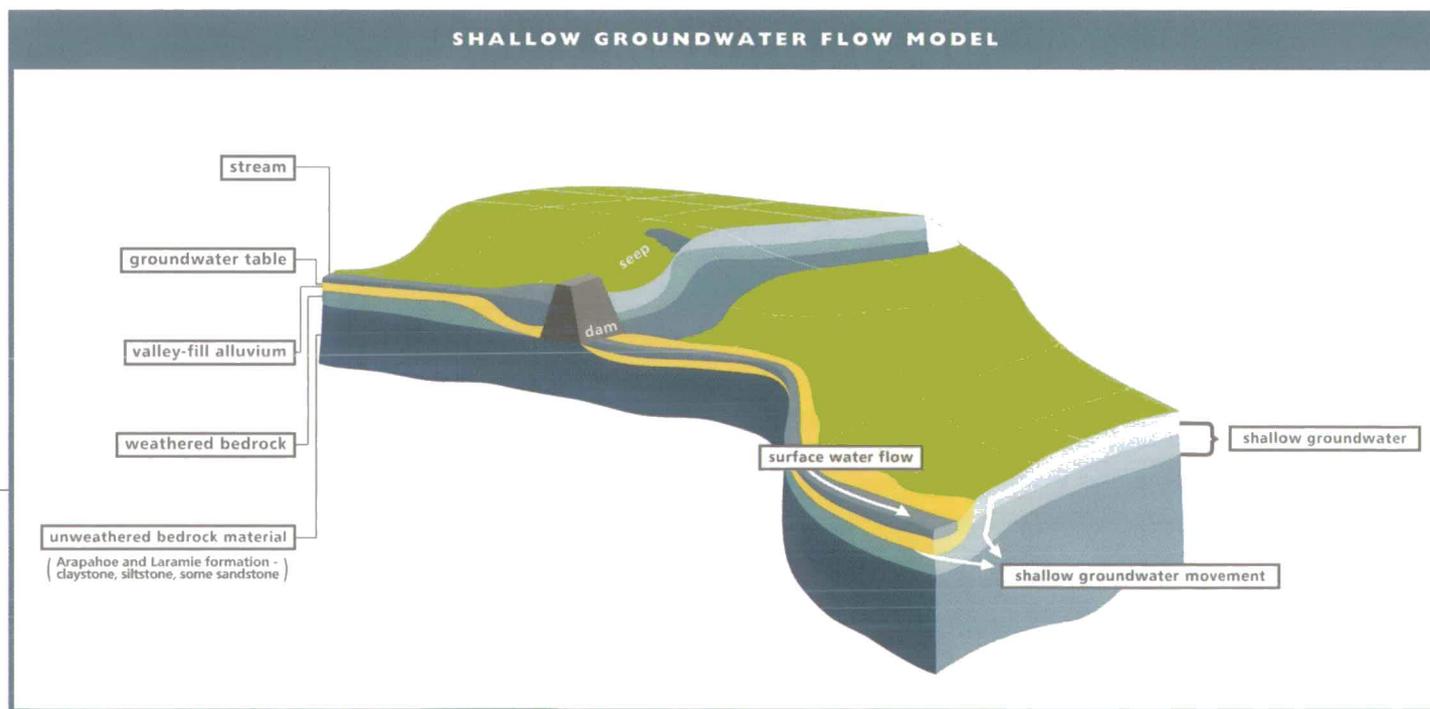
**DISCUSSION: SURFACE WATER INTERACTION WITH SUB-SURFACE WATER** For plutonium, flow between surface water and shallow sub-surface alluvial water is a relatively minor transport pathway to and from surface water, comprising 1 percent or less of the total input or output load for any of the areas studied. In contrast, uranium transport in the shallow sub-surface is a relatively major pathway. Model estimates for uranium-238 in shallow sub-surface flow ranged from 7 percent of the output load in lower Walnut Creek to 83 percent of the input load in the South Interceptor Ditch basin.

**DISCUSSION: EXTREME EVENTS** Model estimates of erosion indicate the plutonium load delivered from the South Interceptor Ditch basin is greater relative to other watersheds during extreme events. The plutonium load delivered from the 100-year, 6-hour storm event (97.1 mm) at the downstream end of the South Interceptor Ditch is approximately four times larger than the load delivered off site in Walnut Creek during the same storm. The explanation for the model-predicted impact of large storms is that the highest levels of plutonium contamination on Site are within the South Interceptor Ditch watershed. The hillslopes are well vegetated and have little runoff or erosion and plutonium transport, until an extreme storm event occurs. Remediation of soils within the South Interceptor Ditch watershed will reduce actinide loads transported in extreme events.

# GROUNDWATER PATHWAY

**INTRODUCTION** Flowing beneath the ground surface, groundwater represents another pathway by which actinides can potentially be transported. This study focuses on "shallow" alluvial groundwater because geologic conditions at RFETS limit the depth of groundwater potentially impacted by Site contamination. Shallow groundwater refers to water flowing in the Site's alluvium and weathered bedrock geologic units and is found from just below the ground surface to depths of approximately 30 meters (100 feet), as shown in the figure below.

Shallow groundwater and surface water are inextricably linked. Water from stream channels infiltrates downward, recharging the shallow groundwater. Seeps discharge shallow groundwater to the surface. Therefore, it is not surprising that an actinide's solubility, which controls actinide transport in surface water, also dictates actinide transport in shallow groundwater. Insoluble actinides, such as plutonium, americium and uranium in the IV oxidation state, are relatively immobile in the soil and groundwater environment due to their low aqueous solubility and tendency to strongly sorb on soil media (Cleveland et al., 1976 and Honeyman and Santschi, 1997). However, work at RFETS, as well as studies in the literature, have shown that insoluble actinides can sorb to natural, sub-micrometer-sized colloid particles that can potentially facilitate actinide movement (Santschi, 2000). Another transport process similar to that observed in surface water involves more soluble actinides, such as uranium in the IV oxidation state, that move in solution with the shallow groundwater flow.



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Beneath areas with shallow groundwater flows in the alluvium and weathered bedrock geologic units, there is a thick, highly-impermeable, unweathered section of bedrock that inhibits downward groundwater flow. Because the shallow groundwater is inhibited from flowing vertically downward, it preferentially moves laterally along the unweathered bedrock surface and generally flows from west to east. The shallow groundwater flow is directed toward streams, where it either discharges as baseflow into the stream, evapotranspires to the atmosphere or continues as shallow groundwater flowing downstream within the more permeable valley-fill alluvium material just below the ground surface. Yet deeper, below the unweathered bedrock unit, is the regional Laramie-Fox Hills aquifer, approximately 200 to 300 meters (650 to 1,000 feet) below the Site. A U.S. Geological Survey study and a separate, peer-reviewed site investigation both indicate this aquifer will not be impacted by site activities because of the intervening unweathered bedrock layer, specifically the Laramie Formation, that has claystones with low hydraulic conductivities (Hurr, 1976; RMRS, 1996).

**METHODOLOGY FOR QUANTIFYING ACTINIDE TRANSPORT** Calculating actinide quantities transported off site each year in shallow groundwater requires quantifying: 1) the volume of shallow groundwater flowing off site; and 2) concentrations of different actinides in the shallow groundwater.

The volume of shallow groundwater flowing off site, or shallow groundwater flux, was calculated using the site-wide water balance model that uses the "MIKE SHE" computer code. This hydrologic model simulates all of the significant integrated hydrologic flow processes including overland flow, channel flow and sub-surface flow in the saturated and unsaturated zones. Lateral shallow groundwater flow off-site is computed for saturated flow within the unconsolidated alluvial and weathered bedrock material. For actinide transport analysis, off site shallow groundwater flux volumes were estimated for water year 2000 (from October 1999 through September 2000) for the Walnut Creek and Woman Creek groundwater basins. In addition to using model results for a normal precipitation year, shallow groundwater flux was estimated using precipitation data for January through May of 1995. Approximately 340 mm (13.5 in), or twice the average amount, of precipitation fell during this period. These model results provide insight into shallow groundwater flows during wet conditions.

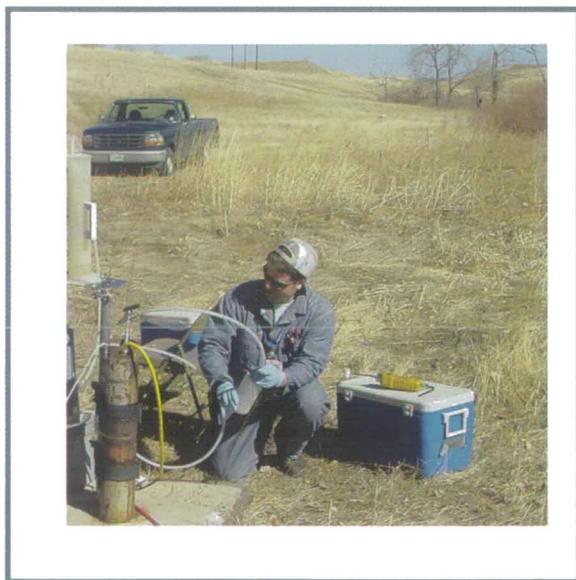
Shallow groundwater actinide measurements, collected from alluvial wells near Walnut and Woman Creeks at the Site's eastern boundary, were used to determine the concentration of actinides in shallow groundwater flowing off site. The estimated annual shallow groundwater flux volumes for the Walnut and Woman Creek basins were multiplied by the average actinide concentrations within each basin to estimate the actinide loads transported off site in shallow groundwater.

# GROUNDWATER PATHWAY

**DISCUSSION: PLUTONIUM AND AMERICIUM IN SHALLOW GROUNDWATER** Determination of plutonium and americium concentrations in shallow groundwater at the Site is complicated by residual surface soil contamination potentially introduced down boreholes during drilling and well installation operations. Shallow groundwater samples collected using traditional bailing techniques may suspend these contaminated drilling-artifact soil materials, thereby producing shallow groundwater samples with artificially high plutonium or americium concentrations. As a result of potential well construction and sampling biases, new clean or "aseptic wells" were drilled and efforts to improve sampling protocols undertaken. This work is currently ongoing. Therefore, plutonium and americium concentrations in shallow groundwater wells used in this analysis may represent a "worst case" scenario. Mean plutonium activities in alluvial wells at the site boundary were 0.035 pCi/L (+/- 0.018 pCi/L) in the Walnut Creek shallow groundwater basin and 0.003 pCi/L (+/- 0.004 pCi/L) in the Woman Creek shallow groundwater basin.

**DISCUSSION: URANIUM IN SHALLOW GROUNDWATER** Uranium-233/234 and uranium-238 isotopes are the dominant actinides found in groundwater in terms of total activity because of their natural abundance, particularly in the RFETS region. Though the concentration of uranium in groundwater at RFETS is within the natural range, shallow groundwater flowing from the Site can have uranium from man-made sources. Special analytical techniques, such as ICP/MS, must be used to study isotopic ratios in the groundwater and determine whether any of the uranium has origins from man-made sources. For natural uranium, the ratio of uranium-235/uranium-238, by mass, is approximately 0.0072. A ratio less than 0.0072 indicates the presence of man-made uranium-238, or "depleted" uranium, whereas a ratio greater than 0.0072 indicates the presence of man-made uranium-235, or "enriched" uranium. Additionally, ICP/MS analysis can detect the presence of uranium-236, a reactor product that is not found in natural uranium.

Samples collected at site wells from July 1999 to August 2000 were analyzed using ICP/MS. Most samples indicated uranium from natural sources. However, alluvial groundwater samples collected near the site boundary in both the Walnut and Woman Creek groundwater basins had uranium-235/uranium-238 mass ratios slightly less than the 0.0072 ratio found naturally. The small variation from the natural ratio, though potentially related to analytical uncertainty, indicates the shallow groundwater in these basins may have a small fraction of man-made "depleted uranium" as part of the total uranium concentration. In addition, the same Walnut Creek boundary location had detectable levels of uranium-236, an isotope that comes only from a man-made uranium source (RMRS, 2000).



Actinide concentrations in groundwater are determined by analyzing samples collected from wells. Most of the uranium found in groundwater at RFETS is from natural sources. Special analytical techniques are used to determine if any fraction comes from man-made uranium sources.

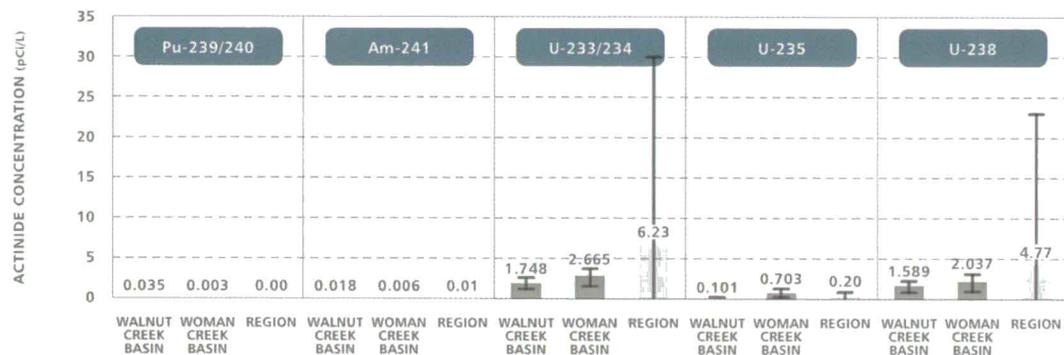
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### GROUNDWATER CONCENTRATIONS

Chart 6 displays shallow groundwater actinide concentrations in the RFETS Walnut and Woman Creek groundwater basins. Site measurements are compared with background concentrations of actinides measured in Front Range regional upper hydrostratigraphic unit groundwater, or shallow groundwater, that is not impacted by RFETS.

CHART 6

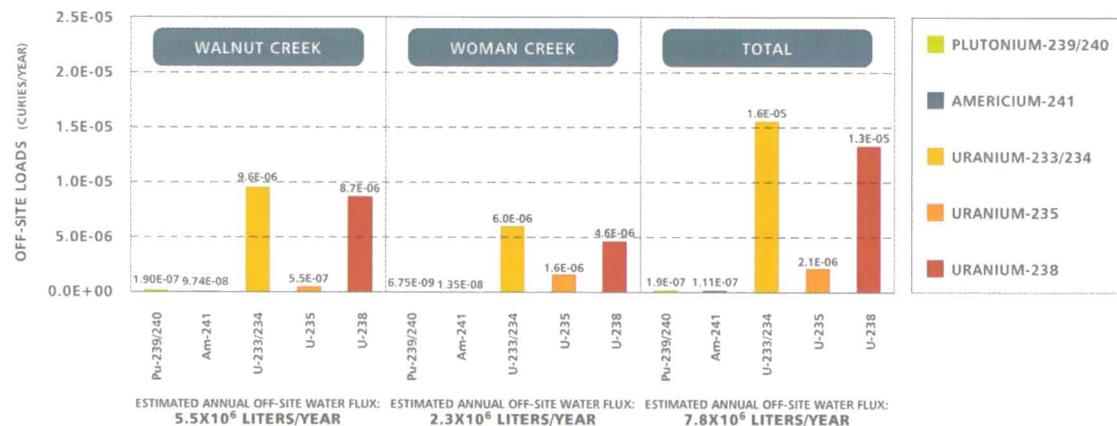
### SHALLOW GROUNDWATER ACTINIDE CONCENTRATIONS - WALNUT AND WOMAN CREEK GROUNDWATER BASINS COMPARED WITH REGIONAL BACKGROUND ACTIVITY IN GROUNDWATER



NOTE: ERROR BARS REPRESENT MEAN ± 1 STANDARD DEVIATION ANALYTICAL ERROR OF ALL RESULTS

CHART 7

### SHALLOW GROUNDWATER ACTINIDE LOADS - ESTIMATED ANNUAL OFF-SITE TRANSPORT BASED ON WATER YEAR 2000 PRECIPITATION



### GROUNDWATER LOADS

Shallow groundwater actinide loads transported off site in the Walnut and Woman Creek groundwater basins are summarized in Chart 7. The model-estimated average annual volume of shallow groundwater yielded off site is displayed for each basin at the bottom of the chart.

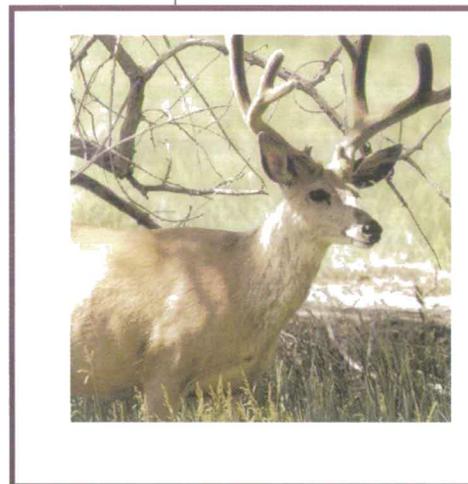
### GROUNDWATER FLUX - WET CONDITIONS

Model estimates of increased shallow groundwater flux during extreme precipitation conditions were calculated for May 1995, when 194 mm (7.65 in) of precipitation occurred, or roughly three times the May norm. The estimated flux of shallow groundwater flowing off site increased by approximately 100 percent in the Walnut Creek drainage and approximately 50 percent in the Woman Creek drainage. This provides some basis for estimating the impacts of extreme precipitation events on shallow groundwater flow and related actinide transport.

# BIOLOGICAL PATHWAY

**INTRODUCTION** Movement of actinides via the biological pathway can occur by a variety of mechanisms that range from transport of soil and actinides by insects to actinide transport by deer that have ingested vegetation with actinide-bearing soil on plant surfaces. A large body of scientific literature addresses quantitative estimates of actinide intake and movement by different biological entities. Much of this research was specific to RFETS, including an extensive series of radioecology studies conducted from the 1960s through the 1990s by the Department of Radiology and Radiation Biology at Colorado State University (Whicker, 1979; Little, et. al., 1980; Webb, et al., 1993). These studies generally concentrated on areas contaminated with plutonium and other actinides in various compartments of the RFETS ecosystem and used field measurements and laboratory analyses of actinides in plant and animal tissues.

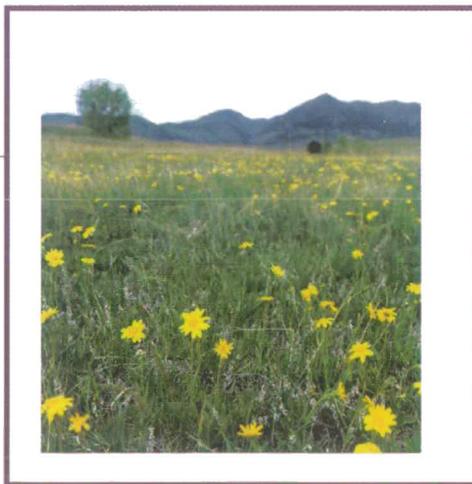
Site-specific research has been conducted on mule deer as a biological pathway for actinide movement for several reasons, including their mobility, amount of soil intake and their relative abundance, with a herd size of approximately 140 (Kaiser-Hill, 2000). Quantifying the off-site transport of actinides by mule deer provides a reference for comparing the effects of the overall macro-biological transport pathway. Other biological transport pathways and mechanisms, such as vegetation uptake of actinides and biogeochemical processes, are not quantified here but are addressed later in the Discussion section of this text (Page 22).



**Mule deer have been the focus of research as the most likely mechanism for biological actinide transport at RFETS.**

## METHODOLOGIES FOR QUANTIFYING ACTINIDE TRANSPORT

Two different methods were used to quantify actinide transport off site via the biological pathway. The first method is based on a site-specific study that estimated less than  $1 \times 10^{-7}$  (one ten-millionth) of the plutonium inventory in soil is moved around the Site by mule deer each year and most of this is redeposited on DOE-controlled property (Whicker, 1979). This value, combined with data on the plutonium inventory in soil and average soil activity, provided a basis for calculating the amount of soil moved by mule deer. The second actinide transport estimation method is based on RFETS data quantifying the average amount of soil consumed by mule deer, over the year, to be approximately 16 grams per day (Arthur and Alldredge, 1979).



**The Buffer Zone serves as attractive habitat for the Site's approximately 140 mule deer. Tracking data indicate approximately 5 percent of the herd leave the Site each year.**

The estimated soil quantities moved or ingested by mule deer on site were used with additional data to quantify the amount of soil transported off site by mule deer. The other information included telemetry data that indicate approximately 5 percent of the deer herd leave the Site annually (Symonds and Alldredge, 1992). The time for a deer to completely cycle forage before its bowel is empty is approximately 48 hours (Alldredge and Reeder, 1972). This variable is important because most plutonium ingested by deer grazing in contaminated areas passes through the deer's gut, because of plutonium's low solubility and is redeposited to the ground in the form of fecal pellets (Whicker, 1979). Based on the amounts of soil transported off site by mule deer, the quantities of plutonium, americium, uranium-238, uranium-235 and uranium-233/234 transported off site were estimated using area-weighted average soil concentrations of these actinides.

**CHART 8**

**BIOLOGICAL PATHWAY ACTINIDE LOADS – ESTIMATED OFF-SITE TRANSPORT BY MULE DEER**



**BIOLOGICAL PATHWAY LOADS**  
 Estimates of actinide loads transported off site by mule deer, calculated using two different methods, are summarized in Chart 8.

**DISCUSSION** Estimates of plutonium activity transported off site by mule deer range from approximately 200 to 1,000 pCi per year. Areas most frequented at RFETS by mule deer are more heavily vegetated hillside grasslands, shrublands and woodlands (Kaiser-Hill, 2000). These areas provide greater erosion protection than sparsely vegetated areas and therefore limit indirect actinide movement caused by deer disturbing the soil. The limited erosion potential in heavily vegetated areas also reduces movement of deer pellets by erosion processes.

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# BIOLOGICAL PATHWAY



Site studies suggest there is limited redistribution of plutonium by biota in aquatic systems.

**DISCUSSION: TERRESTRIAL FAUNA** Plutonium is not a biologically essential element, nor does it serve as an analog for any other essential element (Higley and Whicker, 1999). There is little accumulation of plutonium in the tissues of arthropods, small mammals, snakes and mule deer. In general, biota investigations in the 903 Pad area showed that plutonium concentrations in biota were significantly lower than in soils. Arthropods and small mammals showed plutonium concentrations 100 times less than the concentrations in soil, with no significant differences in seven tissue types analyzed. The concentration hierarchy followed a downward trend from dead plant litter to fresh vegetation to animal compartments. Higher values for plant litter are expected since the litter is more closely associated with the surface soil and is prone to the accumulation of soil particulate matter. Generally, actinide sources in the environment have resulted in minor transfer of these elements into food webs, regardless of transport process.

**DISCUSSION: OTHER HIGHLY MOBILE SPECIES** Several other mobile species undoubtedly transport small quantities of actinides off site. Species such as waterfowl and other birds, coyotes and insects may transport actinides off site. However, data for these species are not available and would be difficult and in some cases logistically nearly impossible to obtain. Redistribution of contaminated soil by burrowing animals such as pocket gophers is a recognized phenomenon but is believed to only have a local effect on actinide redistribution (Whicker, 1979). Using the deer data and normalizing by the deer biomass, it is estimated that off-site transport by other selected terrestrial species is comparable to transport by deer, or possibly lower.



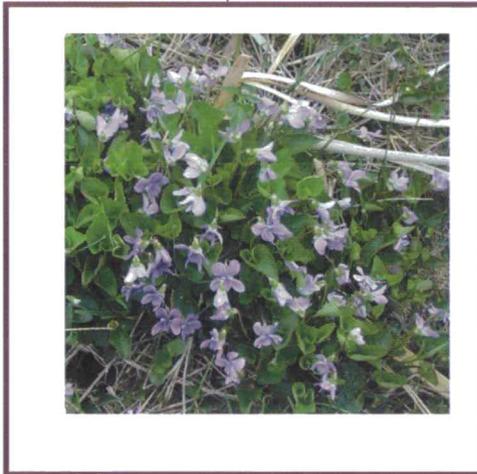
Studies conducted by CSU researchers show little accumulation of plutonium in animal tissues.

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**DISCUSSION: AQUATIC STUDIES** Limited aquatic studies at RFETS indicate a very limited potential for biota to redistribute plutonium in aquatic systems. Paine (1980) found an increase in trophic-level concentration of plutonium does not occur. There appears to be a selective mechanism, which discriminates against plutonium at the phytoplankton to zooplankton level. The highest concentration in crawfish was found in the exoskeleton. Whole fish had detectable activity, but fish flesh showed none. These results indicate low bioavailability of the plutonium because of its low solubility and chemical partitioning to solid particles.

**DISCUSSION: TERRESTRIAL VEGETATION** The uptake of plutonium into plant tissues is normally very minor because of its insoluble nature. The majority of plutonium measured in plant material is associated with surficial dust particles (Higley and Whicker, 1999).

**DISCUSSION: SOIL MICROBES** Microorganisms in soils, sediments and ponds may play a role in the regulation of actinide movement that occurs through surface soil erosion and colloidal transport processes. Potential interactions between indigenous microorganisms and actinides include bioreduction, bioprecipitation, biosorption and solubilization due to production of microbial metabolic products. Site-specific data on the microbial ecology of RFETS, however, do not exist, nor do studies detailing specific microbiological processes on actinide mobility in the surface soils, sub-surface material or surface water at the Site.



Plant tissues uptake very minor amounts of plutonium because of their insoluble nature.

# PATHWAY COMPARISON

**SUMMARY OF ACTINIDE LOADS** Estimates of average annual actinide loads transported off site by each of the major pathways addressed in this report are summarized and compared in this section. In cases where more than one method was used to estimate off-site loads for a specific pathway, the method yielding the highest estimated off-site load was used for the comparison. Because quantities of actinides transported off site vary by several orders of magnitude depending on the actinide and transport pathway, a logarithmic scale is used to display the results (Chart 9). Therefore, each horizontal line represents an actinide load that is larger, by a factor of 10, than the line below. Actinide transport pathways are compared by order of magnitude due to the uncertainties associated with analytical measurements and model estimation results.

## PATHWAY COMPARISON

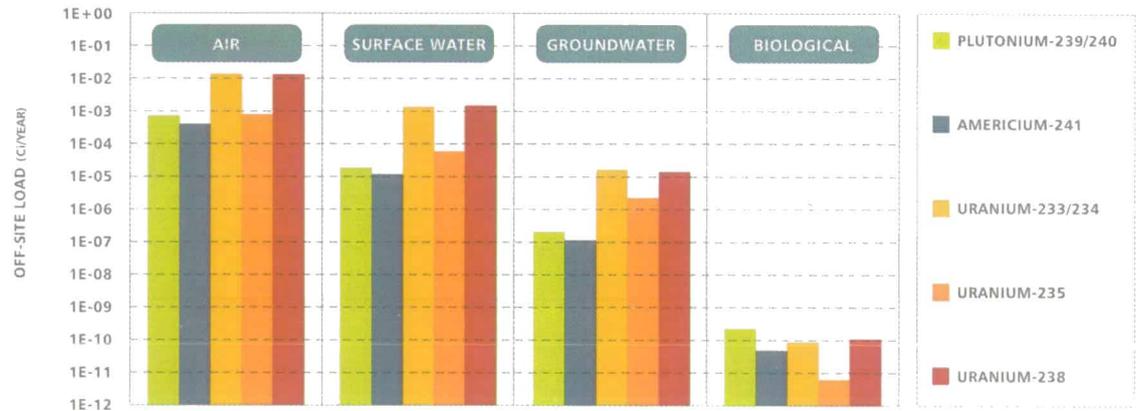
For all actinides, air and surface water are the dominant transport mechanisms. For plutonium, the estimated annual airborne load transported off site exceeds the surface water load by roughly a factor of 40. For americium, the trend of the results is the same, which is logical because both plutonium and americium are transported in a similar manner.

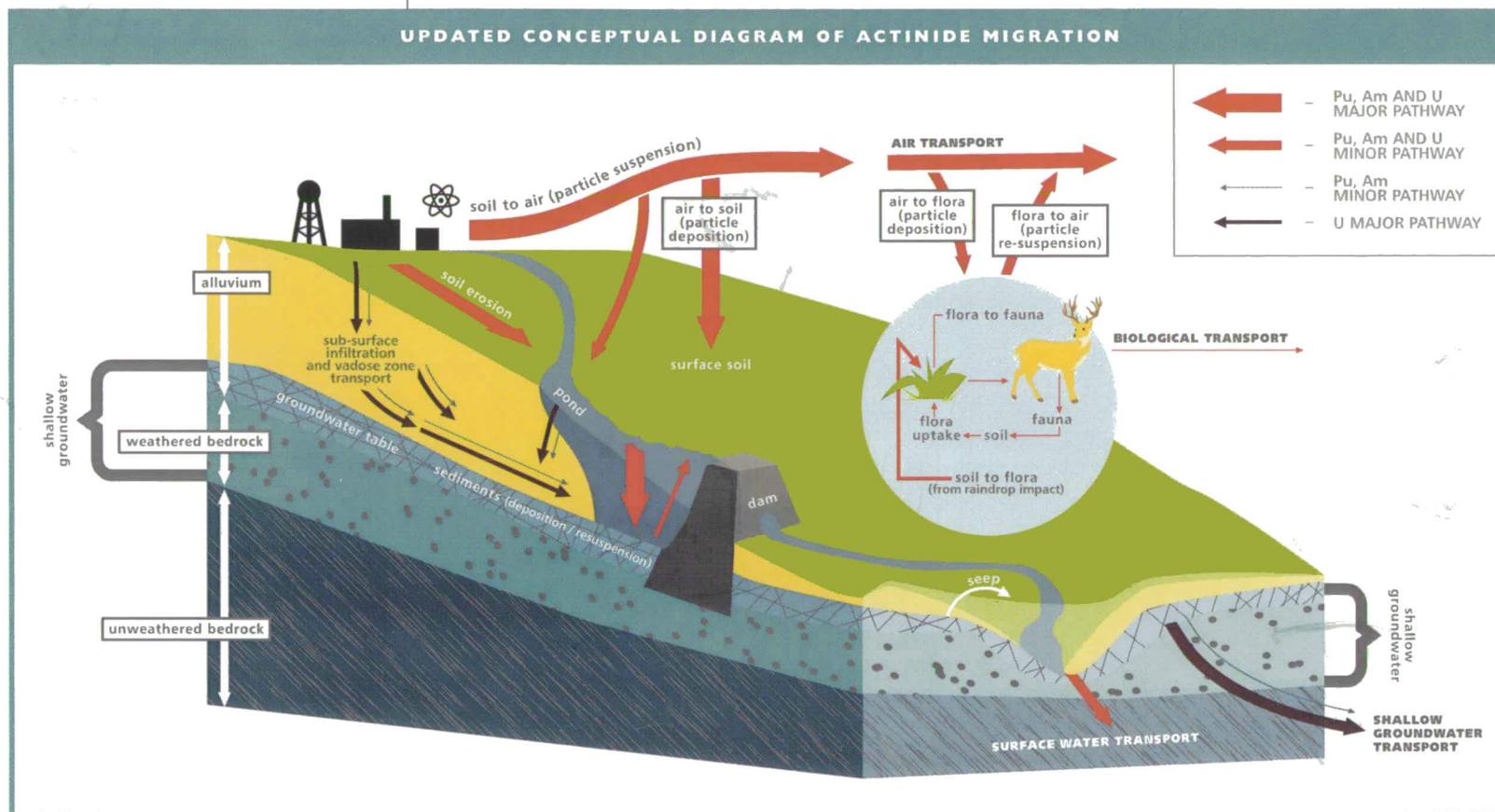
For shallow groundwater, estimated plutonium and americium loads are approximately two orders of magnitude less, or 1/100th, of the load conveyed in surface water. These shallow groundwater loads are, however, potentially biased high because of residual low-level surface soil contamination introduced down boreholes during drilling and well-installation operations. The ratio between surface water and groundwater in transporting loads of plutonium and americium off site is approximately the same as the ratio between volumes of surface water and shallow groundwater flowing off site.

The biological pathway is also minor relative to the air and surface water pathways. It is estimated to transport approximately five orders of magnitude less, or 1/100,000, of the plutonium load compared with the surface-water pathway.

CHART 9

ALL PATHWAYS – ESTIMATED OFF-SITE ANNUAL ACTINIDE LOADS





**TRANSPORT PROCESSES - PLUTONIUM AND AMERICIUM COMPARED WITH URANIUM**

Processes that transport plutonium and americium in the environment at RFETS are summarized in the diagram above. Larger arrows indicate more dominant pathways and smaller arrows indicate lesser pathways. The insoluble nature of plutonium and americium causes these actinides to be largely transported as particles attached to soil that is eroded by wind and water. Sub-surface transport of plutonium and americium is a relatively minor pathway, as is transport by biological mechanisms.

Uranium transport processes at RFETS are also shown above. Though not highly soluble, uranium is more soluble than plutonium and americium and is therefore more easily transported in the sub-surface. Hence, the arrows indicating a sub-surface pathway for uranium are larger than those for sub-surface plutonium or americium transport.

## PATHWAY SUMMARY & CONCLUSIONS

**AIR TRANSPORT PATHWAY** Transport of actinides through the air occurs largely by wind erosion of actinide-containing particulate matter from site soil and dust-laden vegetation. The general direction of airborne actinide transport follows the prevailing winds, from the northwest to the southeast. More importantly, higher winds, which transport much larger loads than lower winds, occur almost exclusively from the northwest quadrant.

For perspective on the quantity of airborne actinides measured at the Site, the air monitoring location with the highest total annual airborne actinide concentration from 1997 through 1999 was station S-140 in the southeast corner of the Site. This location had an airborne actinide level equal to approximately 1.4 percent of the 10 millirem regulatory standard governing airborne radionuclide concentrations at DOE facilities.

**SURFACE WATER TRANSPORT PATHWAY** The central Industrial Area, which drains to South Walnut Creek, yields the largest loads of plutonium and americium in surface water per square meter of drainage area. The Industrial Area has large impervious surfaces that generate large volumes of runoff during storms, which causes erosion and actinide loading in surface water. In contrast, the South Interceptor Ditch drainage has areas near the 903 Pad with the highest known levels of plutonium activity in soil, but the basin is largely well-vegetated and therefore generates less runoff that can cause erosion and transport actinides. The surface water plutonium load discharged per square meter of the South Interceptor Ditch basin (3.8 pCi/m<sup>2</sup>/year) is roughly one-tenth of the load per square meter of watershed compared to the central Industrial Area.

However, for extreme conditions, the South Interceptor Ditch may yield proportionately higher actinide loads. Model results indicate a hypothetical 100-year, 6-hour storm event (97.1 mm) would cause significant erosion in the South Interceptor Ditch basin and result in plutonium loads to the channel that are two to three orders of magnitude higher than observed in the Walnut Creek basin. Remediation of soils within the South Interceptor Ditch watershed will reduce actinide loads transported during extreme events.

The detention ponds on North and South Walnut Creeks serve to settle out particles and generally remove 80 to 90 percent of the annual plutonium and americium load that flows into the ponds. This corresponds with site research that demonstrates approximately 10 percent of the plutonium and americium in surface water is sorbed to colloid particles that are not likely to settle in the ponds. Another important observation regarding plutonium transport involves the lower section of Walnut Creek. The average annual plutonium load measured in Walnut Creek near the site boundary is approximately 30 percent greater than the plutonium load measured upstream, below the detention ponds. Site investigations indicate the plutonium source in this area is diffuse legacy contamination in soils and sediments.

Uranium activities are relatively uniform in surface water across the Site. As a result, the uranium load delivered from different basins is largely a function of each basin's water yield. Though surface water across the Site has uranium concentrations below the Maximum Contaminant Level for drinking water, high resolution analytical techniques are planned to determine if uranium from man-made sources is impacting site surface water.

**GROUNDWATER TRANSPORT PATHWAY** At RFETS, potential groundwater actinide transport involves lateral, shallow groundwater flow in the alluvium and weathered bedrock geologic units. Shallow groundwater at the Site does not percolate down into the regional Laramie-Fox Hills aquifer. A thick, intervening layer of impermeable claystones in the Laramie Formation prevents vertical movement from the shallow groundwater down to the regional aquifer.

Shallow groundwater and surface water are linked. Plutonium and americium are relatively immobile in the soil and groundwater because of their low solubility and tendency to sorb onto soil. However, work at RFETS as well as studies in the literature have shown that insoluble actinides can sorb to natural, sub-micrometer-sized colloid particles that can facilitate actinide movement. In addition to colloidal transport, sub-surface actinide transport can occur when more soluble actinides, such as uranium in the (VI) oxidation state, move in solution.

Low levels of plutonium and americium have been detected in shallow groundwater wells at the eastern site boundary. However, determination of plutonium and americium levels in shallow groundwater is complicated by residual surface soil contamination potentially introduced down boreholes during drilling and well installation. New clean or "aseptic wells" were drilled and efforts to improve sampling protocols are currently ongoing. For this analysis, plutonium and americium activity measured in shallow wells may represent activities higher than what actually exists in the shallow groundwater.

Uranium-233/234 and uranium-238 isotopes are the dominant actinides found in shallow groundwater in terms of total activity because of their natural abundance. Uranium in RFETS shallow groundwater is generally within the range of uranium detected naturally. Data from high-resolution ICP/MS analyses indicate that uranium in most areas of the Site is from natural sources. However, shallow groundwater samples at the site boundary in the Walnut and Woman Creek groundwater basins have a uranium-235/uranium-238 ratio that is slightly less than found naturally. Though potentially related to analytical uncertainty, these results indicate alluvial groundwater in these basins potentially has a signature indicating a small fraction of the uranium is "depleted" uranium.

**BIOLOGICAL TRANSPORT PATHWAY** RFETS-specific studies and other scientific literature indicate that plutonium has low bioavailability, due to its insolubility. Consequently, uptake into plant and animal tissues is minor. There is little accumulation of plutonium in the tissues of arthropods, small mammals, snakes or mule deer.

Mule deer have been studied as a biological pathway for actinide movement because of their mobility, amount of soil intake and size of the herd. Based on the estimated plutonium inventory in soil and data on deer mobility, the plutonium activity transported off site by deer movement is estimated to be approximately  $2 \times 10^{-10}$  to  $1 \times 10^{-9}$  Ci annually.

**CONCLUSIONS** Quantified analyses of RFETS actinide pathways generally support the conceptual model which identified soil and sediment transport processes as the primary mechanisms for plutonium and americium transport. Measured and modeled data confirm that wind and water erosion are the dominant plutonium and americium transport pathways, though the magnitude of airborne transport is larger than previously suggested in the qualitative conceptual model study.

Modeled data also support the conceptual model in terms of shallow groundwater transport being a relatively minor pathway for plutonium and americium because of the low solubility and strong soil sorption characteristics of these actinides. Data also support the conceptual model regarding the importance of sub-surface uranium transport, due to its higher solubility. Analyses indicate most of the uranium in shallow groundwater is from natural sources. Uranium loads transported off site in shallow groundwater are small compared to surface water. However, discharges of shallow groundwater to the surface contribute a major fraction of the surface water uranium load in specific stream channels.

## IMPLICATIONS FOR SITE CLOSURE

An objective of the Pathway Analysis Report is to provide recommendations for long-term protection of the environment, with emphasis on actinide surface water quality, during and after site closure, as specified in the Rocky Flats Cleanup Agreement. Based on the characterization of current actinide sources and quantitative analysis of actinide transport mechanisms, the following general implications apply to near-term site remediation, final site closure design and long-term site management and stewardship.

**NEAR-TERM SITE REMEDIATION** Field measurements and modeling analyses indicate air and surface water are the major transport pathways for plutonium and americium. Soil disturbance increases the potential for soil erosion and contaminant transport. For example, Environmental Protection Agency (EPA) emissions factors indicate heavy construction equipment activities can increase airborne particulate emissions by roughly a factor of 20. Plutonium and americium in surface soil east of the 903 Pad is evidence of widespread contamination believed to have been dispersed when disturbed soils were exposed to a few high wind events in the 1960s. Current understanding of transport processes combined with historic lessons reinforce the importance of implementing soil erosion controls, such as protecting soil stockpiles and limiting excavation on windy days, to minimize airborne actinide transport during remedial activities.

Similarly, soil erosion and transport by surface water is a major potential pathway for plutonium and americium movement. Appropriate erosion control measures should be implemented during site remediation, including techniques such as minimizing vegetation disturbance and redirecting runoff away from excavations. A surface water management and detention pond system, with the capacity to settle out plutonium and americium, should be maintained during active site remediation.

**Minimizing soil erosion by wind and water is a key concept for controlling actinide movement during short-term remediation activities and for long-term Site management.**



Groundwater is not a major pathway for plutonium and americium transport, but operation and maintenance of the existing groundwater treatment systems will protect surface water from potential sub-surface uranium transport. The biological pathway is a minor transport mechanism for actinides and does not require altered management during site remediation other than excluding wildlife from active remediation sites.

**FINAL CLOSURE DESIGN** When site remediation is complete, surficial actinide sources with the highest activities are likely to have been removed. These remedial actions will reduce the reservoir of available actinides and diminish the magnitude of airborne actinide transport from these areas.

Removal of large impervious surfaces from the Industrial Area will result in reduced surface water runoff with a corresponding reduction in soil erosion and actinide transport. The combination of reduced runoff and diminished actinide sources will reduce the actinide load transported by the surface water pathway. In addition to remediation of localized actinide sources, other diffuse, low-level actinide sources that contribute to surface water contaminant loads, as observed in lower Walnut Creek, should be managed as needed for long-term protection of surface water quality.

Minimizing wind and water erosion should remain as a central theme in the final site closure design, with attention given to the long-term functionality of erosion control features. In addition to general erosion protection measures, such as establishing a vegetation cover resistant to drought or other extreme ecological conditions, location-specific controls for surface water erosion should be considered for the final site configuration. Such measures include: (1) re-contoured or terraced slopes; (2) re-routed runoff; and (3) a surface water detention system with the capacity to entrap and settle particles that transport plutonium and americium.

Groundwater is a minor pathway for plutonium and americium, but can be an important transport pathway for uranium. Remediation of man-made uranium sources that impact surface water should provide long-term protection of surface water quality.

Biological mechanisms also have a minor direct influence on actinide movement, but they can indirectly influence actinide transport by causing soil disturbance that promotes erosion with resulting air and surface water actinide transport. Therefore, the final closure configuration design should minimize potential erosion effects caused by animals burrowing or otherwise disturbing the soil in parts of the Industrial Area with residual contamination.

**LONG-TERM MANAGEMENT** After final site closure, efforts to reduce soil erosion caused by wind and water should be continued by minimizing soil disturbance and maintaining stable slopes, particularly in areas with residual actinide activity. This approach includes using appropriate controls for managing biological resources and human impacts after the Site is converted into a National Wildlife Refuge. If post-closure monitoring identifies residual actinide activity that impacts surface water quality, the best available technology should be used to appropriately characterize and mitigate the actinide source.

## FURTHER READING

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**FURTHER READING** (continued)

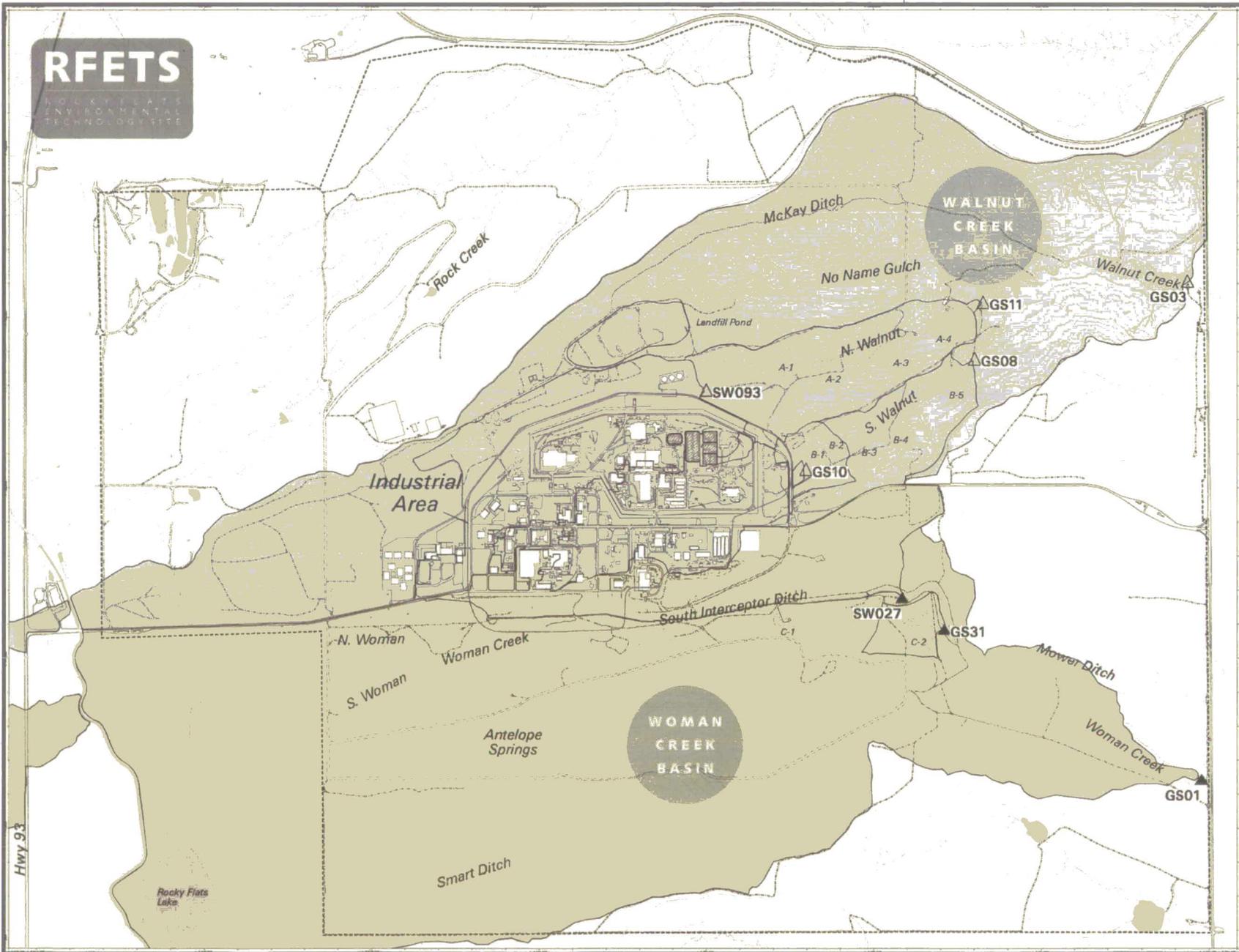
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