

**PARTICULATE, COLLOIDAL, AND DISSOLVED-PHASE  
ASSOCIATIONS OF PLUTONIUM, AMERICIUM, AND  
URANIUM IN WATER SAMPLES FROM WELL 1587 AND  
SURFACE-WATER SITES SW-51 AND SW-53 AT THE  
ROCKY FLATS PLANT, COLORADO**

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and W.H. Orem**

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# Particulate, Colloidal, and Dissolved-Phase Associations of Plutonium, Americium, and Uranium in Water Samples from Well 1587 and Surface-Water Sites SW-51 and SW-53 at the Rocky Flats Plant, Colorado

By R A. Harnish, D M. McKnight, J F Ranville, V C Stephens, and W H Orem

## Abstract

The potential for off-site transport of plutonium in ground or surface water has been a concern in the remediation efforts at the Rocky Flats nuclear facility near Denver, Colorado. The transport and fate of dissolved radionuclide species are inherently different from those for particulate or colloidal phases involving radionuclides. To distinguish among dissolved, colloidal, and particulate phases, water samples were collected and were filtered in series using spiral-flow filtration and tangential-flow ultrafiltration. One of the surface-water samples had a high content of larger particles, in this sample, plutonium and americium were distributed mainly in the particulate phase. Uranium was distributed mainly in the dissolved phase. For the other surface-water sample and the ground-water sample, the colloidal fraction also was important in plutonium and americium distribution. The filtrates from the 10,000-Dalton ultrafilter were fractionated, using ion-exchange resins and XAD-8 resin for isolation of fulvic acids. Most of the dissolved plutonium was associated with the fulvic acid for all three samples. A portion of the americium and uranium activity (5 percent to 21 percent) was associated with fulvic acid, but most americium and uranium were present in an anionic form not associated with fulvic acid, indicating the possible importance of other organic ligands or inorganic anionic species.

## INTRODUCTION

The U.S. Department of Energy administers the operation of the Rocky Flats Plant, a government-owned, contractor-operated facility for nuclear-material processing located in northern Jefferson County, Colorado, about 26 km northwest of downtown Denver. The plant opened in 1952, and plutonium (Pu) processing continued until 1990, when the principal mission of the plant changed from manufacture and processing of plutonium components to pollutant identification and remediation. Pursuant to the U.S. Department of Energy's environmental restoration program for the Rocky Flats Plant, and in cooperation with the U.S. Department of Energy, in 1991, the U.S. Geological Survey (USGS) began a study of the potential for facilitated transport of Pu, americium (Am), and uranium (U) by colloidal-size particles at two surface-water sites and one ground-water site at the plant. This study was designed to determine the predominant phases that control the transport of Pu, Am, and U in surface and ground water at the Rocky Flats Plant and, in particular, the potential for transport of these elements as colloids or as complexes with humic substances. The results of an initial sampling of the water from Well 1587 in November 1991 are described in a previous report by Harnish and others (1994). In summary, the conclusions in that study about actinide distribution were that:

1. About 65 percent of Pu 239 and 240 was associated with particulate and colloidal phases
2. The particulate fraction (greater than 5  $\mu\text{m}$ ) was rich in iron oxyhydroxides, in smaller colloids, clay minerals and organic material were important
3. Copper and zinc were present in the smallest colloid fraction.

The potential exists for formation of complexes involving actinides and carboxylic-acid groups in naturally occurring fulvic acid. Plutonium, for example, forms complexes with organic carboxylates, and complexing agents, such as NTA and EDTA, which are used in some decontamination activities. Because of the heterogeneity in fulvic acids and the importance of ligand-to-metal ratios in controlling complexation, studies of these interactions in an actual environmental setting are potentially more definitive than laboratory studies

*Were they used here?*

## Geohydrologic Setting

The Rocky Flats Plant is located east of the Front Range of the Rocky Mountains on a broad, east-sloping plain of alluvial fans between the stream-cut valleys of North Walnut Creek and Woman Creek (figs 1 and 2). Details of the geologic and hydrologic characteristics of the area within the plant boundaries have been described in previous reports (Rockwell International, 1987, EG&G, 1991, Harnish and others, 1994). Well 1587 is a shallow alluvial monitoring well in the Woman Creek Basin, about 90 m east of the 903 Pad (fig 2). The well is cased with PVC pipe 10 cm in diameter, extends about 6.9 meters through the alluvium to bedrock, and is screened through the lower 4.9 m of alluvium. The seep at site SW-51 originates about 180 m east-southeast of the 903 Pad (fig 2), then becomes a shallow stream that empties into a drainage ditch. The surface-water seep at site SW-51 is formed where ground water enters permeable formations on a slope and is ephemeral in response to rainfall and infiltration in the vicinity. The seep at site SW-53 is a marshy area fed by surface water in a depression about 275 m east-southeast of the 903 Pad area. All three sampling locations are downgradient from the 903 Pad area, a potential source of actinide contamination. Physical and chemical measurements (table 1) indicate that these are well buffered, slightly alkaline, calcium/magnesium bicarbonate type waters.

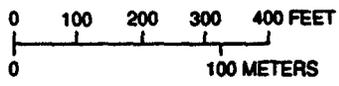
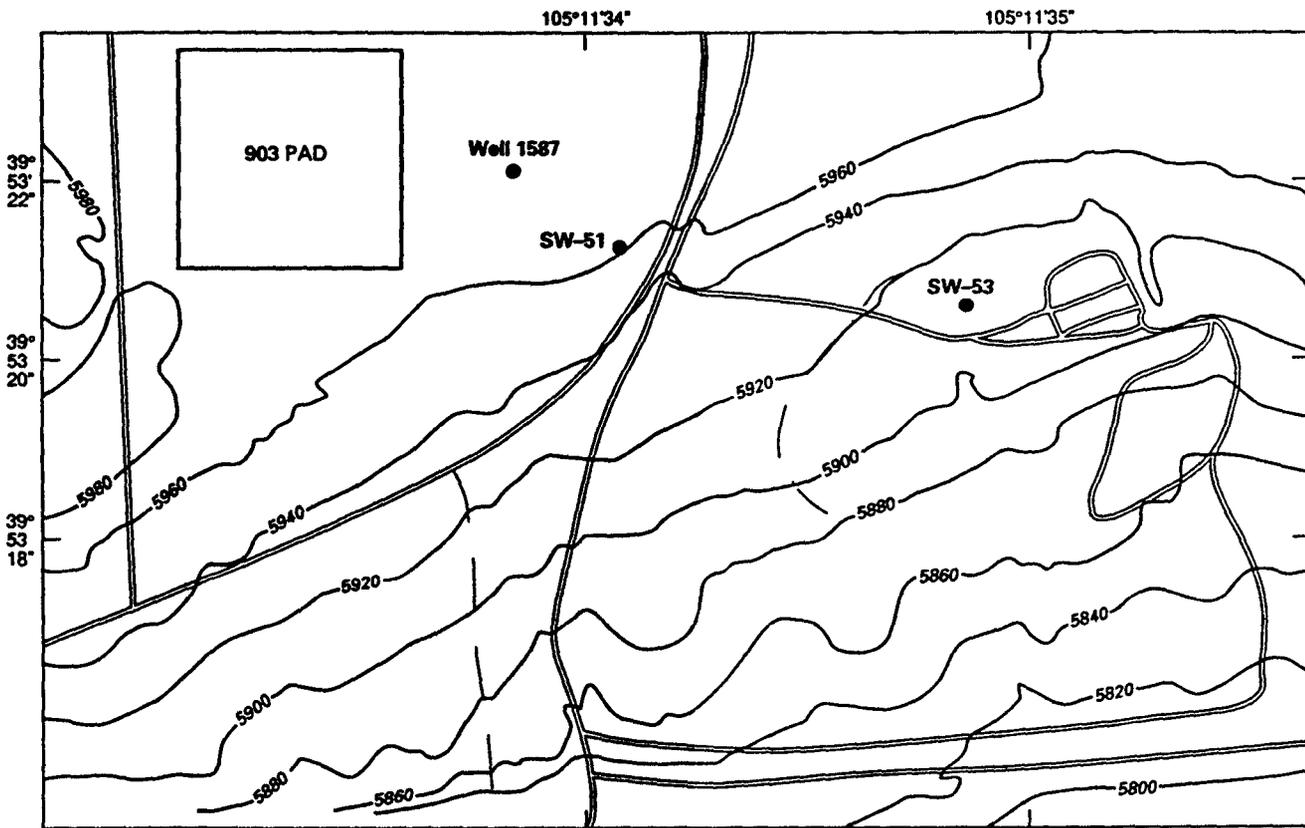
## Terminology Used in this Report

Filtration in environmental sampling traditionally has used 0.45- $\mu$ m filters for separating the particulate phase from the dissolved phase. This choice of filter size is arbitrary because size distributions for environmental particles in water form a continuum from less than 1 nm to hundreds of micrometers (Buffle and others, 1989, 1992) (table 2). For the present study, five filter pore sizes were used for particle isolation and fractionation, and the following terminology is used to distinguish among the fractions:

- 1 The term "particle" refers to all phases of a sample that are not dissolved and includes colloids and particulates.
- 2 The term "particulate" refers to the phase that is retained by a 5.0- $\mu$ m filter (used for samples from Well 1587 and SW-51) or a 3.0- $\mu$ m filter (used for the sample from 787SW-53).
- 3 The term "colloid" includes three distinct particle fractions.
  - a. Particles that pass through a 5.0- $\mu$ m (or 3.0- $\mu$ m) filter, but are retained on a 0.45- $\mu$ m filter
  - b. Particles that pass through a 0.45- $\mu$ m filter, but are retained on a 100,000-Dalton filter
  - c. Particles that pass through a 100,000-Dalton filter, but are retained on a 10,000-Dalton filter

A 100,000-Dalton filter corresponds approximately to a 5- to 8-nm (0.005- to 0.008- $\mu$ m) filter. A 10,000-Dalton filter corresponds approximately to a 1-nm (0.001- $\mu$ m) filter.

- 4 The term "dissolved" denotes the phase that passes through a 10,000-Dalton filter.



**EXPLANATION**  
 — 5920 — Topographic contour in feet  
 Datum is sea level  
 SW-51 ● Sampling location

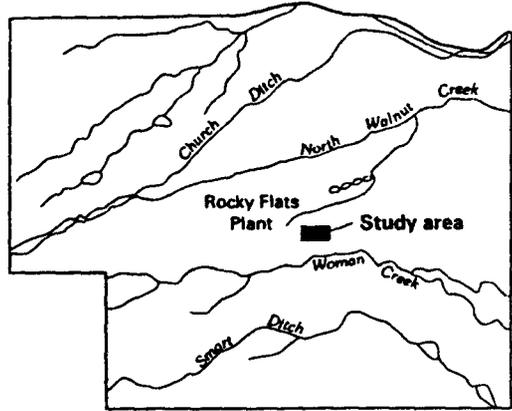


Figure 2. Locations of Well 1587, site SW-51, and site SW-53 at the Rocky Flats Plant.

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**Table 2.** Classification and nominal physical characteristics of suspended material, adapted from Buffle and others, 1992

[ $\mu\text{m}$ , micrometer; >, greater than, cm/s, centimeter per second, km/yr, kilometer per year; <, less than, -, not applicable]

Phase	Diameter ( $\mu\text{m}$ )	Molecular weight (Dalton)	Sedimentation rate
Particulate	62-2,000	$>10^8$	0.4-20 cm/s
Particulate	5-62	$>10^8$	0.5-124 km/yr
Colloid	2-5	$>10^8$	0.1-0.5 km/yr
Colloid	1	$10^8$	$<0.1$ km/yr
Colloid	0.45	$10^7$	$<0.1$ km/yr
Colloid	0.1	$10^6$	$<0.1$ km/yr
Colloid	0.001	$10^4$	$<0.1$ km/yr
Dissolved	$<0.001$	$<10^4$	-

- In addition, the following terms refer to filtration fractions "filtrate" is the water and compounds of any size that pass through a filtration membrane, and "retentate" is a material of any size that is retained by a filtration membrane
- The term "fulvic acid" is operationally defined as dissolved organic acid isolated on XAD-8 resin under conditions described by Thurman and Malcolm, (1981)

## Acknowledgments

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## METHODS

### Sample Collection

The task of isolating colloids from natural water is complicated by the unstable nature of these particles and their low concentrations. Ideally, the isolated particles are unaltered from their natural state in terms of size, morphology, charge characteristics, surface area, chemical composition, and sorbed species. A practical goal is a minimal perturbation during the sampling and processing of the particles. This need for minimal perturbation necessitates sample acquisition and filtration protocols that minimize physical disruption of the particles and that avoid physicochemical processes, such as coagulation, that alter colloid characteristics (Ryan, 1988). Exposure to the atmosphere, exposure to light, and departures from in-situ water temperature can profoundly alter the physicochemical characteristics of the water sample. These considerations are especially critical when sampling ground

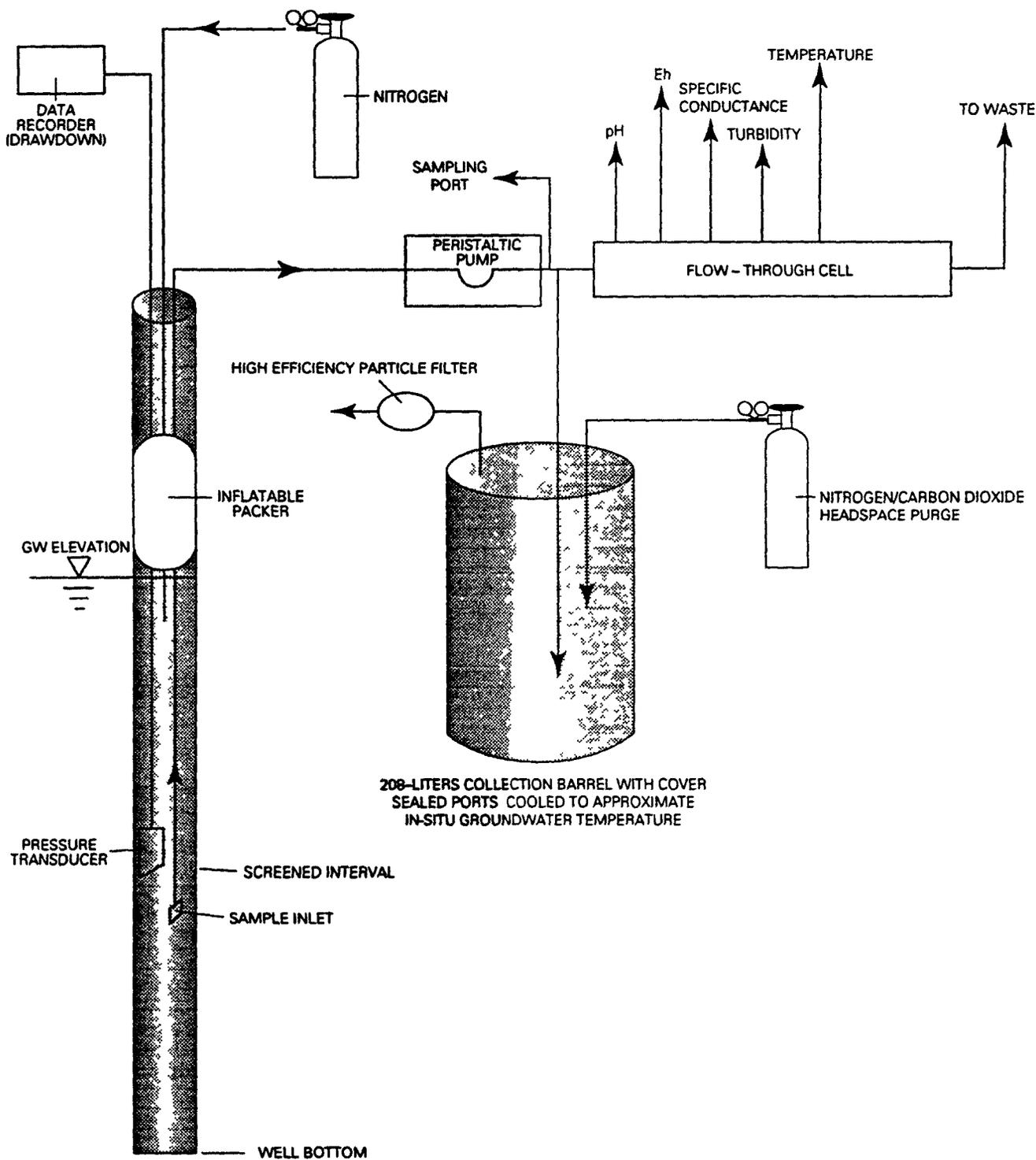


Figure 3 Sampling setup for collection of ground water from Well 1587 at the Rocky Flats Plant.

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The filtration rate ranged from 400 to 500 mL/min for the 5.0- and 3.0- $\mu\text{m}$  filtration to 100 to 300 mL/min for the 0.45- $\mu\text{m}$ , 100,000- and 10,000-Dalton filtrations. The filtration sequence is shown in figure 5. The unfiltered sample was first filtered by spiral-flow filtration through a 5.0- $\mu\text{m}$  polycarbonate filter for Well 1587 and seep SW-51 and 3.0- $\mu\text{m}$  glass-fiber filter for seep SW-53. Particles greater than 5  $\mu\text{m}$  were recovered from the 5.0- $\mu\text{m}$  filters by placing the filters in a petri dish and gently rinsing the filter surface. Retentates thus obtained were refrigerated at 4°C. The 3.0- $\mu\text{m}$ -depth filtration of the sample from SW-53 precluded recovery of the greater than 3.0- $\mu\text{m}$  particles by rinsing, and analysis of the retentate for radionuclides at this site was accomplished by acid leaching of the filter.

The filtrate from the 5.0- $\mu\text{m}$  (or 3.0- $\mu\text{m}$ ) filtration then was filtered through several 0.45- $\mu\text{m}$  polyvinylidene fluoride filters by tangential filtration, producing a 0.45- $\mu\text{m}$  filtrate, a 0.45- $\mu\text{m}$  concentrated retentate, and particles adhering to the filters. Following filtration, the filtration systems were disassembled, and adhering colloids were removed from the filters by placing a filter in a Teflon bag along with some retentate and gently rubbing off the colloids, this process was repeated for all filters of each size range. The collected colloids were combined with the retentate from the fraction and were refrigerated at 4°C. At each step of the filtration, samples were taken of filtrates and retentates for radiochemical and chemical analysis. Two subsequent tangential-filtration steps (100,000 and 10,000 Dalton) were done in the same manner for the isolation of colloids. Filtration protocols are listed in table 3.

The final 10,000-Dalton filtrate was processed through a series of XAD-8, anion-exchange, and cation-exchange resins. The resin XAD-8 is a nonionic macroporous sorbent that has been used to isolate fulvic acids from aquatic systems (Thurman and Malcolm, 1981). Methods for the isolation of aquatic fulvic acids have been reviewed by Aiken (1985). Protocols for resin extraction used in this study are listed in table 4. Parts of the 10,000-Dalton filtrate, acidified to pH 1.8, were passed through a column of XAD-8. The fulvic acids retained on the resin were eluted in 0.1 N NaOH and then acidified. The acidified eluates were combined and reconcentrated on the XAD-8 column. The final eluate was hydrogen saturated on a cation-exchange resin, and the fulvic acid was lyophilized for analysis.

The eluate from the XAD-8 extraction was adjusted to its original pH (7.66 for Well 1587 water and 7.76 and 7.57 for SW-51 and SW-53 surface water) using NaOH, in preparation for subsequent extraction by anion- and cation-exchange resins. One-hundred liters of post-XAD eluant from each site was extracted sequentially in a batch mode using 80 grams of Biorad AG1-X8 anion-exchange resin (100–200 mesh, chloride form) and then with 80 grams of Biorad AG50W-X8 cation-exchange resin (100–200 mesh, hydrogen form). The resins were subsequently ashed and submitted for radionuclide analysis.

Another 100-L part of the 10,000-Dalton filtrate from each site, which had not been previously extracted with XAD-8, was extracted sequentially by the same anion- and cation-resin treatment protocol. Because fulvic acids contain carboxylic-acid groups that are negatively charged at the neutral pH of the water samples, the fulvic acids also are removed from solution by an anion-exchange resin. The resins were then ashed and submitted for analysis of radionuclide activity.

Laboratory-grade deionized water was filtered by the same filtration protocol as the field water samples and then analyzed at each filtration step for Pu 239 and 240, Am 241, and U 233 and 234 activity. The filters were digested and analyzed for the same radionuclides. The 10,000-Dalton filtrate then was passed through XAD-8, cation-, and anion-exchange resins, eluates and the resins were similarly analyzed for radionuclide activity. In no case did detected activities of Pu 239 and 240, Am 241, or U 233 and 234 substantially exceed detection limits (tables 5–7), confirming that the filtration equipment and protocol did not substantially affect the reported activities of these actinides.

**Table 3. Protocols used for filtration of water from Well 1587 and surface-water sites SW-51 and SW-53**

[ $\mu\text{m}$ , micrometers,  $\text{cm}^3$ , cubic centimeters, L, liters, --, no data]

Filter pore size	Filter type	Filter material	Number of filters used	Filter area ( $\text{cm}^2$ )	Volume filtered (L)	Retentate volume (L)
Well 1587						
5.0 $\mu\text{m}$	Poretics	Polycarbonate	35	2,240	833	0.34
0.45 $\mu\text{m}$	Millipore	Polyvinylidene fluoride	8	3,715	828	0.42
100,000 Daltons	Millipore	Polyethersulfone	8	3,715	825	--
10,000 Daltons	Millipore	Cellulose	8	3,715	820	0.80
SW-51						
5.0 $\mu\text{m}$	Poretics	Polycarbonate	14	896	300	0.30
0.45 $\mu\text{m}$	Millipore	Polyvinylidene fluoride	8	3,715	296	0.32
100,000 Daltons	Millipore	Polyethersulfone	8	3,715	292	0.52
10,000 Daltons	Millipore	Cellulose	8	3,715	287	0.62
SW-53						
3.0 $\mu\text{m}$	Balston	Glass fiber	1	--	120	--
0.45 $\mu\text{m}$	Millipore	Polyvinylidene fluoride	6	2,786	116	1.36
100,000 Daltons	Millipore	Polyethersulfone	6	2,786	112	1.08
10,000 Daltons	Millipore	Cellulose	6	2,786	108	0.95

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yield  
trace?

Table 5. Radiochemical analysis of field blanks plutonium 239 and 240

[ $\mu\text{m}$ , micrometers; pCi/L, picocuries per liter; pCi/sample, picocuries per sample; pCi/gram, picocuries per gram]

Sample	Result	Units	Analytical Error	Detection Limit
<b>Water blanks</b>				
Unfiltered deionized water	0 001	pCi/L	0 003	0 005
5 0- $\mu\text{m}$ filtered deionized water	0 004	pCi/L	0 004	0 005
0 45- $\mu\text{m}$ filtered deionized water	0 006	pCi/L	0 004	0 007
100,000-Dalton filtered deionized water	0 001	pCi/L	0 003	0 005
10,000-Dalton filtered deionized water	0 006	pCi/L	0 005	0 008
Post-XAD-8 10,000-Dalton filtered deionized water	0 002	pCi/L	0 004	0 005
Post anion-exchange resin 10,000-Dalton filtered deionized water	0 004	pCi/L	0 005	0 006
Post cation-exchange resin 10,000-Dalton filtered deionized water	0 001	pCi/L	0 003	0 006
<b>Filter blanks</b>				
5 0- $\mu\text{m}$ filter (new)	0 005	pCi/sample	0 013	0 025
0 45- $\mu\text{m}$ filter (new)	0 006	pCi/sample	0 008	0 008
100,000-Dalton filter (new)	0 003	pCi/sample	0 005	0 007
10,000-Dalton filter (new)	0 005	pCi/sample	0 010	0 018
<b>Resin blanks</b>				
XAD-8 resin (new)	0	pCi/gram	0 05	0 08
Anion-exchange resin (new)	0 001	pCi/gram	0 006	0 005
Cation-exchange resin (new)	0 001	pCi/gram	0 004	0 004

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**Table 7. Radiochemical analysis of field blanks uranium 233 and 234**

[ $\mu\text{m}$ , micrometers; pCi/L, picocuries per liter; pCi/sample, picocuries per sample, pCi/gram, picocuries per gram]

Sample	Result	Units	Analytical Error	Detection Limit
<b>Water blanks</b>				
Unfiltered deionized water	0.22	pCi/L	0.19	0.22
5.0- $\mu\text{m}$ filtered deionized water	0.088	pCi/L	0.13	0.16
0.45- $\mu\text{m}$ filtered deionized water	0.27	pCi/L	0.23	0.30
100,000-Dalton filtered deionized water	0.31	pCi/L	0.24	0.36
10,000-Dalton filtered deionized water	0.23	pCi/L	0.21	0.25
Post-XAD-8 10,000-Dalton filtered deionized water	0.12	pCi/L	0.20	0.22
Post anion-exchange resin 10,000-Dalton filtered deionized water	0.18	pCi/L	0.19	0.26
Post cation-exchange resin 10,000-Dalton filtered deionized water	0.15	pCi/L	0.25	0.20
<b>Filter blanks</b>				
5.0- $\mu\text{m}$ filter (new)	0.022	pCi/sample	0.035	0.050
0.45- $\mu\text{m}$ filter (new)	0.007	pCi/sample	0.026	0.059
100,000-Dalton filter (new)	0.004	pCi/sample	0.024	0.064
10,000-Dalton filter (new)	-0.004	pCi/sample	0.004	0.053
<b>Resin blanks</b>				
XAD-8 resin (new)	0	pCi/gram	0.03	0.12
Anion-exchange resin (new)	0.003	pCi/gram	0.003	0.01
Cation-exchange resin (new)	0.01	pCi/gram	0.04	0.08

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**Table 8. Detection limits for inductively coupled plasma/atomic-emission spectrometry analysis**

<b>Analyte</b>	<b>Detection limit (micrograms per liter)</b>
Aluminum	50
Arsenic	2
Barium	20
Calcium	1,000
Iron	20
Lithium	10
Magnesium	1,000
Manganese	10
Potassium	1,000
Silicon	100
Sodium	1,000
Strontium	5
Vanadium	10
Zinc	10

**Table 9. Detection limits for anion analyses**

<b>Analyte</b>	<b>Detection limit (milligrams per liter)</b>
Bicarbonate	5
Carbonate	5
Fluoride	0.5
Chloride	3
Sulfate	5
Nitrate and nitrite	0.1

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**Table 11. Measurements of pH made during the course of ultrafiltration of water from surface-water sites SW-51 and SW-53**

[ $\mu\text{m}$ , micrometer; pH, in standard units SW-51 sample total processing time = 16 hours, SW-53 sample total processing time = 9 hours]

Elapsed time	Unfiltered	5.0- $\mu\text{m}$ filtrate <sup>1</sup>	0.45- $\mu\text{m}$ filtrate	100,000-Dalton filtrate	10,000-Dalton filtrate
pH in SW-51 sample					
1 hour	7.73	7.74	7.75	7.75	7.74
7 hours	7.75	7.73	7.76	7.74	7.76
14 hours	7.73	7.74	7.74	7.72	7.73
pH in SW-53 sample					
1 hour	7.54	7.56	7.53	7.54	7.53
4 hours	7.58	7.54	7.55	7.54	7.52
8 hours	7.54	7.55	7.54	7.55	7.53

<sup>1</sup> 0- $\mu\text{m}$  filtrate for SW-53

Scanning electron microscope (SEM) examination of retentates from the surface-water samples, however, indicated some evidence of post-collection aggregation, although to a lesser extent than the Well 1587 sample. The processes of retentate concentration and preparation of the sample for SEM analysis can affect apparent particle sizes and argues for in-line filtration for the acquisition of SEM samples.

### Particle Recovery

Three considerations are critical to assessing the potential of particle-facilitated transport of radionuclides in natural water. First, the particles must be present in sufficient quantity to have a significant effect on transport. Second, the particles must be reactive with the contaminant of concern. Third, the particles must be mobile under the physiochemical conditions of the flow regime. The first consideration, detailing particle recovery from water from the three sites, is included in table 12. For Well 1587, a total of 833 L of water was collected and filtered. By summing the particle concentration of each size fraction, the particle concentration of the whole-water sample was calculated to be 1.32 mg/L (table 12). Nine percent of the total particle mass was recovered in the particulate phase, 91 percent of the total particle mass was present in the three colloidal phases. For seep SW-51, 300 L of water was collected and filtered. The particle concentration of the whole-water sample was about 3 mg/L (table 12). Eighteen percent of the total particle mass was recovered in the particulate phase, about 82 percent of the total particle mass was present in the three colloidal phases.

For SW-53, 120 L of water was collected and filtered. The particle concentration of the whole water sample was calculated to be about 100 mg/L. Eighty-two percent of the total particle mass was recovered in the particulate phase; 18 percent of the particle mass was present in the three colloidal phases.

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The particle-recovery data indicated the disparate aquatic environments from which the samples were collected. The sample from Well 1587 was typical of most ground water in that the concentration of particulate species was low. The flowing surface water at site SW-51, which was in contact with bed sediments and terrestrial vegetation, carried about triple the particulate and colloid concentration of the ground water. The relatively slow flowing water of the wetland environment at site SW-53, which had extensive aquatic-plant populations, contained particulate concentrations about two orders of magnitude greater than the ground water. In the low-particulate water of Well 1587 and site SW-51, colloids were the predominant phase. In the wetland water at site SW-53, colloids seem to compose a smaller percentage of the total particle concentration, although particles of 3 to 5  $\mu\text{m}$  were defined as particulates in this sample, and therefore created a bias toward underestimation of colloid concentrations in this sample. Colloids existed at much higher concentrations in water from SW-53 than from Well 1587 and SW-51. Therefore, at all of the sites, colloids existed in potentially sufficient quantity to facilitate the transport of radionuclides.

### Inorganic Chemistry

The results of the analysis of the unfiltered and filtered water samples for inorganic species are listed in table 13. These samples are enriched in Ca and Mg, compared to Na, which indicates the geologic setting. The higher Ca concentrations are consistent with the measurement of calcite in the particle fractions in the initial study (Harnish and others, 1994). For all the major cations, the dissolved species were the predominant form. This result is similar to the result from the initial study (Harnish and others, 1994).

The dissolved inorganic species detected in the unfiltered samples from SW-53 are listed in table 13. For SW-51, only Al, Fe, and Ba had concentrations greater than the detection limit and, for Well 1587, only Al and Ba were detected in the unfiltered sample. This pattern can be explained by the much greater abundance of particles in the water sample from SW-53. The Al concentration detected in the sample from Well 1587 was essentially unchanged by filtration, indicating that the Al was truly dissolved.

For the sample from SW-51, the Al and Fe concentrations were decreased substantially by the first filtration through the 50- $\mu\text{m}$  filter (table 13). This result indicated that the particulates from these two sites may contain hydrous Al and Fe oxides. Such oxides would have surfaces with the capability to sorb radionuclides.

The association of trace metals with the particulate and colloidal fractions was more complicated for the sample from SW-53. Aluminum and the trace metals that had low concentrations in the unfiltered samples (Zn, As, and V) were removed by the first (3- $\mu\text{m}$ ) filtration step (table 13). The decreases in Al and Fe concentrations at the first filtrate were greater than tenfold, but the decrease in Mn was only about fourfold. This result indicated that the SW-53 particulates were possibly enriched in hydrous Al, Fe, and Mn oxides.

Measurable concentrations of Fe were in the 3- $\mu\text{m}$  filtrate (table 13), but the next filtration step decreased concentrations to undetectable values. In contrast, dissolved Mn was not decreased to undetectable levels even by the final 10,000-Dalton filtration step. These results indicated that the colloids between 3 and 0.45  $\mu\text{m}$  may contain hydrous Fe oxides and that the colloid fractions likely did not contain substantial concentrations of Mn oxides. It is not known why Mn concentrations increased in the 100,000-Dalton and 10,000-Dalton filtrates at site SW-53.

## Radiochemical Analyses

### Filtrate Analysis

Results of radiochemical analyses of unfiltered samples and filtrate samples of ground water from Well 1587 and surface water from sites SW-51 and SW-53 are listed in table 14 and are reported in picocuries per liter. Plutonium 239 and 240 were detected in all unfiltered samples and filtrate samples from all three sites, and the highest activity concentrations were detected in the unfiltered samples (table 14). Total (unfiltered) concentrations were highest in the surface-water samples from SW-51 and SW-53 (about 1.45 and 4.14 pCi/L), the concentration was about 0.26 pCi/L in the water from Well 1587. Plutonium 239 and 240 concentration activities were significantly lower in the 5- $\mu$ m and 3- $\mu$ m filtrates and in subsequent filtrates. These results indicated that Pu is particle-reactive, preferentially associating with the particulate size fraction in these water samples. More than 92 percent of the Pu 239 and 240 concentration in the Well 1587 sample was associated with the particulate fraction greater than 5  $\mu$ m. Similarly, 80 percent of the Pu 239 and 240 activity in the SW-51 sample and 79 percent of the Pu 239 and 240 activity in the SW-53 sample were associated with the particulate fraction. The only colloid fractions that compose a substantial part of total Pu 239 and 240 activity were the 0.45- to 3- $\mu$ m fraction from SW-53 where about 20 percent of total concentration was associated with this fraction.

The unfiltered concentration of about 0.26 pCi/L in water from Well 1587 was an order of magnitude greater than the concentration (0.0235 pCi/L) measured in the November 1991 sampling of the same well (Harnish and others, 1994). This disparity may be due to mobilization of Pu by the greater infiltration rate and shear forces that could detach Pu-bearing particles from aquifer solids or surface soils prior to infiltration of water in the spring rainy season. Alternatively, the disparity may be due to the difficulty of obtaining representative samples in systems containing large particles and particle-reactive contaminants. Large particles are less likely to be uniformly distributed in the sample, and, thus, a single sampling of a water for a particle-associated species is less likely to be representative of mean concentration of that species. Americium 241 concentrations in the filtrates from the three sites were generally near or less than detection limits (table 14). No interpretation of size-distribution association was possible. Uranium 233 and 234 were detected in the unfiltered and filtrate samples from all three sites (table 14), and the uniformity of concentrations in all of the fractions indicated that most of the U 233 and 234 in these samples was in the dissolved phase.

### Particle Analysis

Results of radiochemical analysis of the dried retentates from the three samples and of the grab samples of algae obtained at sites SW-51 and SW-53 are listed in table 15. These concentrations are a measure of the extent of reactivity of each particle-size fraction with Pu 239 and 240, Am 241, and U 233 and 234. The data do not indicate relative abundances of particles among the size fractions and, thus, are not a measure of the contribution of each size fraction to total radionuclide load in the water. The mass of the recovered particulate fraction and the largest colloid fraction from SW-53 are from a 3- $\mu$ m filter and are not directly comparable to the same fractions collected at the other two sample sites.

Plutonium 239 and 240 activities associated with the particulate fractions were remarkably consistent among the three sample sites (about 7.25, 7.68, and 8.23 pCi/g for Well 1587, SW-51, and SW-53) (table 15). Plutonium activities associated with the colloidal fractions, however, indicated considerable variation among these sites. The largest colloidal fraction (0.45 to 5  $\mu$ m for SW-51 and 0.45 to 3  $\mu$ m for SW-53) was associated with the highest Pu 239 and 240 activity concentrations in samples from SW-51 and SW-53 (28.05 and 23.33 pCi/g). Plutonium activity in the Well 1587 0.45- to 5- $\mu$ m colloidal fraction was much lower (2.23 pCi/g). The sample from SW-51 had high Pu activities for the two smaller colloid fractions (10.72 pCi/g for the 100,000-Dalton to 0.45- $\mu$ m fraction

**Table 15** Results of radiochemical analysis of dried retentates and algal samples recovered in the filtration of water from Well 1587 and surface-water sites SW-51 and SW-53

[Pu, plutonium, Am, americium, U, uranium,  $\mu\text{m}$ , micrometer; pCi/g, picocuries per gram, ND, not detected, >, greater than, --, no data]

Phase	Size	Activity (pCi/g)		
		Pu 239 and 240	Am 241	U 233 and 234
<b>Well 1587</b>				
Particulate	>5 $\mu\text{m}$	7.25 $\pm$ 1.99	1.34 $\pm$ 0.82	ND
Colloid	0.45 $\mu\text{m}$ –5 $\mu\text{m}$	2.23 $\pm$ 0.74	0.54 $\pm$ 0.38	ND
Colloid	100,000 Daltons - 0.45 $\mu\text{m}$	--	--	--
Colloid	10,000 Daltons - 100,000 Daltons	1.07 $\pm$ 0.32	0.31 $\pm$ 0.17	0.98 $\pm$ 0.77
<b>SW-51</b>				
Particulate	>5 $\mu\text{m}$	7.68 $\pm$ 0.93	11.45 $\pm$ 2.59	5.44 $\pm$ 4.15
Colloid	0.45 $\mu\text{m}$ –5 $\mu\text{m}$	28.05 $\pm$ 4.32	5.44 $\pm$ 1.67	5.27 $\pm$ 5.10
Colloid	100,000 Daltons - 0.45 $\mu\text{m}$	10.72 $\pm$ 1.65	3.10 $\pm$ 0.89	2.83 $\pm$ 2.17
Colloid	10,000 Daltons - 100,000 Daltons	13.09 $\pm$ 1.98	0.89 $\pm$ 0.38	2.20 $\pm$ 1.93
Algae <sup>1</sup>		12.79 $\pm$ 0.76	2.78 $\pm$ 0.22	0.38 $\pm$ 0.15
<b>SW-53</b>				
Particulate	>3 $\mu\text{m}$	8.23 $\pm$ 0.40	1.81 $\pm$ 1.02	0.10 $\pm$ 0.04
Colloid	0.45 $\mu\text{m}$ –3 $\mu\text{m}$	23.33 $\pm$ 2.41	3.49 $\pm$ 0.59	2.99 $\pm$ 1.29
Colloid	100,000 Daltons - 0.45 $\mu\text{m}$	0.53 $\pm$ 0.22	0.38 $\pm$ 0.17	2.89 $\pm$ 1.75
Colloid	10,000 Daltons - 100,000 Daltons	0.34 $\pm$ 0.18	0.09 $\pm$ 0.08	3.19 $\pm$ 1.43
Algae <sup>1</sup>		1.13 $\pm$ 0.13	0.38 $\pm$ 0.09	ND

<sup>1</sup>Grab sample.

343 64

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Table 16. Results of radiochemical analysis of recovered retentates, particles adhering to filters, and dissolved phase in water from Well 1587

[Pu, plutonium, Am, americium, U, uranium, pCi/L, picocuries per liter;  $\mu\text{m}$ , micrometer; ND, not detected, >, greater than, <, less than, -- no data]

Phase	Size	Recovered retentates	Particles adhering to filters	Total	Percent of total activity in size fraction	Percent of particle mass in size fraction
Pu 239 and 240 Activity (pCi/L)						
Particulate	>5 $\mu\text{m}$	8.70E-4 $\pm$ 2.39E-4	2.88E-5 $\pm$ 3.50E-6	8.99E-4 $\pm$ 2.43E-4	--	9
Colloid	0.45 $\mu\text{m}$ -5 $\mu\text{m}$	4.46E-4 $\pm$ 1.48E-4	3.45E-4 $\pm$ 5.50E-6	7.91E-4 $\pm$ 1.54E-4	--	16
Colloid	100,000-Daltons - 0.45 $\mu\text{m}$	--	7.75E-5 $\pm$ 1.24E-5	7.75E-5 $\pm$ 1.24E-5	--	28
Colloid	10,000-Daltons - 100,000-Daltons	6.74E-4 $\pm$ 2.02E-4	1.10E-5 $\pm$ 2.10E-6	6.85E-4 $\pm$ 2.04E-4	--	47
Dissolved	<10,000-Daltons			<sup>1</sup> 1.20E-2 $\pm$ 2.0E-3	--	
Total		1.99E-3 $\pm$ 5.89E-4	4.60E-4 $\pm$ 2.35E-5	1.52E-2 $\pm$ 2.61E-3	--	100
Am 241 Activity (pCi/L)						
Particulate	>5 $\mu\text{m}$	1.61E-4 $\pm$ 9.84E-5	2.82E-5 $\pm$ 2.63E-5	1.89E-4 $\pm$ 1.25E-4	--	9
Colloid	0.45 $\mu\text{m}$ -5 $\mu\text{m}$	1.08E-4 $\pm$ 7.60E-5	3.32E-5 $\pm$ 1.87E-5	1.41E-4 $\pm$ 9.47E-5	--	16
Colloid	100,000-Daltons - 0.45 $\mu\text{m}$	--	1.05E-5 $\pm$ 4.30E-6	1.05E-5 $\pm$ 4.30E-6	--	28
Colloid	10,000-Daltons - 100,000-Daltons	1.95E-4 $\pm$ 1.07E-4	1.02E-5 $\pm$ 9.40E-6	2.97E-4 $\pm$ 1.16E-4	--	47
Dissolved	<10,000-Daltons			<sup>2</sup> 9.59E-4 $\pm$ 5.66E-4	--	
Total		4.64E-4 $\pm$ 2.81E-4	8.10E-5 $\pm$ 5.87E-5	1.60E-3 $\pm$ 9.06E-4	--	100
U 233 and 234 (pCi/L)						
Particulate	>5 $\mu\text{m}$	ND	2.49E-4 $\pm$ 1.02E-4	2.49E-4 $\pm$ 1.02E-4	--	9
Colloid	0.45 $\mu\text{m}$ -5 $\mu\text{m}$	ND	ND	ND	--	16
Colloid	100,000-Daltons - 0.45 $\mu\text{m}$	--	ND	ND	--	28
Colloid	10,000-Daltons - 100,000-Daltons	6.17E-4 $\pm$ 8.5E-4	ND	6.17E-4 $\pm$ 8.5E-4	--	47
Dissolved	<10,000-Daltons			<sup>1</sup> 5.50E-1 $\pm$ 1.32E-2	--	
Total		6.17E-4 $\pm$ 4.85E-4	2.49E-4 $\pm$ 1.02E-4	5.51E-1 $\pm$ 1.38E-2	--	100

<sup>1</sup>Direct analysis of less than 10,000-Dalton filtrate.

<sup>2</sup>Sum of anion- and cation-exchange resin extraction of less than 10,000-Dalton filtrate

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**Table 17. Results of radiochemical analysis of recovered retentates, particles adhering to filters, and dissolved phase in water from surface-water site SW-51**

[Pu, plutonium, Am, americium, U, uranium, pCi/L, picocuries per liter;  $\mu\text{m}$ , micrometer; ND, not detected, >, greater than, <, less than, --, no data]

Phase	Size	Recovered retentates	Particles adhering to filters	Total	Percent of total activity in size fraction	Percent of particle mass in size fraction
<b>Pu 239 and 240 Activity (pCi/L)</b>						
Particulate	>5 $\mu\text{m}$	4.15E-2 $\pm$ 4.97E-3	9.54E-3 $\pm$ 1.43E-3	5.10E-2 $\pm$ 6.40E-3	28	18
Colloid	0.45 $\mu\text{m}$ –5 $\mu\text{m}$	1.29E-2 $\pm$ 1.01E-3	1.20E-2 $\pm$ 1.12E-3	2.49E-2 $\pm$ 3.13E-3	14	16
Colloid	100,000-Daltons - 0.45 $\mu\text{m}$	8.79E-3 $\pm$ 1.35E-3	1.27E-2 $\pm$ 7.60E-3	2.15E-2 $\pm$ 8.95E-3	12	27
Colloid	10,000-Daltons - 100,000-Daltons	1.54E-2 $\pm$ 2.30E-3	1.11E-2 $\pm$ 6.63E-4	2.65E-2 $\pm$ 2.96E-3	14	39
Dissolved	<10,000-Daltons	--	--	<sup>1</sup> 5.90E-2 $\pm$ 8.26E-3	32	--
<b>Total</b>		7.86E-2 $\pm$ 1.06E-2	4.53E-2 $\pm$ 1.08E-2	1.83E-1 $\pm$ 2.97E-2	100	100
<b>Am 241 Activity (pCi/L)</b>						
Particulate	>5 $\mu\text{m}$	6.18E-3 $\pm$ 1.39E-3	1.21E-3 $\pm$ 1.27E-4	7.39E-3 $\pm$ 1.52E-3	33	18
Colloid	0.45 $\mu\text{m}$ –5 $\mu\text{m}$	2.50E-3 $\pm$ 7.73E-4	2.63E-3 $\pm$ 2.60E-4	5.13E-3 $\pm$ 1.03E-3	23	16
Colloid	100,000-Daltons - 0.45 $\mu\text{m}$	2.54E-3 $\pm$ 7.24E-4	1.31E-3 $\pm$ 1.70E-4	3.85E-3 $\pm$ 8.94E-4	17	27
Colloid	10,000-Daltons - 100,000-Daltons	1.05E-3 $\pm$ 4.41E-4	6.44E-4 $\pm$ 1.10E-5	1.69E-3 $\pm$ 4.52E-4	7	39
Dissolved	<10,000-Daltons	--	--	<sup>2</sup> 4.56E-3 $\pm$ 6.45E-4	20	--
<b>Total</b>		1.23E-2 $\pm$ 3.33E-3	5.79E-3 $\pm$ 5.68E-4	2.26E-2 $\pm$ 4.54E-3	100	100
<b>U 233 and 234 (pCi/L)</b>						
Particulate	>5 $\mu\text{m}$	2.94E-3 $\pm$ 2.23E-3	2.81E-4 $\pm$ 1.98E-4	3.22E-3 $\pm$ 2.43E-3	<1	18
Colloid	0.45 $\mu\text{m}$ –5 $\mu\text{m}$	2.42E-3 $\pm$ 2.36E-3	3.37E-4 $\pm$ 1.21E-4	2.76E-3 $\pm$ 2.48E-3	<1	16
Colloid	100,000-Daltons - 0.45 $\mu\text{m}$	2.32E-3 $\pm$ 1.77E-3	5.64E-4 $\pm$ 1.25E-4	2.88E-3 $\pm$ 1.90E-3	<1	27
Colloid	10,000-Daltons - 100,000-Daltons	2.60E-3 $\pm$ 2.27E-3	6.74E-4 $\pm$ 4.07E-4	3.27E-3 $\pm$ 2.68E-3	<1	39
Dissolved	<10,000-Daltons	--	--	<sup>1</sup> 1.32E+0 $\pm$ 1.20E-1	99	--
<b>Total</b>		1.03E-2 $\pm$ 8.63E-3	1.85E-3 $\pm$ 8.51E-4	1.33E+0 $\pm$ 1.30E-1	100	100

<sup>1</sup>Direct analysis of less than 10,000-Dalton filtrate.

<sup>2</sup>Sum of anion- and cation-exchange resin extraction of less than 10,000-Dalton filtrate

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## Dissolved-Phase Associations with Fulvic Acid

The concentrations of Pu 239 and 240, Am 241, and U 233 in various dissolved phases for the three water samples are listed in tables 19-21. As described in the discussion of particle and filter analysis, the SW-53 sample had the greatest concentration of particulates, and most of the Pu 239 and 240 was associated with this particulate material. For the other two samples, the activity concentration of dissolved Pu 239 and 240 was greater than that of the particulate Pu 239 and 240. The total-dissolved Am 241 activity concentration was less than detection in all three samples. For all samples, most of the U 233 and 234 was in the dissolved phase.

The activity concentration of Pu 239 and 240 complexed by fulvic acid was calculated in two ways. In the first method of calculation (direct), the activity concentration of radionuclide in the isolated, lyophilized fulvic acid was multiplied by the fulvic-acid concentration. In the second method of calculation (difference), the anion and cation activity concentration were summed for the influent filtrate and the XAD-8 effluent. This sum represents an estimate of the dissolved activity concentration. Then, the fulvic acid associated actinide was calculated as the difference between the sum for the influent filtrate and the sum for the XAD-8 effluent. For Pu 239 and 240, the sum of the cation and anion activity is very close to the direct measurements of dissolved Pu 239 and 240 (77 percent to 109 percent of the dissolved concentration). For Am 241, the dissolved activity concentration was less than detection, which is the reason for basing the second calculation on the sum of the cation and anion activity concentrations. The calculation by difference for U 233 and 234 resulted in comparable values to the direct calculation, but the values had a greater error.

For all three water samples, the two calculations for fulvic acid associated Pu 239 and 240 produced very similar results, indicating that between 57 percent and 88 percent of the dissolved Pu 239 and 240 was complexed by fulvic acid. The similarity of the results of the two calculations indicates that the alteration of pH during the isolation procedure does not disrupt the fulvic-acid/Pu complexes. At the very high ligand to metal ratios in these samples, strong complexes would be predominant, and it is not surprising that these complexes would not be disrupted by pH alterations. The activity concentrations of Am 241 associated with the fulvic acid were very low, consistently two orders of magnitude lower than the concentrations of Pu 239 and 240 associated with fulvic acid. In contrast, the activity of fulvic-acid-bound U in all three filtrates was larger than that for fulvic-acid-bound Pu. The activity was, however, not as high a percentage of the total dissolved activity.

## Dissolved-Phase Anionic and Cationic Associations

The 10,000-Dalton filtrates and the effluents from the XAD-8 resin were reacted with the anion- and cation-exchange resins to further access the chemical speciation of the dissolved actinides. The results are presented in tables 19, 20, and 21 and are summarized in table 22. A general pattern is evident. When the samples were reacted with the anion-exchange resin, most of the dissolved radionuclides were removed, whereas only small proportions of the radionuclides were removed by the cation-exchange resin. The finding that the cationic forms of all three radionuclides were consistently the fraction that had the least activity indicated that the dissolved radionuclides are not present as simple ions, such as  $\text{PuO}_2^+$ .

In the case of Pu 239 and 240, these results are consistent with the previous result that the dissolved Pu 239 and 240 was complexed by fulvic acid. Fulvic acids are anionic at the near neutral pH of the samples and would have been sorbed by the anion-exchange resin. The results, therefore, can be explained by the sorption of fulvic acids containing complexed Pu 239 and 240 and provide further support for the importance of fulvic-acid complexation of Pu 239 and 240. However, for Am 241, the activity concentration removed by the anion-exchange resin is ten to twenty times greater than that associated with the fulvic acid. There are several possible explanations for this result. One explanation is that the dissolved Am 241 species are present as inorganic anions. Another explanation is that dissolved Am 241 is complexed by organic ligands that are not isolated as fulvic acid, such as more hydrophilic organic acids (Aiken and others, 1992). For U 233 and 234, the anionic activity concentration also is greater than the fulvic-acid-associated activity concentration, but not to as great an extent as for Am 241. The same two possible explanations also are applicable to this result.

**Table 20** Results of radiochemical analysis of less than 10,000-Dalton dissolved phase in water from surface-water site SW-51 for fulvic-acid, anionic, and cationic associations

[Pu, plutonium, Am, americium, U, uranium, ND, not detected, FAB, fulvic-acid bound, XAD-AEX, post-XAD 8 anion-exchange resin, AEX, anion-exchange resin, XAD-CEX, post-XAD-8 cation-exchange resin, CEX, cation-exchange resin, pCi/L, picocuries per liter]

Sample	Activity of sample (pCi/L)		
	Pu 239 and 240	Am 241	U 233 and 234
Filtrate	5 90E-2±8.26E-3	ND	1.32E+0±1.20E-1
XAD-8 isolate (FAB) <sup>1</sup>	5.21E-2±5 88E-4	2.26E-4±3 15E-5	9 60E-2±2.30E-2
XAD-8 isolate (FAB) <sup>2</sup>	4 97E-2±9.58E-3	4 77E-4±1 96E-3	1.23E-1±6 90E-2
Anion-exchange resin (AEX)	6 05E-2±3.25E-3	4 46E-3±5 58E-4	6.58E-1±4 83E-2
Cation-exchange resin (CEX)	9 97E-4±3 04E-4	1 04E-4±8 72E-5	3.34E-2±5 27E-3
Total AEX and CEX	6 15E-2±3.55E-3	4.56E-3±6 45E-4	6 91E-1±5 36E-2
Post-XAD-8 anion-exchange resin (XAD-AEX)	1 16E-2±5 88E-3	3 93E-3±1 20E-3	5.28E-1±4 12E-2
Post XAD-8 cation-exchange resin (XAD-CEX)	2.14E-4±1 46E-4	1.53E-4±1 08E-4	4 05E-2±5 49E-3
Total FAB and XAD-AEX	6 13E-2±6 47E-3	4 16E-3±1 23E-3	6.24E-1±6 42E-2

<sup>1</sup>Calculated directly from the activity of the XAD-8 isolate ('direct' calculation)

<sup>2</sup>Calculated by difference between the activity of anion- and cation-resin extraction of the 10,000-Dalton filtrate and the activity of post-XAD-8 anion- and cation resin extraction of the 10,000-Dalton filtrate ('difference' calculation).

**Table 22. Summary of fulvic acid, anionic, and cationic-association ratios of actinides in the less than 10,000-Dalton dissolved phase in waer from Well 1587 and surface-water sites SW-51 and SW-53**

[Pu, plutonium, Am, americium, U, uranium, ND, not detected, FAB, fulvic-acid bound, AEX, anion-exchange resin XAD-AEX, post-XAD-8 anion-exchange resin]]

	Ratio of activity in specified fraction to total dissolved activity		
	Pu 239 and 240	Am 241	U 233 and 234
	Well 1587		
FAB activity (direct) <sup>1</sup>	<sup>3</sup> 0 81 <sup>4</sup> (0 62)	<sup>3</sup> 0 05 <sup>4</sup> (ND)	<sup>3</sup> 0 11 <sup>4</sup> (0 09)
FAB activity (difference) <sup>2</sup>	<sup>3</sup> 0 75 <sup>4</sup> (0.57)	<sup>3</sup> 0 13 <sup>4</sup> (ND)	<sup>3</sup> 0 17 <sup>4</sup> (0 14)
Anionic activity (AEX on tables 19-21)	<sup>3</sup> 0 93 <sup>4</sup> (0 71)	<sup>3</sup> 0.95 <sup>4</sup> (ND)	<sup>3</sup> 0 82 <sup>4</sup> (0 67)
Cationic activity (CEX on tables 19-21)	<sup>3</sup> 0 07 <sup>4</sup> (0 05)	<sup>3</sup> 0 05 <sup>4</sup> (ND)	<sup>3</sup> 0 18 <sup>4</sup> (0 15)
AEX/total (FAB and XAD-AEX)	0 91	1 07	0 99
	SW 51		
FAB activity (direct) <sup>1</sup>	<sup>3</sup> 0 85 <sup>4</sup> (0 88)	<sup>3</sup> 0 05 <sup>4</sup> (ND)	<sup>3</sup> 0 14 <sup>4</sup> (0 07)
FAB activity (difference) <sup>2</sup>	<sup>3</sup> 0 81 <sup>4</sup> (0 84)	<sup>3</sup> 0 11 <sup>4</sup> (ND)	<sup>3</sup> 0 18 <sup>4</sup> (0 09)
Anionic activity	<sup>3</sup> 0 98 <sup>4</sup> (1 03)	<sup>3</sup> 0.98 <sup>4</sup> (ND)	<sup>3</sup> 0.95 <sup>4</sup> (0 50)
Cationic activity	<sup>3</sup> 0 02 <sup>4</sup> (0 02)	<sup>3</sup> 0 02 <sup>4</sup> (ND)	<sup>3</sup> 0 05 <sup>4</sup> (0 03)
AEX/total (FAB and XAD-AEX)	0 95	1 07	1 05
	SW-53		
FAB activity (direct) <sup>1</sup>	<sup>3</sup> 0 68 <sup>4</sup> (0 74)	<sup>3</sup> 0 10 <sup>4</sup> (ND)	<sup>3</sup> 0 15 <sup>4</sup> (0 11)
FAB activity (difference) <sup>2</sup>	<sup>3</sup> 0 68 <sup>4</sup> (0 74)	<sup>3</sup> 0 09 <sup>4</sup> (ND)	<sup>3</sup> 0.21 <sup>4</sup> (0 15)
Anionic activity	<sup>3</sup> 0 96 <sup>4</sup> (1 04)	<sup>3</sup> 0 95 <sup>4</sup> (ND)	<sup>3</sup> 0 92 <sup>4</sup> (0 67)
Cationic activity	<sup>3</sup> 0 04 <sup>4</sup> (0 05)	<sup>3</sup> 0 05 <sup>4</sup> (ND)	<sup>3</sup> 0 08 <sup>4</sup> (0 06)
AEX/total (FAB and XAD-AEX)	0 98	0 97	1 05

<sup>1</sup>Calculated directly from activity of XAD-8 isolate (fulvic acid) (FAB<sup>1</sup> on tables 19-21).

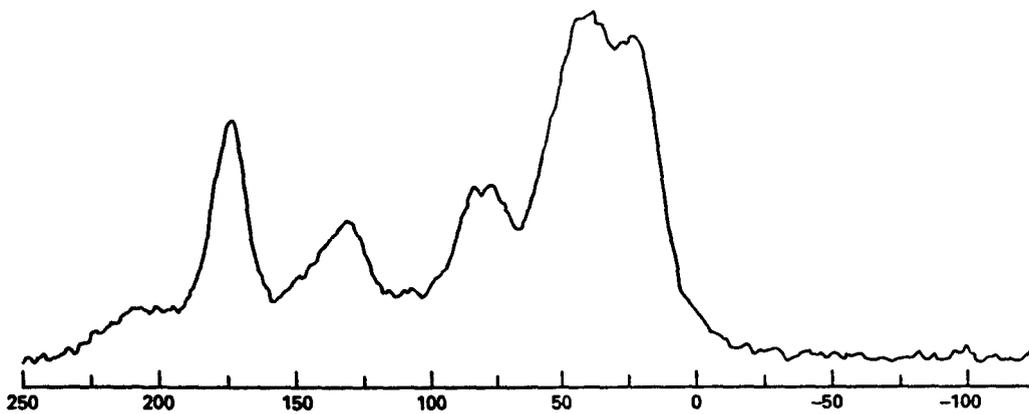
<sup>2</sup>Calculated by difference between activity of anion- and cation-resin extraction of the 10,000-Dalton filtrate and activity of post XAD-8 anion- and cation-resin extraction of the 10,000-Dalton filtrate (FAB<sup>2</sup> on tables 19-21).

<sup>3</sup>Calculated using the sum of anion- and cation-resin extraction of 10,000-Dalton filtrate (total AEX and CEX on tables 19-21).

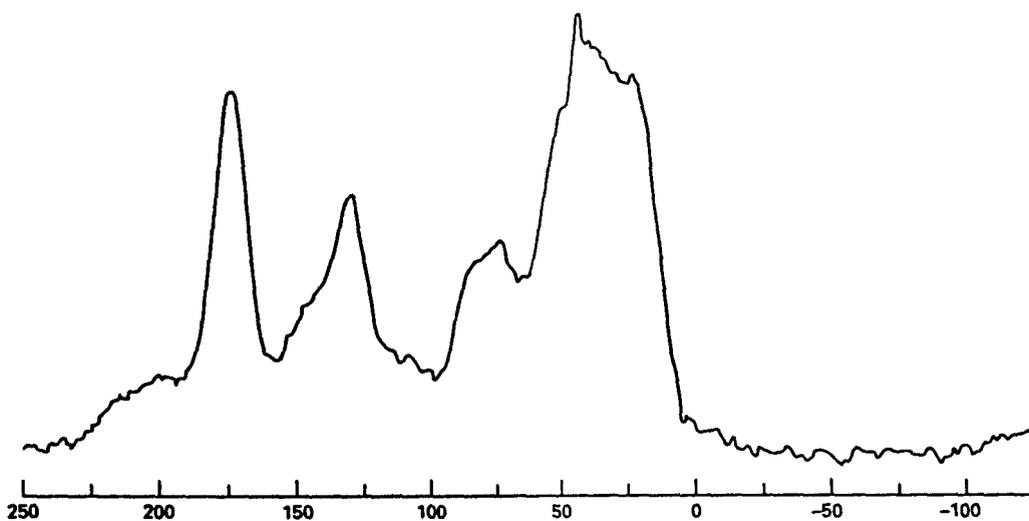
<sup>4</sup>Calculated using direct analysis of 10,000-Dalton filtrate (filtrate on tables 19-21).

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A Well 1587 — XAD-8 ELUATE (FULVIC ACID)



B Well SW-51 — XAD-8 ELUATE (FULVIC ACID)



C Well SW-53 — XAD-8 ELUATE (FULVIC ACID)

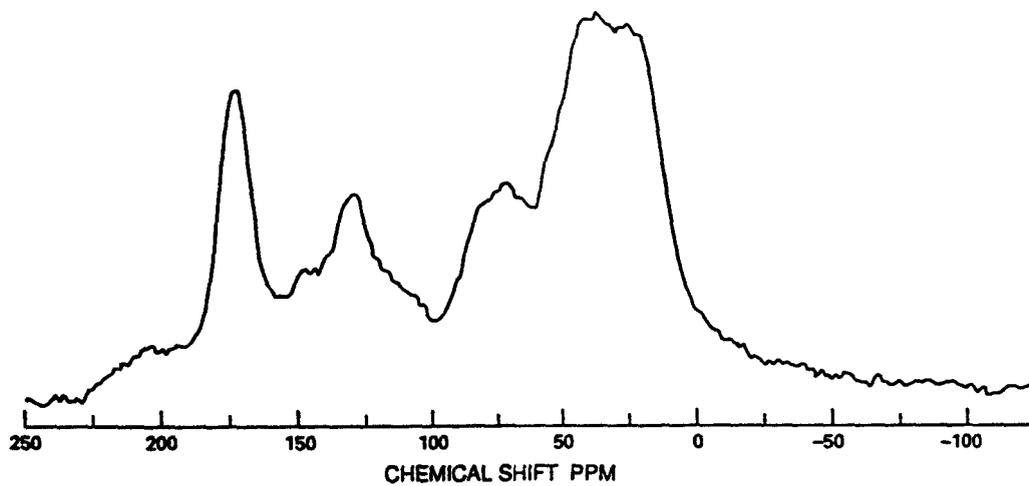


Figure 6.  $^{13}\text{C}$ -Nuclear Magnetic Resonance spectrum of fulvic acid isolated from the dissolved phase of water from A, well 1587, B, SW-51, and C, SW-53

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**Table 25.** Electrophoretic mobility of particles recovered by filtration of water from Well 1587 and surface-water sites SW-51 and SW-53

[ $\mu\text{m/s/V/cm}$ , micrometers per second divided by volts per centimeter; >, greater than,  $\mu\text{m}$ , micrometer; --, no data]

Phase	Size	Electrophoretic mobility ( $\mu\text{m/s/V/cm}$ )		
		Well 1587	SW-51	SW-53
Particulate	>5 $\mu\text{m}$	-1.04 $\pm$ 0.06	-1.05 $\pm$ 0.04	--
Colloid	0.45 $\mu\text{m}$ - 5 $\mu\text{m}$	-1.01 $\pm$ 0.08	-1.03 $\pm$ 0.05	<sup>1</sup> -0.77 $\pm$ 0.05
Colloid	100,000 Daltons - 0.45 $\mu\text{m}$	--	-0.97 $\pm$ 0.06	-0.85 $\pm$ 0.05
Colloid	10,000 Daltons - 100,000 Daltons	-1.24 $\pm$ 0.03	-1.15 $\pm$ 0.05	-1.07 $\pm$ 0.04

<sup>1</sup>0.45  $\mu\text{m}$  - 3  $\mu\text{m}$  for site SW-53

## CONCLUSIONS

- 1 Substantial percentages of total Pu 239 and 240 and Am 241 activity are associated with particulate phases in ground water and particulate and colloidal phases in surface water, despite the large variation in the total concentration of the particulate and colloidal phases
- 2 Uranium 233 and 234 are predominantly in the dissolved phase, again independent of particulate and colloidal concentrations.
- 3 A substantial percentage of dissolved Pu 239 and 240 is complexed by dissolved fulvic acid, and the concentrations of dissolved Pu 239 and 240 increases with dissolved fulvic-acid concentration
- 4 Dissolved cationic forms account for very low percentages of dissolved Pu 239 and 240, Am 241, and U 233 and 234
- 5 Dissolved U 233 and 234 and Am 241 are predominantly anionic in nature, but not associated with dissolved fulvic acids, indicating the possible importance of other organic ligands or inorganic anionic species

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