

Particulate, Colloidal, and Dissolved-Phase Associations of Plutonium and Americium in a Water Sample from Well 1587 at the Rocky Flats Plant, Colorado

by R.A. Harnish, D.M. McKnight and J.F. Ranville

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CONVERSION FACTORS

Multiply	By	To obtain
centimeter (cm)	0.3937	inch
centimeter per second (cm/s)	0.3937	inch per second
kilometer (km)	0.6214	mile
kilometer per year (km/yr)	0.6214	mile per year
liter (L)	0.2642	gallon
liter per minute (L/min)	0.2642	gallon per minute
meter (m)	3.281	foot
micrometer (μm)	3.281×10^{-6}	foot
milligram (mg)	0.03527×10^{-3}	ounce, avoirdupois
milliliter (mL)	0.03382	ounce, fluid
milliliter per minute (mL/min)	0.002642	gallon per minute
millimeter (mm)	0.0394	inch
nanometer (nm)	3.281×10^{-9}	foot

Degree Celsius ($^{\circ}\text{C}$) may be converted to degree Fahrenheit ($^{\circ}\text{F}$) by using the following equation:

$$^{\circ}\text{F} = 9/5(^{\circ}\text{C}) + 32.$$

The following terms and abbreviations also are used in this report:

Dalton: a standard unit of mass equal to one-half the atomic mass of $^{12}_6\text{C}$.

Milligram per liter (mg/L): a solution having one milligram of solute per liter of solution.

Normal (N): a solution having one gram equivalent weight of solute per liter of solution.

Picocuries per liter (pCi/L): solution concentration of radioactivity equivalent to 3.7×10^{-2} disintegrations per second per liter.

Particulate, Colloidal, and Dissolved-Phase Associations of Plutonium and Americium in a Water Sample from Well 1587 at the Rocky Flats Plant, Colorado

By R.A. Hamish, D.M. McKnight and J.F. Ranville

Abstract

In November 1991, the initial phase of a study to determine the dominant aqueous phases that control the transport of plutonium (Pu), americium (Am), and uranium (U) in surface and groundwater at the Rocky Flats Plant was undertaken by the U.S. Geological Survey. By use of the techniques of stirred-cell spiral-flow filtration and crossflow ultrafiltration, particles of three size fractions were collected from a 60-liter sample of water from well 1587 at the Rocky Flats Plant. These samples and corresponding filtrate samples were analyzed for Pu and Am. As calculated from the analysis of filtrates, 65 percent of Pu 239 and 240 activity in the sample was associated with particulate and largest colloidal size fractions. Particulate (22 percent) and colloidal (43 percent) fractions were determined to have significant activities in relation to whole-water Pu activity. Am and Pu 238 activities were too low to be analyzed. Examination and analyses of the particulate and colloidal phases indicated the presence of mineral species (iron oxyhydroxides and clay minerals) and natural organic matter that can facilitate the transport of actinides in ground water. High concentrations of the transition metals copper and zinc in the smallest colloid fractions strongly indicate a potential for organic complexation of metals, and potentially of actinides, in this size fraction.

INTRODUCTION

Particulate and colloidal particles are important in most natural waters for transport, bioavailability, and reactivity of contaminants. Colloidal particles, those particles generally greater than 1 nm and less than 5 μm in diameter, are an especially important compo-

nent because of their high specific surface areas (which facilitate reactions between solids and solution), and their small size (which inhibits settling and allows them to remain suspended for long periods). Because the term "colloidal" indicates only a size classification, the chemical composition and reactivity of colloids in natural waters is varied. Organic colloidal forms include cellular exudates and partially to extensively degraded detrital material that can be associated with mineral phases (Ranville and others, 1991). Inorganic forms include clays and insoluble metal oxides. Colloidal particles have been observed in ground waters (Degueldre and others, 1989; Salbu and others, 1985) and have been shown to be mobile in aquifers (Harvey and others, 1989). Association of contaminants having low solubility values with ground-water colloids is expected to greatly affect contaminant transport (McCarthy and Zachara, 1989; Penrose and others, 1990). Several studies have shown the importance of this process in radionuclide transport (Buddemeier and Hunt, 1988; Penrose and others, 1990).

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, Colo., about 16 miles northwest of downtown Denver. The plant opened in 1952, and plutonium 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 began a study of the potential for facilitated transport of plutonium and americium by colloidal-size particles in the surface and ground water at the site. This study is designed to determine the dominant phases that control the transport of plutonium and americium contaminants in surface and ground water at the Rocky Flats

Plant and, in particular, the potential for colloidal transport of these elements.

This report describes the results from the initial ground-water sampling—specifically: (1) An initial characterization of the chemistry and mineralogy of colloidal material isolated from ground water at well 1587; and (2) determination of the distribution of plutonium and americium between dissolved and colloidal phases.

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 (fig. 1). The geohydrologic units at the plant consist of unconsolidated surficial units (Rocky Flats Alluvium of Quaternary age) underlain by Cretaceous bedrock (fig. 2) (EG&G, 1991b). Water in the Rocky Flats Alluvium is unconfined, and ground-water levels rise in response to spring recharge and individual precipitation events. Ground-water flow direction in the alluvium generally is to the east. Average ground-water velocities are 84 feet per year, based on a horizontal gradient of 0.02 ft/ft, an effective porosity of 0.1, and mean hydraulic conductivity of 4×10^{-4} cm/s (Rockwell International, 1987). Well 1587 is completed in alluvium in the Woman Creek basin, about 300 feet east of the 903 Pad (fig. 3). Well 1587 is a 4-inch diameter, PVC-cased monitoring well extending 22.5 feet through the alluvium to bedrock and is screened through the lower 16.2 feet of alluvium.

Terminology Used in This Report

Application of filtration in environmental sampling has traditionally involved the use of 0.45- μ 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 nanometer to hundreds of micrometers (Buffle and others, 1989, 1992) (table 1). For the present study, three filter pore sizes were used for particle isolation and fractionation, and the following terminology is used in this report to distinguish among the fractions:

1. The term "particle" is used as a general term that refers to all phases of a sample that are not considered to be dissolved and includes colloids and particulates.

2. The term "particulate" refers to the phase that is retained by a 5.0- μ m filter.

3. The term "colloid" includes those particles that will pass through a 5.0- μ m filter but would be retained by a 0.1- μ m filter for larger colloids and by a 10,000-Dalton filter for smaller colloids. A 10,000-Dalton filter corresponds approximately to, but is different from, a 0.001- μ m filter.

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

5. In addition, the following terms refer to filtration fractions: "filtrate" is a compound of any size that passes through a filtration membrane, and "retentate" is a compound of any size that is retained by a filtration membrane of a particular size.

Acknowledgments

The authors wish to thank Scott Grace (U.S. Department of Energy), Gregory Wetherbee (EG&G Rocky Flats, Inc.), and Michael Harris (EG&G Rocky Flats, Inc.) for valuable scientific discussions and logistical assistance in this field effort.

METHODS

Sample Collection

Well 1587 was sampled during November 18–20, 1991. The sample was obtained using a peristaltic pump to collect a 60-L sample that was stored in 20-L polyethylene carboys for transport to a mobile laboratory onsite. Because of the potential for contamination by airborne particles at the site, sample collection and processing were done in a "clean" system: ground water was isolated by use of an inflatable packer at the well, and subsequent air/sample content was minimized by the use of a combination of nitrogen atmosphere, positive pressure, and HEPA (highly toxic particulates) filtration of ambient air. To minimize shear-induced detachment of sorbed particles and resuspension of settled particles in the aquifer matrix, the pumping rate was maintained below 100 mL/min for the duration of sampling. This rate of pumping produced virtually no drawdown in ground-water level after about 4 L of water was withdrawn from the well. Specific conductance, pH, and temperature were mon-

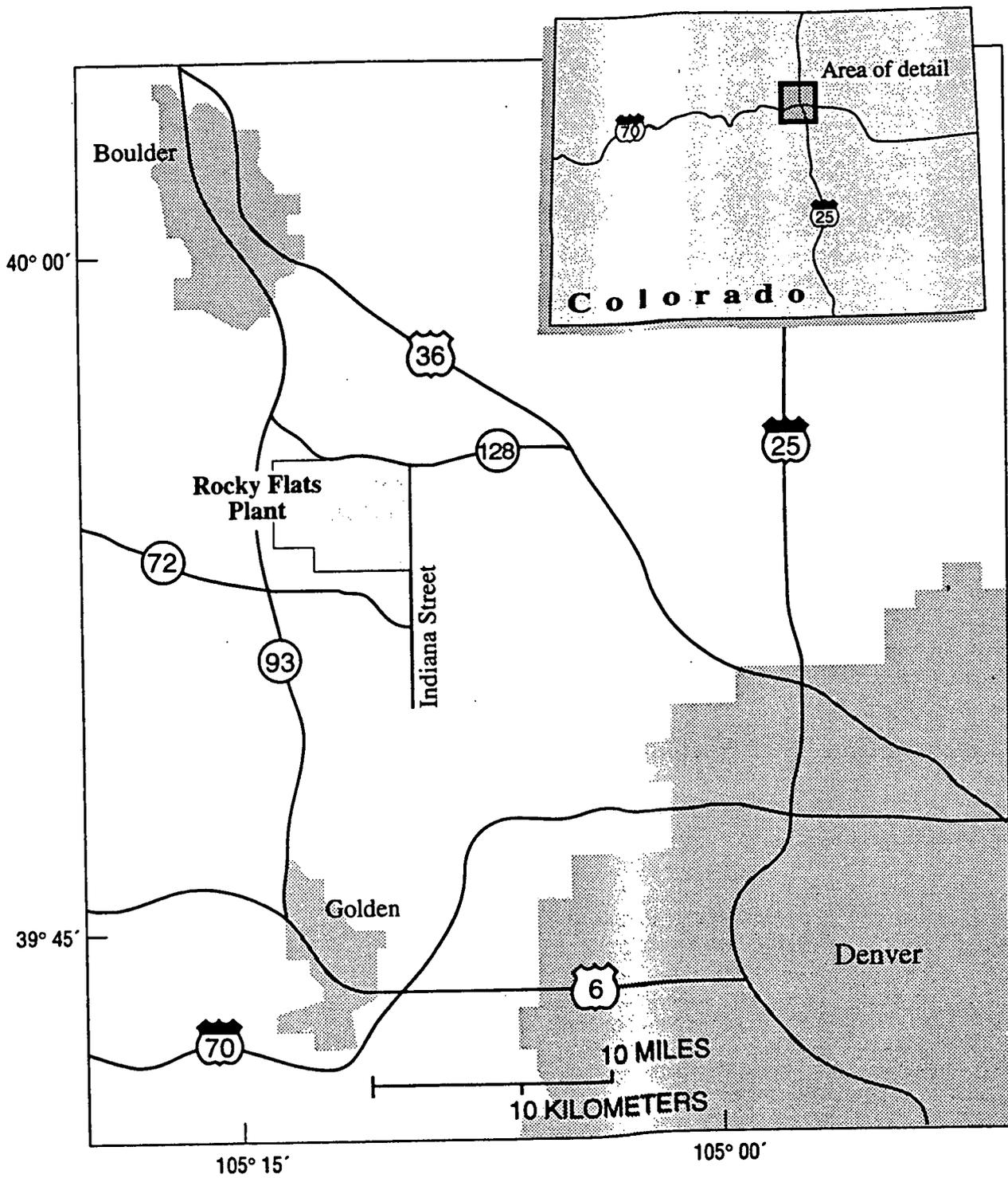


Figure 1. Location of Rocky Flats Plant near Denver, Colorado.

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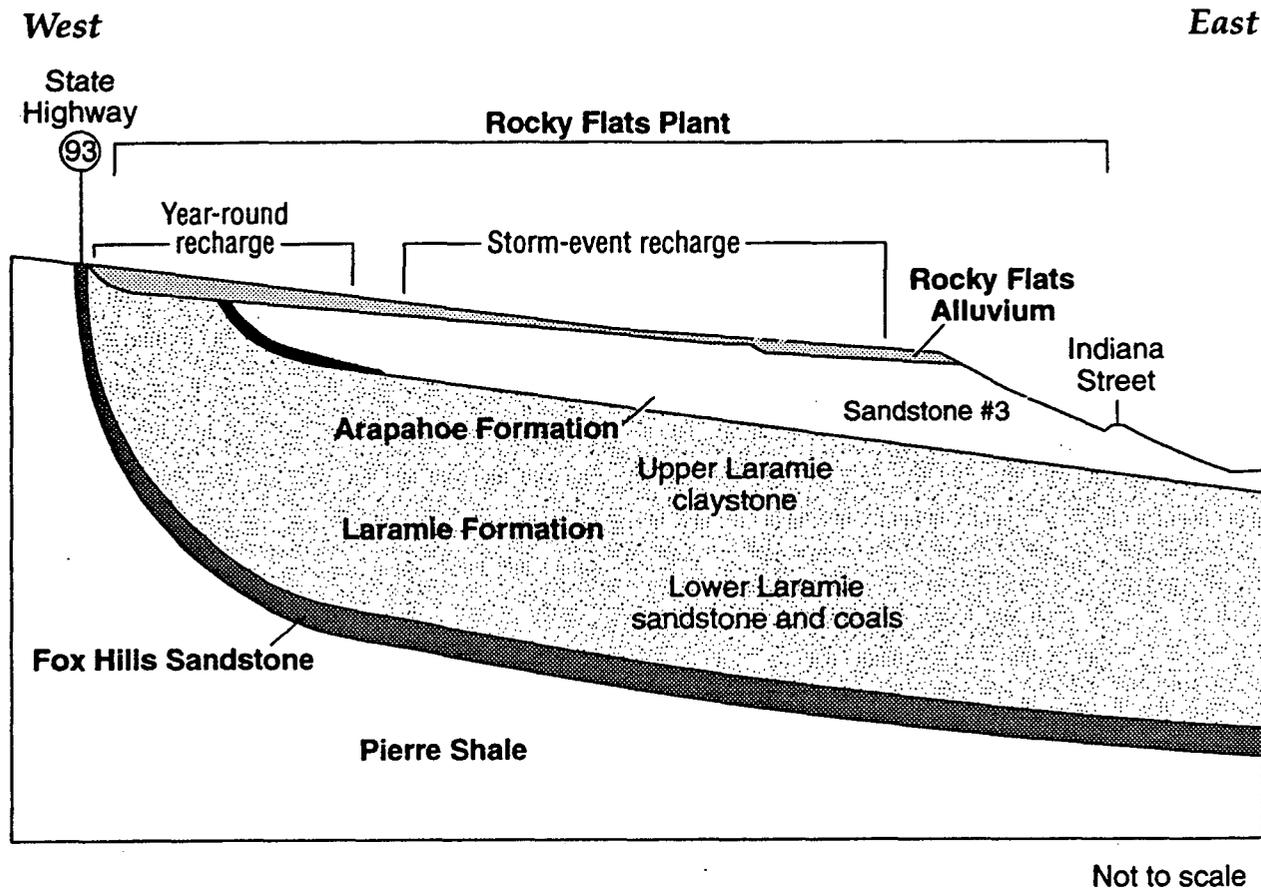


Figure 2. Diagrammatic section of the stratigraphic units underlying Rocky Flats Plant near Denver, Colorado.

itored during initial purging of the well as a check on sufficiency of purging and during sample collection as a check on constancy of sample composition. Values of pH were measured using an ion analyzer and combination pH electrode; temperature was measured with a multiprobe temperature tester, and specific conductance was measured using a conductivity meter with platinum flow-through cell. Twelve well-casing volumes (9,600 mL) were pumped and discarded during initial purging before pH, specific conductance, and temperature stabilized and sample collection was begun (fig. 4). A total sample volume of 60 L was collected and then stored at ground-water temperature under subdued light until processing. Processing was continuous upon collection of sample and was begun within several hours of collection.

Sample Processing

The task of isolating colloids from natural waters is complicated by the unstable nature of these particles

and their low concentration. The goal is to isolate particles that represent the size ranges desired in such a way that the particles are unaltered from their natural state in terms of size, morphology, charge characteristics, surface area, chemical composition, and sorbed species. This goal is unlikely to be achieved, and the most that can be hoped for is a minimal perturbation during processing of the particles. This need for minimal perturbation necessitates the use of filtration methods and sample-handling protocols designed to minimize physical disruption of the particles and to avoid physiochemical processes such as coagulation that alter colloid characteristics. Crossflow (or tangential-flow) ultrafiltration is a relatively new technique that has been used principally in biotechnical applications to concentrate large amounts of cells and macromolecules. In recent years, a number of studies have applied this technique to the isolation of environmental particles (Whitehouse and others, 1986; Gutman, 1987; Hernandez and Stallard, 1988; Barthel and others, 1989; Leenheer and others, 1989; Ranville and others, 1991). The goal of crossflow ultrafiltration is to pre-

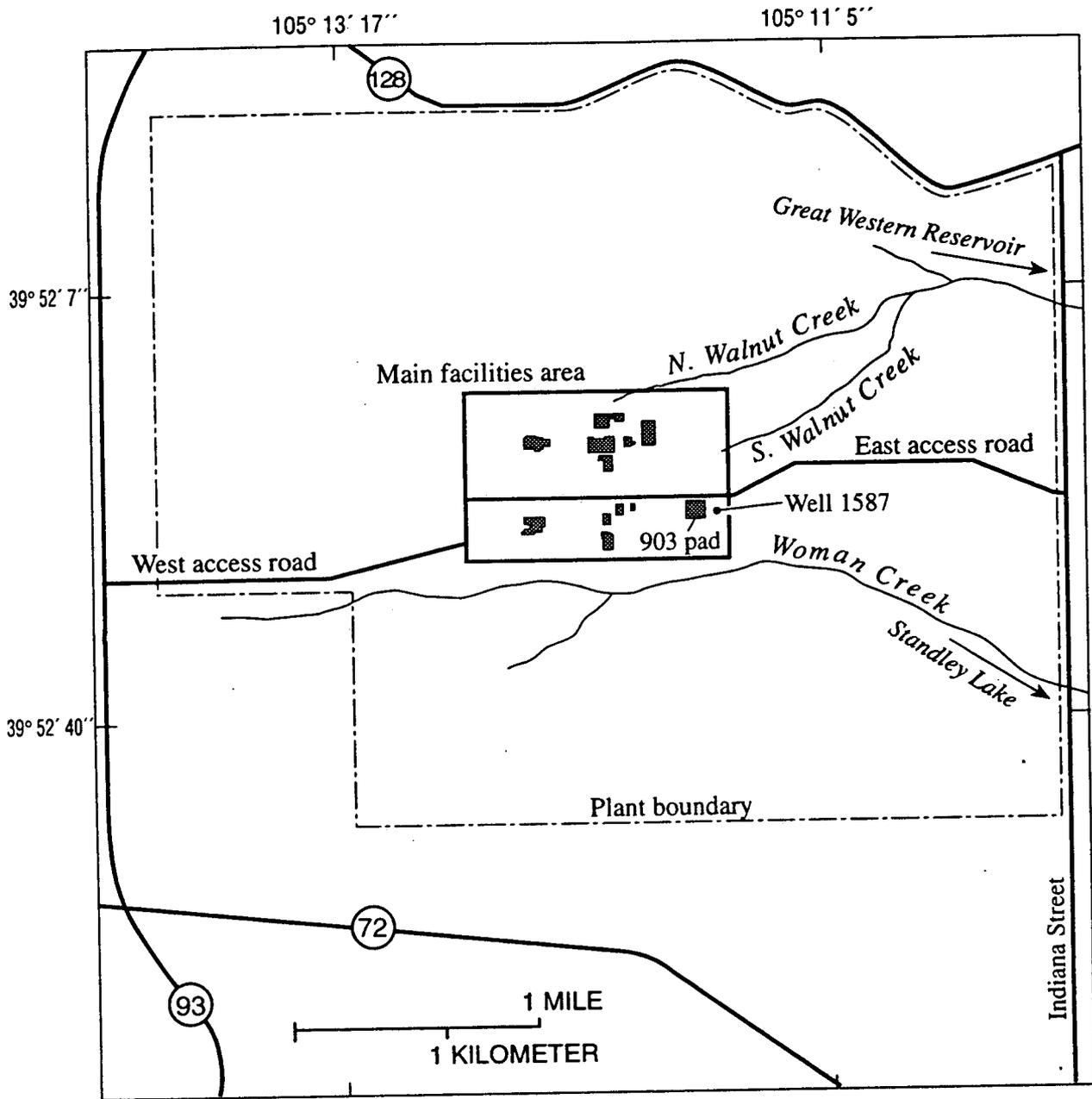


Figure 3. Location of well 1587 at Rocky Flats Plant near Denver, Colorado.

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vent the formation of a filter cake on the membrane surface, which results in clogging. This clogging occurs in conventional filtration and is known as dead-end filtration. Not only does filter clogging contribute to sieving effects and diminishing effective pore diameter, but it also can alter physiochemical properties of the colloids themselves. In crossflow filtration, prevention of clogging is accomplished by the geometry of the filtration apparatus that recirculates the sample over the filter to sweep particles from the membrane surface (fig. 5). By applying a cross-membrane pressure, usually by restricting the return flow, filtrate is forced through the membrane. An optimal pressure that balances filtration flux with minimum particle penetration into the membrane provides the least filter clogging. Thin-channel spiral-flow filtration, also used in this study for particulate isolation, operates by a similar mechanism of cross-filter circulation and similarly minimizes concentration polarization at the filter surface compared to dead-end filtration.

Table 1. Operational classification and nominal physical characteristics of suspended material used in this report

[μm , micrometer; >, greater than; cm/s, centimeter per second; km, kilometer; km/yr, kilometer per year; <, less than; --, not applicable]

Phase	Diameter (μm)	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	0.01	10^4	<0.1 km/yr
Dissolved	<0.01	$<10^4$	--

Particulate and colloid samples were isolated using the techniques of spiral-flow filtration and cross flow ultrafiltration. Filtration protocols are summarized in table 2.

Spiral-Flow Filtration

A spiral-flow ultrafiltration unit with 90-mm-diameter, 5.0- μm pore size, polycarbonate, Poretics membrane filter was used for initial filtration of the 60-L sample. Filters were precleaned with 0.1 N NaOH, 0.1 N HNO₃, and rinsed with distilled water, a procedure that has been effective in removing organic

and inorganic contamination from the filters (Ranville and others, 1991). Filtration equipment was acid washed before use in 5 percent HNO₃ and then rinsed with distilled water. Filtration rate ranged from 400 to 500 mL/min, and the 5- μm filtrate was collected in polyethylene carboys for further processing. Particulates greater than 5 μm were recovered from the filters by placing the filters in a petri dish and gently massaging the filter surface with filtrate water. Particle concentrates were refrigerated at 4° Celsius.

Crossflow Filtration

Fifty-five L of 5.0- μm filtrate were further processed using crossflow ultrafiltration. Two filtration systems were used in series to accomplish the collection of (1) the fraction between 0.1 and 5 μm , and (2) the fraction between 10,000 Dalton and 0.1 μm (fig. 6). The 5- μm filtrate from stirred-cell filtration was filtered first through a crossflow ultrafiltration system with three polysulfone membranes (0.1- μm pore size) providing a total filter surface area of 1,393 cm². The filtrate from the 0.1- μm filtration then was filtered through a second crossflow ultrafiltration system fitted with three 10,000-Dalton pore size polysulfone membranes. During the filtration, the crossflow rate ranged from 2.5 to 3.5 L/min, and the filtrate rate ranged from 200 to 350 mL/min. The final colloid retentate volumes were 0.370 L and 0.575 L for the fraction between 0.1 μm and 5 μm and the fraction between 10,000 Daltons and 0.1 μm , respectively. The filter systems were disassembled, and colloids were removed from the filters by placing a filter in a Teflon bag along with some retentate and gently rubbing the colloids off. This process was repeated for all three filters of each size range. The colloids thus recovered were combined with the retentate for that fraction and were refrigerated at 4° Celsius. During filtration, samples of raw water and the three filtrate fractions were collected for chemical analysis; samples for metals and radiological analysis were acidified to pH less than 2 with nitric acid.

Sample Analysis

Scanning electron microscopy-energy dispersive X-ray analysis (SEM-EDX) on subsamples of the colloid retentates and on the various filtrate fractions was done at the U.S. Geological Survey electron microscopy laboratory in Lakewood, Colo. Approximately 5 mL of the colloid isolates were vacuum-filtered at

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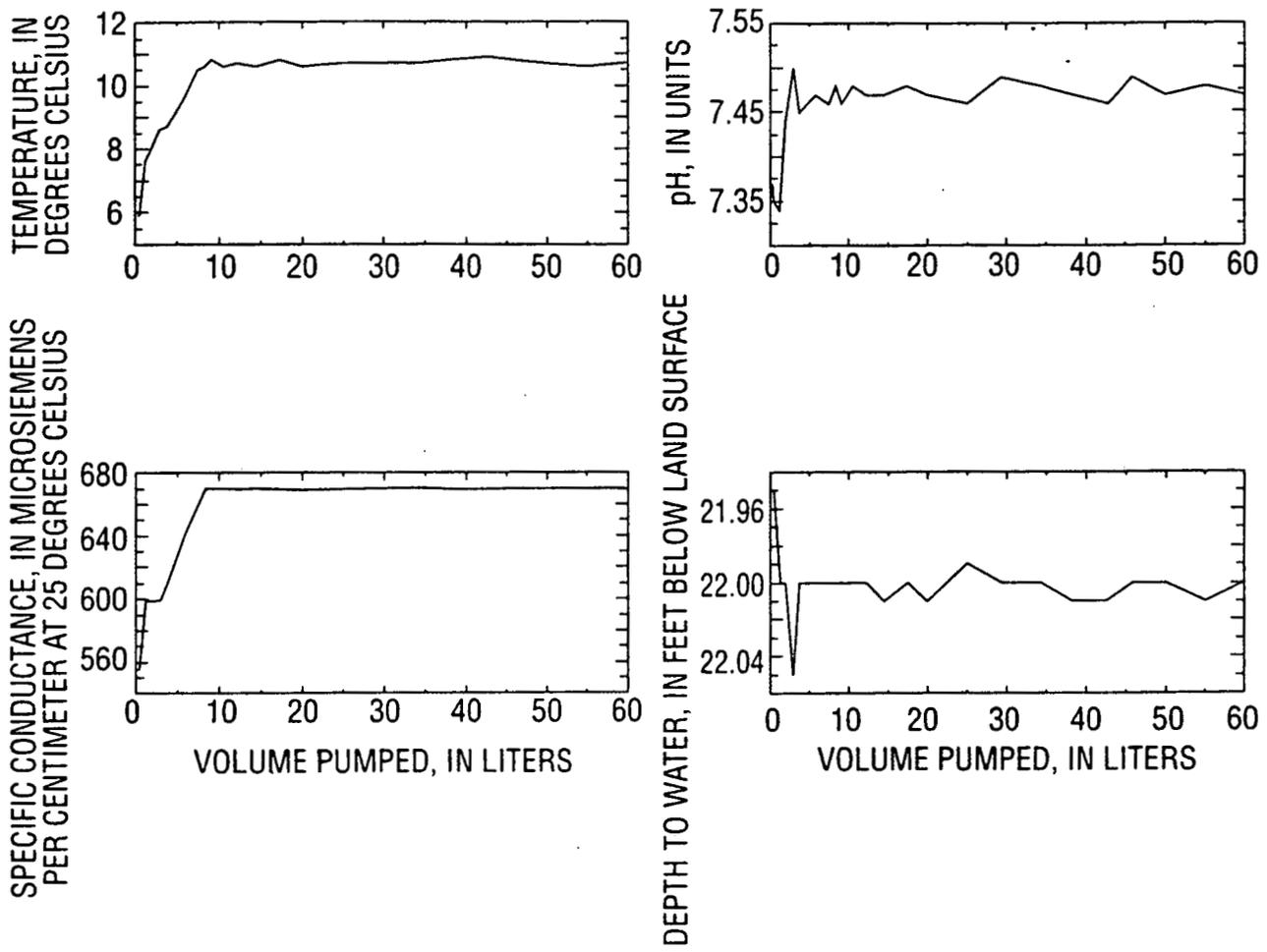


Figure 4. Variation in temperature, pH, specific conductance, and drawdown as a function of volume of water pumped from well 1587.

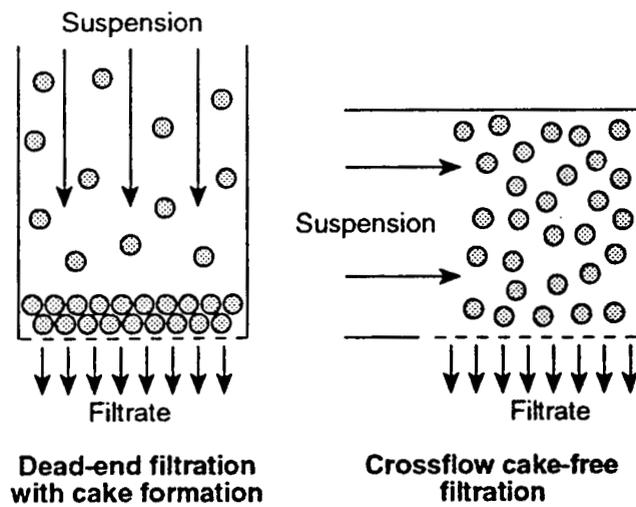


Figure 5. Crossflow and dead-end filtration.

Table 2. Protocols used for filtration of ground water collected at Rocky Flats Plant near Denver, Colorado

[μm , micrometer; cm^3 , cubic centimeters; L, liters]

Filter pore size	Filter type	Filter material	Number of filters used	Filter area (cm^2)	Volume filtered (L)	Retentate volume (L)	Concentration factor
5.0- μm	Poretics	Polycarbonate	6	384	60	0.106	566
0.1- μm	Millipore	Polysulfone	3	1,393	55	0.370	148
10,000-Dalton	Millipore	Polysulfone	3	1,393	50	0.575	87

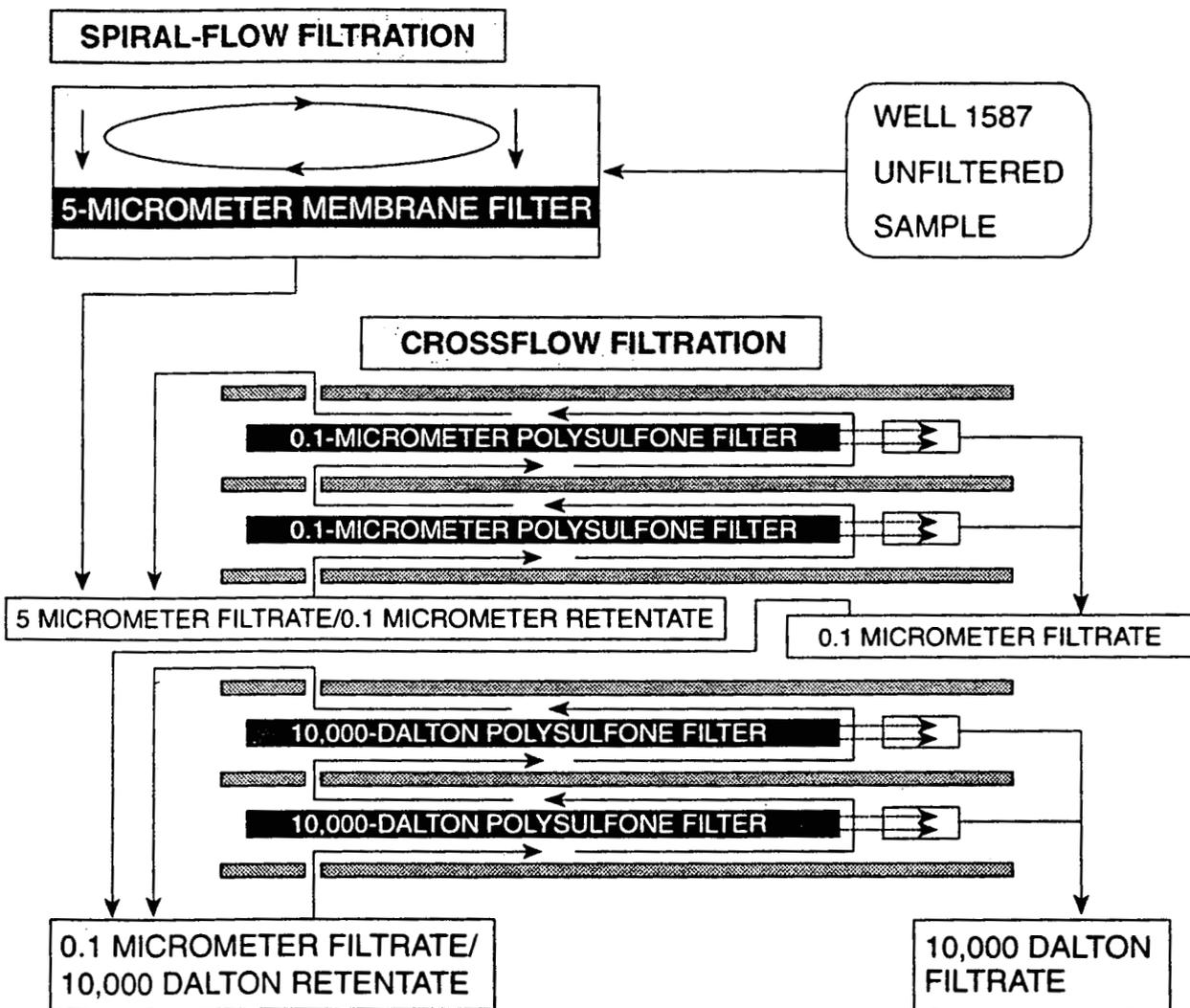


Figure 6. Filtration procedure for processing water sample.

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low vacuum onto a 0.1- μm polycarbonate filter for examination of the collected colloids. Larger volumes (about 100 mL) of the filtrate samples also were sampled in this manner to check filtration integrity. A small area was cut from each filter, mounted to a carbon SEM-EDX stub using carbon paint, and gold-coated for SEM analysis.

Splits of the colloid retentate and the filtrate samples were submitted to IT Corporation laboratories for chemical analysis. Radiological analysis for Pu 239 and 240, Am 241, and Pu 238 was done by IT Laboratories, Richland, Wash., by alpha isotopic separation using an alpha spectrometric detector. Minimum detectable activities were reported as 0.005 pCi/L for Pu 239 and 240, 0.01 pCi/L for Am 241, and 0.01 pCi/L for Pu 238. Samples of colloids for radiological analysis were submitted as suspensions because low colloid concentrations in the retentates precluded recovery of solids by freeze-drying. Metals analysis by inductively coupled plasma-atomic emission spectrometry (ICP-AES) was done by IT Laboratories, Pittsburgh, Pa. Samples of retentates and filtrates were acidified to pH less than 2 with nitric acid and submitted directly for analysis. Detection limits for these analyses are presented in table 3. Radiological and metals analyses both included standard reference sample analysis.

Table 3. Detection limits for inductively coupled plasma-atomic emission spectrometry analysis

Analyte	Detection limit (micrograms per liter)
Aluminum	9.0
Antimony	8.0
Barium	1.0
Beryllium	1.0
Cadmium	1.0
Calcium	7.0
Chromium	4.0
Cobalt	2.0
Copper	3.0
Iron	2.0
Lithium	2.0
Magnesium	11.0
Manganese	1.0
Molybdenum	3.0
Nickel	3.0
Potassium	61.0
Silver	2.0
Sodium	9.0
Strontium	1.0
Tin	14.0
Vanadium	2.0
Zinc	1.0

RESULTS OF FILTRATION AND ANALYSES

Radionuclides in ground water can be present either as particulate, colloidal, or dissolved phases. The particulates and colloids are particle phases, whereas the dissolved phase would behave as solutes. The following discussion presents results of particle isolation, SEM-EDX, radiochemical analysis, and inorganic chemical analysis as they relate to the potential for colloid-facilitated transport of Pu and Am.

Particle Isolation

Three particle retentates were obtained by spiral-flow and crossflow filtration of the 60-L sample that corresponded to the filter pore-size cutoffs used: (1) The fraction greater than 5 μm ; (2) the fraction between 0.1 μm and 5 μm ; and (3) the fraction between 10,000 Dalton and 0.1 μm . Retentate volumes and concentration factors are presented in table 2. Visual examination of the final concentrates indicated that the particle-retentate concentrations were extremely low and that processing of the particle fractions by freeze-drying was impractical; therefore, no direct measurement of colloid mass concentrations was obtained. The greater than 5- μm fraction appeared orange-red, which indicates the presence of iron oxyhydroxides. Particles present in the other two size fractions appeared white to brown, which indicates carbonate and organic compounds; particle numbers were extremely low in the smaller size fractions. Visual examination of the two smaller fractions also showed the presence of filtration artifacts—large particles suspected to be polysulfone particles detached from the filter surface.

Scanning Electron Microscope-Energy Dispersive X-ray Analyses

Scanning electron microscopy was used to determine morphological characteristics and aggregation, and energy dispersive X-ray analysis was used to determine bulk chemical composition of mineral colloids in the greater than 5- μm fraction and the fraction between 5 μm and 0.1 μm . Scanning electron photomicrographs of particles isolated from the greater than 5- μm fraction (figs. 7 and 8) and energy dispersive X-ray spectra of a whole-field scan of these particles (fig. 11) show a large pool of particles composed primarily of iron and smaller amounts of silica, aluminum, calcium, and potassium, which indicates the presence of iron

oxide minerals, calcite, clays, and other detrital aluminosilicates. Large amorphous particles and apparent surface coatings that give no X-ray signal could be organic particles and coatings (fig. 8). An aggregate composed mostly of iron and aluminosilicate minerals is shown in figure 7. An iron-bearing mineral form that is partially covered, perhaps by an organic layer, is shown in figure 8.

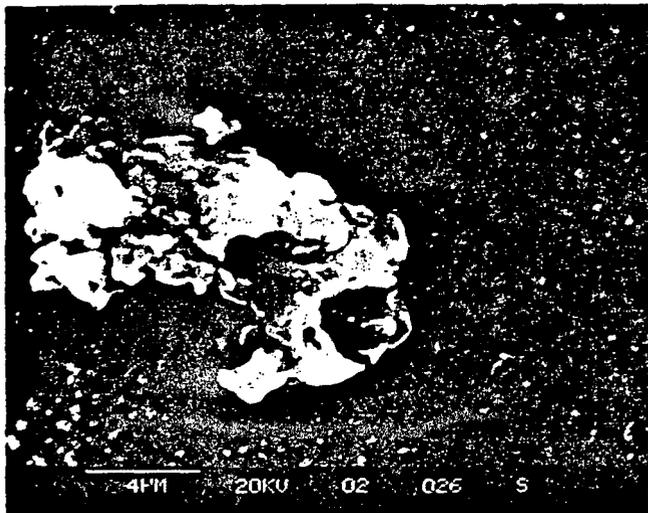


Figure 7. Scanning electron photomicrograph of greater than 5-micrometer aggregate composed mostly of iron and aluminosilicate minerals.



Figure 8. Scanning electron photomicrograph of greater than 5-micrometer iron-bearing particle.

Micrographs and spectra of the isolate fraction between 5 µm and 0.1 µm have many of the same elements as the larger size fraction, although qualitatively, iron forms are less abundant and calcites and aluminosilicates are more abundant.

Figure 9 shows the field of particles dominated by calcite crystals and aluminosilicates; some iron forms are present, as are some apparently organic colloids. Large, irregular, folded sheets visible in the micrograph appear to be particles of the filter membrane that have dislodged during filtration. A large aggregate that probably formed as a post-collection artifact and was composed of clay minerals, iron forms, calcite, and apparent organic forms is shown in figure 10. Energy dispersive X-ray scans of particles randomly picked onsite (figs. 12, 13) show a dominance of calcium, potassium aluminosilicate minerals, and diminished signals for iron in this size fraction.

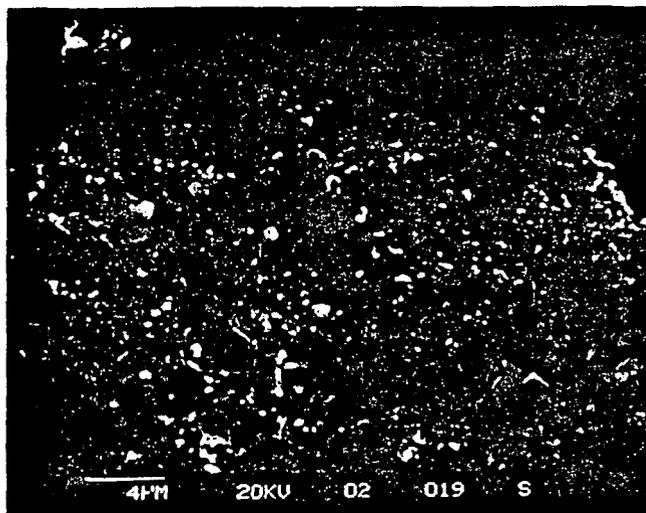


Figure 9. Scanning electron photomicrograph of particles in the 0.1- to 5-micrometer size range.

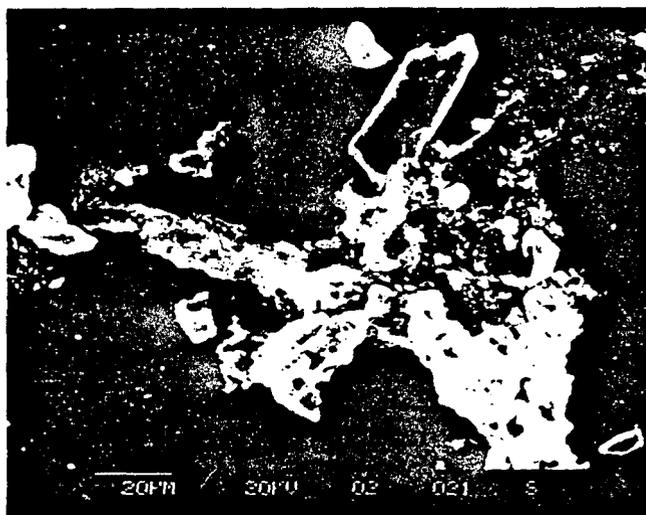


Figure 10. Scanning electron photomicrograph of an aggregate in the 0.1- to 5-micrometer size range probably formed as a post-collection artifact.

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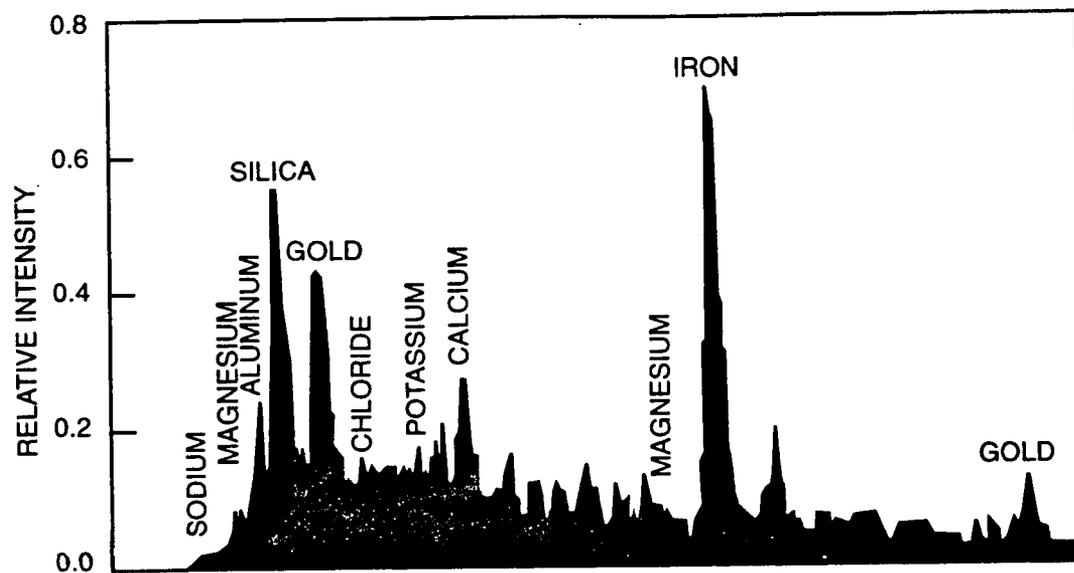


Figure 11. Energy dispersive X-ray spectra of particles in the greater than 5-micrometer size fraction.

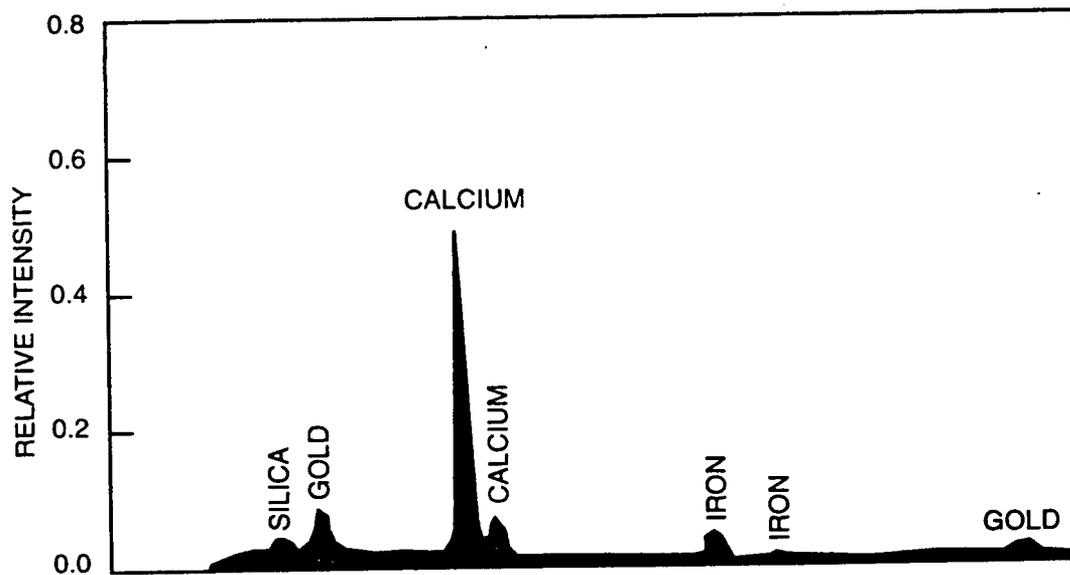


Figure 12. Energy dispersive X-ray spectra of particles in the 0.1- to 5-micrometer size fraction.

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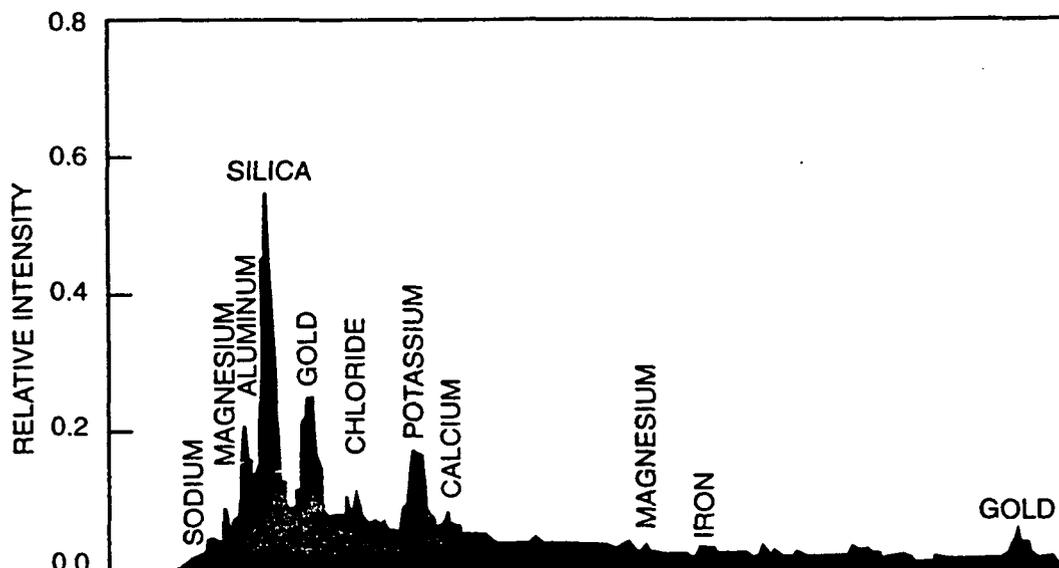


Figure 13. Energy dispersive X-ray spectra of particles in the 0.1- to 5-micrometer size fraction.

The presence of some large aggregates in the fraction between 5 μm and 0.1 μm indicates a problem with post-collection aggregation and emphasizes the need for onsite preparation of SEM-EDX samples at the time of sample collection. The presence of what probably are filter-material particles in this fraction is a filtration artifact and requires further examination of filter pre-cleaning protocols and post-filtration handling.

Radiochemical Analyses

Radiochemical analyses for Pu 239 and 240, Am 241, Pu 238, and U 238 were done on the three particle retentates and four filtrate samples obtained. Results of these analyses for Pu and Am are listed in tables 4 (filtrates) and 5 (particle retentates). For filtrates and particle retentates in all size fractions, radioisotope activity was low. The analytical errors in these measurements are large compared to the measured activities; this is typical of alpha spectrometric measurements of extremely low activities such as these. Alpha spectrometric counting produces a gaussian distribution, and error terms listed here are 2-sigma ranges, indicating the 95-percent confidence limit. None of the U 238 results exceeded the minimal detectable activity.

Data for the radiochemical analysis of the filtrate samples (table 4) indicate that the primary activity measured was for the Pu 239 and 240 species; activity of Am 241 in excess of analytical error occurred only

in the 10,000-Dalton filtrate fraction. In no instance was Pu 238 detected in activities higher than overall analytical error. Despite low activities, however, it is clear that for Pu 239 and 240, activity in the unfiltered sample is higher than activity in the 5- μm filtrate, which, in turn, has higher activity than the 0.1- μm and 10,000-Dalton filtrate. The value for the 10,000-Dalton filtrate is greater than the value for the 0.1- μm filtrate; however, it is well within the range associated with the error for the value from the 0.1- μm filtrate. Therefore, the two values are not in disagreement. These data show that the plutonium that could be removed by filtration was removed by the 0.1- μm filter and are consistent with a substantial portion of the Pu 239 and 240 being in particulate and colloidal form.

Radiochemical analyses of the particle retentates (table 5) indicate a similar pattern: low overall activities for all isotopes in all fractions; significant (approximately 2 times overall error) activity only for Pu 239 and 240 in the 5- μm retentate fraction, and no Pu 238 activity in excess of overall error. Results for radioisotope analyses of these fractions is expressed as picocuries per sample.

To assess the size-fraction distribution of particle-associated radioisotope activity, two methods of calculation are used (tables 6, 7). In the first method, size-fraction distributions are calculated as the difference between filtrate activities for the larger size fractions. For example, in this "difference method" the activity of a radionuclide in the greater than 5- μm par-

Table 4. Results of radiochemical analysis of unfiltered sample and filtrate samples of ground water from well 1587 at Rocky Flats Plant near Denver, Colorado

[Am, americium; Pu, plutonium; μm , micrometer]

Sample	Activity (picocuries per liter)		
	Am 241	Pu 239 and Pu 240	Pu 238
Unfiltered	0.0035 \pm 0.00701	0.0235 \pm 0.0118	0.00247 \pm 0.00349
5- μm filtrate	0 \pm 0.00981	0.0184 \pm 0.00985	0.00405 \pm 0.00504
0.1- μm filtrate	0.00649 \pm 0.00921	0.00552 \pm 0.00522	0.00142 \pm 0.00319
10,000-Dalton filtrate	0.017 \pm 0.0147	0.00832 \pm 0.00593	0.00117 \pm 0.00426

Table 5. Results of radiochemical analysis of particulates and colloids in ground water from well 1587 at Rocky Flats Plant near Denver, Colorado

[Am, americium; Pu, plutonium; μm , micrometer]

Sample	Activity (picocuries per sample)		
	Am 241	Pu 239 and Pu 240	Pu 238
5- μm retentate	0.0217 \pm 0.0211	0.041 \pm 0.0232	0.0 \pm 0.00948
0.1- μm retentate	0.0101 \pm 0.0117	0.00277 \pm 0.0096	0.00 \pm 0.00832
10,000-Dalton retentate	0.00814 \pm 0.0115*	0.00973 \pm 0.00806*	0.000591 \pm 0.00582*

*Activity of the 10,000-Dalton retentate sample is reported as picocuries per liter.

Table 6. Distribution of particulate, colloidal, and dissolved-phase-associated plutonium by size fraction determined from analysis of filtrates

[>, greater than; μm , micrometer; <, less than; ND, not detected]

Phase	Size	Pu 239 and Pu 240	
		Activity (picocuries per liter)	Percent of total activity
Particulate	> 5- μm	0.0051 \pm 0.0216	22
Colloid	0.1- μm - 5- μm	0.0101 \pm 0.0151	43
Colloid	10,000-Dalton* - 0.1- μm	ND	ND
Dissolved	<10,000-Dalton	0.0083 \pm 0.0059	35
	Total:	0.0235 \pm 0.0118	100

* See text for explanation of calculation.

Table 7. Distribution of particulate, colloidal, and dissolved-phase-associated plutonium by size fraction determined from analysis of retentates

[>, greater than; μm , micrometer; <, less than; ND, not detected]

Phase	Size	Pu 239 and Pu 240	
		Activity (picocuries per liter)	Percent of total activity
Particulate	> 5- μm	0.00067 \pm 0.00037	7
Colloid	0.1- μm - 5- μm	ND	ND
Colloid	10,000-Dalton - 0.1- μm	0.00024 \pm 0.00021	3
Dissolved	<10,000-Dalton	0.0083 \pm 0.0059	90
	Total:	0.0092	100

ticulate phase is calculated by subtracting the activity in the 5- μm filtrate from the activity in the unfiltered sample. In the second method, size-fraction distributions are calculated directly from particle retentate activities for each fraction, corrected for dissolved-phase contributions. Assuming that the colloidal retentates represent a complete recovery of the particles existing in the various size fractions, the two methods of calculation should result in the same results for size-fraction distribution of radioisotope activity. However, comparison of calculations by the two methods for the well 1587 sample indicates marked differences (tables 6, 7).

The Pu 239 and 240 activity of the whole-water sample is 0.0235 pCi/L. As calculated by the "difference" method, particulate (greater than 5- μm) Pu 239 and 240 activity is calculated to be 0.0051 pCi/L (table 6). The Pu 239 and 240 activity in the 0.1- μm to 5- μm colloid phase is calculated to be 0.0101 pCi/L as follows. With the removal of Pu in the particulate (greater than 5- μm) and 0.1- μm to 5- μm colloid phases, the filtrates from the 0.1- μm and the 10,000-Dalton filtration have lower Pu concentrations which have a higher relative error compared to the unfiltered and 5- μm filtered fractions. In the continuation of the difference calculation, a value of 0.0083 pCi/L was used for both filtrates; this was done because this value was the greater of these two filtrate measurements. The value of 0.0083 pCi/L is well within the measurement error for the 0.1- μm filtrate and is the value for the final 10,000-Dalton filtrate. This analysis is consistent with the data in showing a failure to detect plutonium activity in the 10,000-Dalton to 0.1- μm colloid fraction, as shown in table 6. Colloidal-associated activity thus is calculated to contribute 43 percent of

total water plutonium 239 and 240 activity. Particulate-associated activity accounts for 22 percent of total water plutonium 239 and 240 activity, and the remainder resides in the dissolved phase, less than 10,000-Dalton in size. Although the errors associated with the analyses and these calculations of the mean values are large, they are a statistical measure of the central tendency represented by these data and indicate that plutonium 239 and 240 activity in the aqueous phase of this ground water is distributed primarily in particulate and colloidal size fractions (figure 14).

In contrast, size-fraction distribution as calculated directly from the particle retentate analysis (table 7) gives a markedly different result. For the 0.1- μm to 5- μm colloid phase, the error was much greater than the measurement, and in table 7 the activity is listed as not detected. As calculated from particle retentate analysis, the total plutonium 239 and 240 activity in the whole-water sample is calculated to be 0.0092 pCi/L (the sum of the particulate and colloidal fraction activities and 10,000-Dalton filtrate activity); the activity of the 10,000-Dalton to 0.1- μm colloidal fraction is calculated to be 0.00024 pCi/L. Colloidal-associated activity thus is calculated to contribute 3 percent of total water plutonium 239 and 240 activity. Of the particle-associated activity, most is expected to be in the greater than 5- μm fraction (7 percent of whole-water activity) and 3 percent in the 10,000-Dalton to 0.1- μm fraction. Thus, in absolute magnitudes of radioisotope loading and in estimates of size-fraction distribution, the two methods of calculation differ: the difference calculations indicate greater absolute plutonium activities in the colloidal phase and the whole-water sample and, significantly, indicate that particulate and colloidal phases are the dominant phases associated with plutonium in this ground water.

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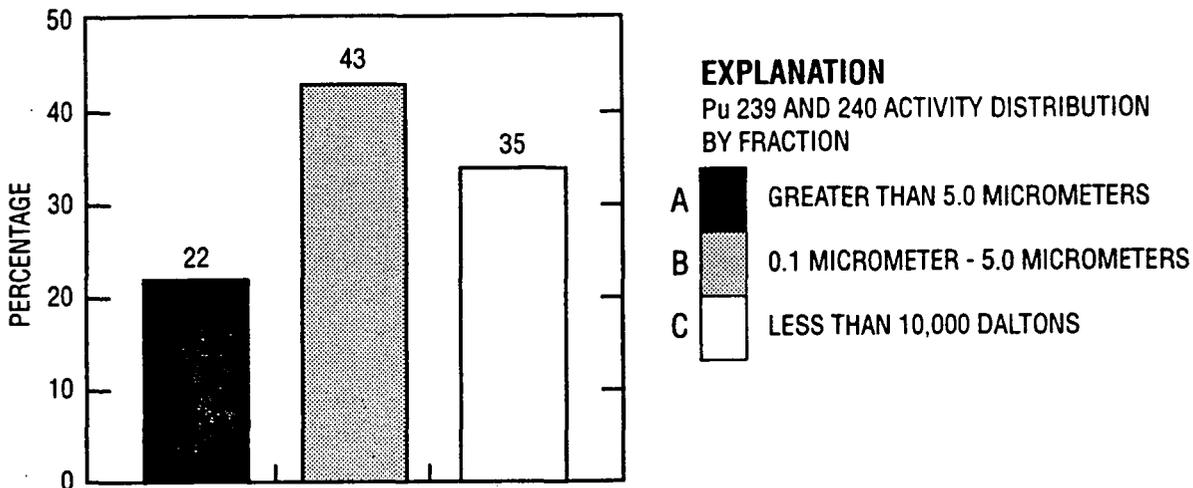


Figure 14. Plutonium 239 and 240 activity distribution by size fraction, calculated by difference in filtrate activities.

In contrast, the particle retentate analysis method predicts total and colloidal-associated activities to be more than an order of magnitude less than calculated by difference, and the predominant plutonium 239 and 240 activity is associated with the dissolved phase. The results are clearly in dispute. However, preliminarily, the results calculated by difference are the more realistic assessment of the fractionation results; therefore, colloidal-associated plutonium is the dominant plutonium phase in this system. This conclusion is justified because calculations of fractional mass calculated by difference are intrinsically not recovery-dependent; that is, it is assumed that plutonium-activity differences in the filtrates from various fractions result from particle associations, but quantitative recovery of these particles is not required. In contrast, calculations based on direct analysis of recovered particles are recovery-dependent, and although they represent direct evidence for colloid-associated nuclides, they are more subject to experimental error. It is likely that the discrepancies between the "difference" and "direct" data can be attributed to: (1) Error associated with the near-detection-limit nuclide levels in the sample; (2) differences in the counting statistics for particle retentates compared to larger volume solutions; and (3) incomplete recovery of the particles from the fractions. Filters used in the three filtration steps were not analyzed for radionuclides, and it is conceivable that particles and associated nuclides remained on the filters.

Inorganic Chemistry

Thermodynamic modeling of inorganic chemical analysis and measured physical properties of waters indicates that the major ion chemistry of water in well 1587 principally is controlled by carbonate-mineral dissolution, and the water can be classified as Ca + Mg + HCO₃ type. On the basis of these characteristics and by using typical anion data for water in well 1587 (EG&G, 1991a, b), the WATEQ4F model (Ball and Nordstrom, 1991) was used to identify the possible existence of colloidal phases at equilibrium in water from well 1587.

The mineral phases that are predicted to be supersaturated, their molecular formulas, and the calculated magnitude of supersaturation are listed in table 8. Given the constraints and limitations of such equilibrium speciation models, the listed phases need to be considered as only potentially present. With this provision, the calculations indicate a colloid population that includes aluminosilicates (clay minerals), calcium carbonate, aluminas, manganese oxyhydroxides, and iron oxyhydroxides can exist in water from well 1587. The calculations do not address the potential presence of organic colloids or mixed organic/mineral assemblages that are ubiquitous in most natural waters.

In the absence of a direct measure of particle mass in each of the fractions, calculations based on a model mineral assemblage (kaolinite, Fe(OH)₃, and CaCO₃) were undertaken to obtain estimates of the colloidal pool distribution in the sample from well 1587. By assigning a suite of three dominant mineral forms

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Generally, $\pm .5$ is considered "near equilibrium"

... sample of

Table 8. Mineral phases predicted by calculations by the model WATEQ to be supersaturated in ground water from Rocky Flats Plant near Denver, Colorado

[Log IAP/KT is the saturation index, defined as the logarithm of the ratio of ion activity product to solubility product constant; IAP, ion activity product; KT, solubility product constant]

Mineral name	Molecular formula	Log IAP/KT
Aluminosilicates		
Allophane (a)	$\{Al(OH)_3\} SiO_2$.788
Allophane (P)	$\{Al(OH)_3\} SiO_2$	1.672
Annite	$KFe_3 Al Si_3 O_{10}$	20.049
Beidellite	$(Na, K, Mg)_{0.33} Al_{2.33} Si_{3.678} O_{10} (OH)_2$	4.374
Halloysite	$Al_2 Si_2 O_5 (OH)_4$.397
Illite	$K_6 Mg_{0.25} Al_{2.3} Si_{3.5} O_{10} (OH)_2$	2.793
Kaolinite	$Al_2 Si_2 O_5 (OH)_4$	5.64
Muscovite	$KAl_3 Si_3 O_{10} (OH)_2$	9.376
Leonhardite	$Ca Al_2 Si_4 O_{12}$	2.526
Leonhardite	$Ca Al_4 Si_3 O_{24}$	13.31
Pyrophyllite	$Al_2 Si_4 O_{10} (OH)_2$	6.263
Montmorillonite	$Ca_{0.17} Al_{2.33} Si_{3.67} O_{10} (OH)_2$	4.185
Montmorial AB	$(H, Na, K)_{0.42} Mg_{0.45} Fe^{3+} Al_{1.47} Si_{3.82} O_{10} (OH)_2$	2.192
Montmorial BF	$(N NaK)_{0.28} Mg_{0.29} Fe^{3+} Al_{1.58} Si_{3.93} O_{10} (OH)_2$	2.942
Calcium carbonates		
Aragonite	$CaCO_3$.023
Calcite	$CaCO_3$.178
Aluminas		
Boehmite	$AlOOH$	1.478
Diaspore	$AlOOH$	3.31
Gibbsite	$Al(OH)_3$	2.16
Iron oxyhydroxides		
Cupric ferrite	$Cu Fe_2 O_4$	13.56
Cuprous ferrite	$Cu Fe O_2$	3.972
	$Fe(OH)_{2.7} Cl_{0.3}$	7.256
Ferrihydrite	$Fe(OH)_3$	2.38
Goethite	$Fe OOH$	8.271
Hematite	$Fe_2 O_3$	17.353
Maghemite	$Fe_2 O_3$	8.157
Magnetite	$Fe_3 O_4$	8.386
Manganese oxyhydroxides		
Birnessite	MnO_2	6.372
Bixbyite	$Mn_2 O_3$	4.687
Hausmannite	$Mn_3 O_4$.472
Manganite	$Mn OOH$	3.506
Pyrolusite	$Mn O_2$	6.065
Other minerals		
Barite	$BaSO_4$.666
Balsaluminite	$Al_4 (OH)_{10} SO_4 + 10H^+$	3.685
FCO_3 apatite	$Ca_{9.496} Na_{0.36} Mg_{0.144} (PO_4)_{4.8} (CO_3)_{1.2} F_{2.48}$	9.559

predicted to be present in this system by WATEQ4F calculations, molar concentrations of the predominant metals in the greater than 5- μm fraction were calculated and applied to the molecular formulas of the predicted mineral phases. This calculation provided an initial estimate of the concentrations and concentration percentages of each of the predicted mineral phases for each of the three particle-size phases collected. By assuming that this mineral assemblage is present in each of the three fractions and that it dominates the colloid pool, the colloid-pool concentration for each fraction was estimated. Results of the calculations are presented in table 11, and, in general, predict that iron oxide forms will predominate in the particulate (greater than 5- μm) fraction, calcium carbonate in the colloidal fraction (0.1- μm to 5- μm), and that clay minerals assume a larger fractional concentration in the colloidal fraction (10,000-Dalton to 0.1- μm). These approxima-

tions agree qualitatively with SEM-EDX observations. Estimated particle concentrations in ground water predict that the particle pool of particulates in the greater than 5- μm fraction will be 25 times greater than the colloid pool in the size range 0.1- μm to 5- μm , and 16 times greater than the colloid pool 10,000-Dalton to 0.1- μm . These figures refer to estimated masses of particles, and in terms of particle numbers, surface area, surface activity, and other variables related to the reactivity of the colloids, the two smaller size fractions could conceivably be of comparable or greater importance than the larger size fraction in facilitated transport of radionuclides.

Samples of filtrate fractions and particle fractions were analyzed for metals and major ions. Results of these analyses are presented in table 9 (inorganic analysis of filtrates) and table 10 (inorganic particle composition).

Table 9. Results of analysis for inorganic constituents in water from well 1587 at Rocky Flats Plant near Denver, Colorado

[μm , micrometer]

Constituent	Concentration (micrograms per liter)			
	Unfiltered	5- μm filtrate	0.1- μm filtrate	10,000-Dalton filtrate
Al	67	57	55	54
Sb	29	21	4	25
Ba	172	172	172	172
Be	1	1	1	1
Cd	3	1	1	1
Ca	123,000	122,000	123,000	123,000
Cr	13	13	12	13
Co	2	2	2	2
Cu	8	14	6	9
Fe	57	48	27	54
Mg	10,300	10,200	10,300	10,300
Mn	12	12	11	13
Ni	3	3	3	3
K	974	911	904	904
Ag	2	2	2	2
Na	6,870	6,810	6,750	6,770
V	7	7.5	7	8
Zn	41	23	34	33
Mo	3	3	3	3
Sn	14	14	14	14
Sr	443	438	443	443
Li	5	5	5	5

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Table 10. Inorganic composition of particulates and colloids in water from well 1587 at Rocky Flats Plant near Denver, Colorado

[>, greater than; μm , micrometer; $\mu\text{g/L}$, microgram per liter; conc., concentration]

Inorganic species	> 5- μm fraction		0.1- to 5- μm fraction		10,000-Dalton to 0.1- μm fraction		Total particulates and colloids	
	Conc. ($\mu\text{g/L}$)	percent by weight	Conc. ($\mu\text{g/L}$)	percent by weight	Conc. ($\mu\text{g/L}$)	percent by weight	Conc. ($\mu\text{g/L}$)	percent by weight
Al	4.71	3.83	0.67	4.84	2.02	5.84	7.40	4.32
Sb	0.25	0.20	0.89	6.44	2.18	6.30	3.32	1.94
Ba	0.58	0.47	0.06	0.43	0.09	0.26	0.73	0.43
Be	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cd	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ca	15.75	12.82	1.33	9.62	0.00	0.00	17.08	9.98
Cr	0.05	0.04	0.21	1.52	0.38	1.10	0.64	0.37
Co	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cu	0.60	0.49	0.35	2.53	11.01	31.84	11.96	6.98
Fe	91.63	74.58	1.78	12.87	2.66	7.69	96.07	56.09
Mg	2.01	1.64	0.78	5.64	0.00	0.00	2.79	1.63
Mn	0.43	0.35	0.02	0.14	0.01	0.03	0.46	0.27
Ni	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
K	2.02	1.64	4.37	31.60	9.87	28.54	16.26	9.49
Ag	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na	3.17	2.58	2.81	20.32	4.17	12.06	10.15	5.93
V	0.25	0.20	0.00	0.00	0.00	0.00	0.25	0.15
Zn	1.30	1.06	0.52	3.76	2.15	6.22	3.97	2.32
Mo	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sr	0.11	0.09	0.04	0.29	0.04	0.12	0.19	0.11
Li	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
TOTAL	122.86		13.83		34.58		171.27	

Table 11. Calculated distribution of model minerals kaolinite, calcium carbonate, and ferrihydrite in particle and colloid fractions in water from well 1587 at Rocky Flats Plant near Denver, Colorado

[>, greater than; μm , micrometer]

Mineral phase	Composition (percent by weight)		
	> 5- μm fraction	0.1- μm to 5- μm fraction	10,000-Dalton to 0.1- μm fraction
Kaolinite	8	31	66
Calcium carbonate	17	35	0
Ferrihydrite	75	34	33
	Concentration (milligram per liter)		
Estimated total	0.2	0.0092	0.0145

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Filtrate analysis by ICP-AES (table 9) indicates that water from well 1587 is a Ca + Mg type, which indicates that the parent materials predominate in the Rocky Flats Alluvium. As expected, concentrations of some inorganic species decrease as particle components are removed during processing from unfiltered to 10,000-Dalton filtered samples. Because particle concentrations are extremely small and inorganic analyte concentrations very large in this ground water, no attempt was made to calculate inorganic colloid composition by difference.

Inorganic Particle Composition

Table 10 lists the inorganic particle composition of each fraction and the weight percent of each analyzed element in the particulate fractions. Particle composition for each fraction also is shown in figures 15-17.

Greater Than 5- μ m Fraction: Iron (75 percent by weight in this fraction), calcium (13 percent), and aluminum (4 percent) are the most abundant metals in the greater than 5- μ m fraction. The predominance of iron in greater than 5- μ m particles in an oxic environment indicates the presence of iron oxyhydroxides, highly surface-active minerals with high affinity for actinide elements. Clay minerals (for example aluminosilicates) also are surface-active and potential phases for Pu and Am sorption.

0.1- μ m to 5- μ m Fraction: Particles in this fraction are predominantly potassium (32 percent), iron (13 percent), and sodium (20 percent). Aluminum contributes 5 weight percent to the fraction and calcium about 10 percent. As a signature element for various aluminosilicate minerals, the increase in colloidal potassium in this fraction and in the less than 0.1- μ m fraction indicate the presence of clay minerals in these fractions. Iron (oxyhydroxides) and calcium (carbonates) are present in this fraction in significant proportion, and sodium, also detected in certain clay minerals (for example montmorillonite) is significant in terms of weight percent in this fraction.

10,000-Dalton to 0.1- μ m Fraction: Copper (32 percent), potassium (29 percent), sodium (12 percent), iron (8 percent), aluminum (6 percent), and zinc (6 percent) are most abundant in this fraction. The presence of copper in increased concentrations in this fraction is indicative of the potential importance of organic materials in controlling the speciation of trace metals in this fraction. The 10,000-Dalton filter can retain organic material that corresponds to humic substances—higher molecular-weight organic acids that are biologically

refractory. These humic and fulvic acids can bind trace metals, with relatively high formation constants to form metal-humic complexes. Carboxylic acids are the dominant acidic functional groups in these substances, and the strong metal-binding sites could be bidentate and account for from 10 to 20 percent of the carboxylic acids. Thus, as the concentration of humic and fulvic acids increases in the retentate during ultrafiltration, the concentration of strongly complexed metals concurrently will increase. As the organic-acid concentration increases to the point where the ionized, acidic functional groups become substantial in terms of charge balance, the concentration of weakly bound, or territorially bound metal ions also will increase in the retentate; otherwise, the retentate would be more acidic than the filtrate, and this was not the result in this study. The 10,000-Dalton to 0.1- μ m retentate could have a high organic content that reflects the retention of humic and fulvic acids. If this is in fact the situation, the increased copper concentrations can be explained as copper fulvic-acid complexes and the increased sodium as the principal territorially bound counter-ion. Calculations indicate that a humic-carbon level of about 0.2-mg-carbon/L would be required to complex this concentration of copper, and this concentration of humic carbon is a reasonable figure for ground waters.

Metal Distribution in Particulate and Colloidal Fractions

Metal-distribution data are presented in figure 18 for the distribution of each element in the particulate/colloidal phases. These data represent the contribution of each element analyzed to the composition of the particles collected in each size fraction. In general, metals are concentrated either in the greater than 5- μ m fraction (Al, Ba, Ca, Fe, Mg, Mn, Sr, V) or in the 10,000-Dalton to 0.1- μ m fraction (Sb, Cr, Cu, K, Na, Zn). Although most of these elements also are present at lower concentrations in the 0.1- μ m to 5- μ m fraction, in no instance is this fraction predominant in terms of fractional weight percent of an element. Most metals are present at some concentration in each of the size fractions, the exceptions being calcium and magnesium, which are absent from the 10,000-Dalton to 0.1- μ m fraction, and vanadium, which is present in measurable quantity only in the greater than 5- μ m fraction.

Greater Than 5- μ m Fraction: Metals concentrated in this fraction include: (1) Aluminum, which is very abundant terrestrially as a component of clay minerals, in organic complexes, and rarely as free aqueous ions

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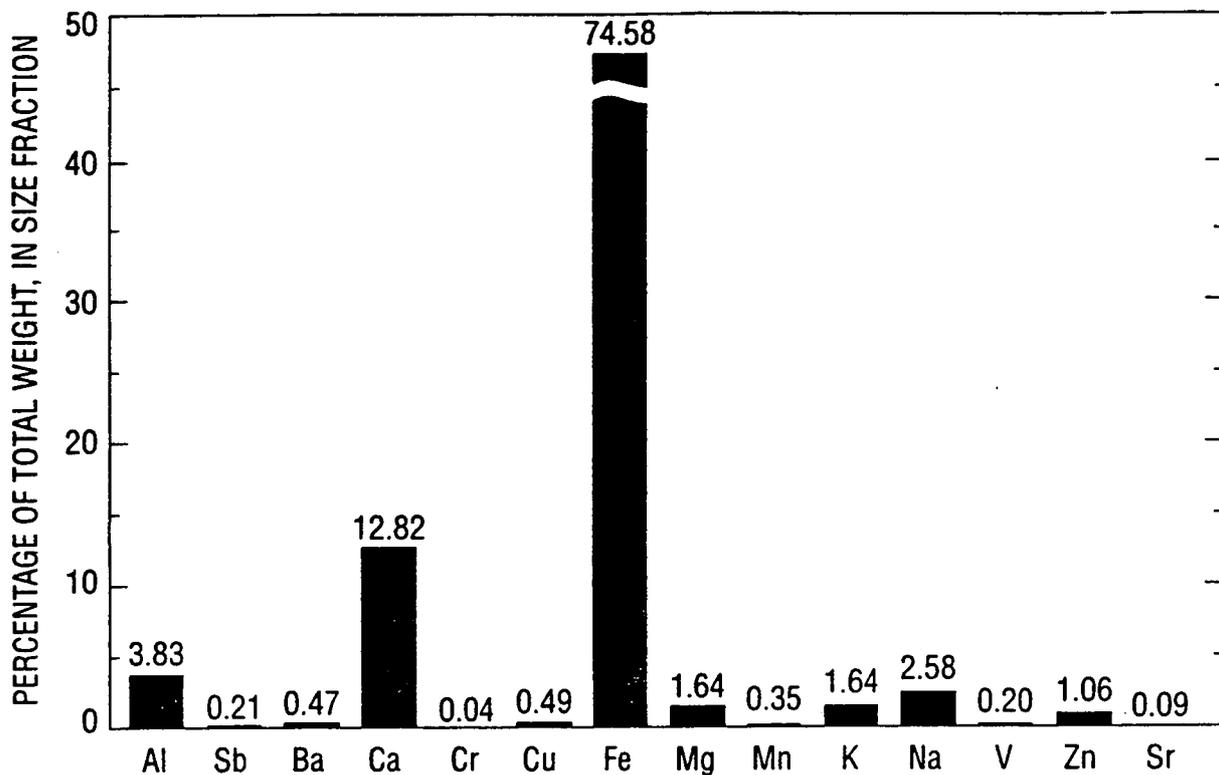


Figure 15. Inorganic particle composition in the greater than 5-micrometer fraction.

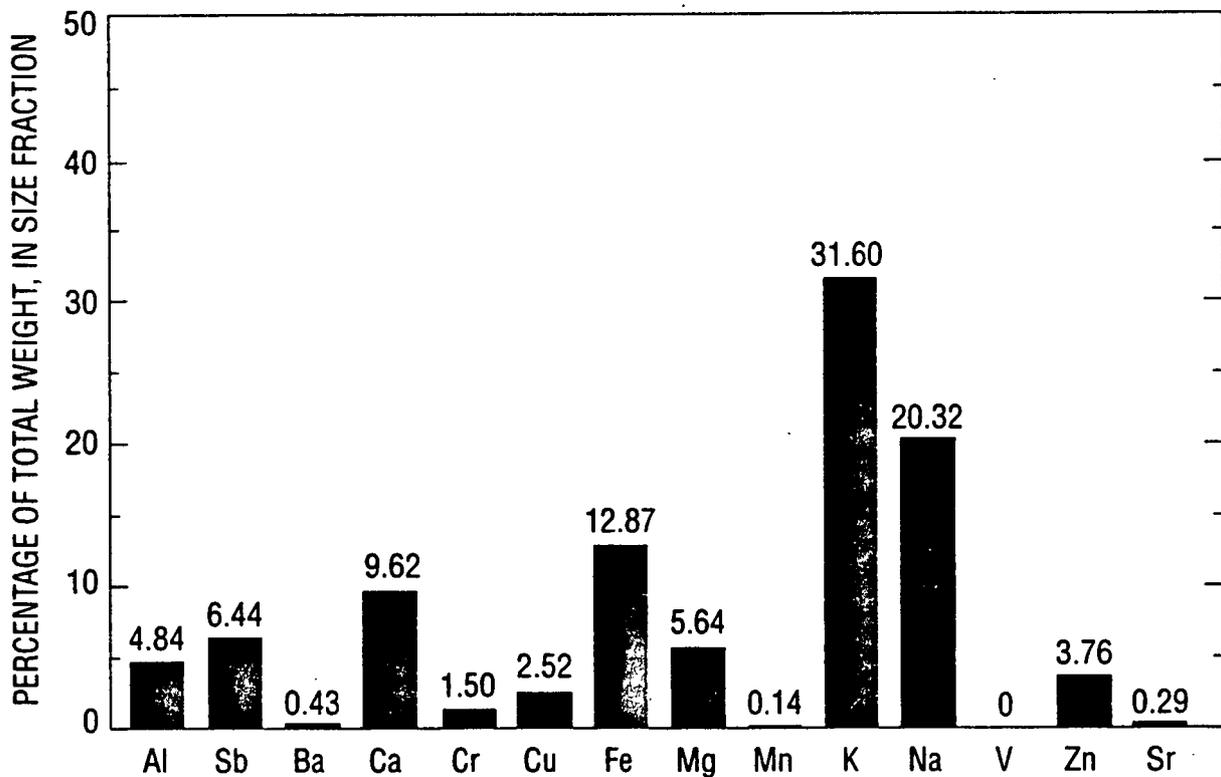


Figure 16. Inorganic particle composition in 0.1- to 5-micrometer fraction.

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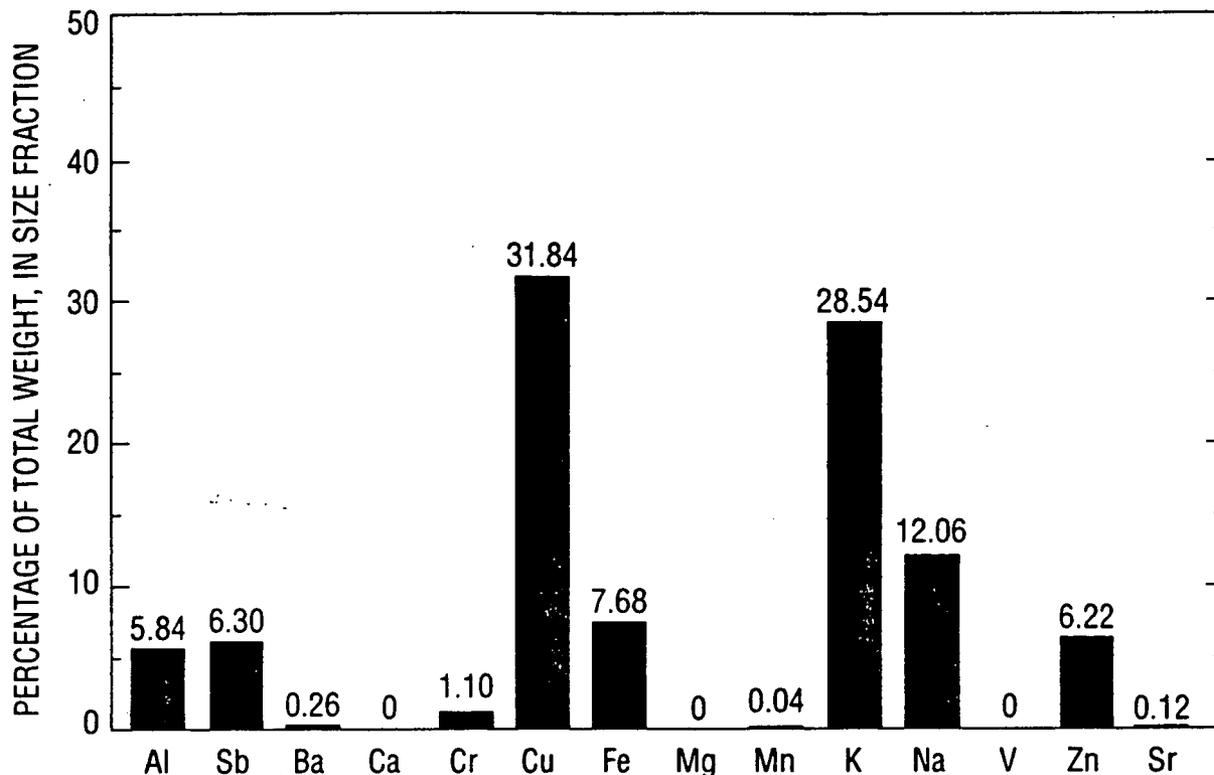


Figure 17. Inorganic particle composition in the 10,000-Dalton to 0.1-micrometer fraction.

in natural waters; (2) the alkaline earth metals barium, calcium, magnesium, and strontium, which are strongly adsorbed by ion exchange to clay minerals and other surfaces having exchange sites, including natural organic matter; (3) the redox-affected elements iron and manganese, which form insoluble and highly surface-active oxides and oxyhydroxides in oxic systems; and (4) vanadium, a transition metal, which has the tendency to form oxyanions.

10,000-Dalton to 0.1- μ m: Metals concentrated in this fraction include: (1) The transition metals copper and zinc, both of which form strong complexes with naturally occurring organic matter (humic and fulvic acids); (2) the alkali metals potassium and sodium, which, though they tend to react very differently in natural systems, participate in exchange reactions with silicate mineral structures; and (3) antimony and chromium.

Metal Distribution in the Unfiltered Sample

The metal distribution in the unfiltered sample including particulate, colloidal, and dissolved phases are shown in figure 19. Several metals show significant

distribution in the particulate/colloidal fraction, including iron (64 percent), aluminum (12 percent), antimony (12 percent), and zinc (11 percent). Vanadium (3 percent) and chromium (5 percent) had smaller concentration percentages in the particulate/colloidal fraction. Two metals, copper (59 percent of concentration distributed in the particulate/colloidal phase) and iron (64 percent of concentration distributed in the particulate/colloidal phase) exist primarily in particle fractions. The rest of the analyzed metals (barium, calcium, magnesium, manganese, potassium, sodium, and strontium) exist nearly totally in the dissolved phase of the unfiltered sample.

CONCLUSIONS

The Rocky Flats Plant near Denver, Colo., is administered by the U.S. Department of Energy. In 1990 the plant stopped plutonium processing and focused on contaminant assessment and potential remediation strategies. In regard to the potential for colloidal-facilitated transport in ground water at well 1587, the following conclusions are made.

1. As calculated from analysis of filtrates, 65 percent of plutonium 239 and 240 activity in the

25

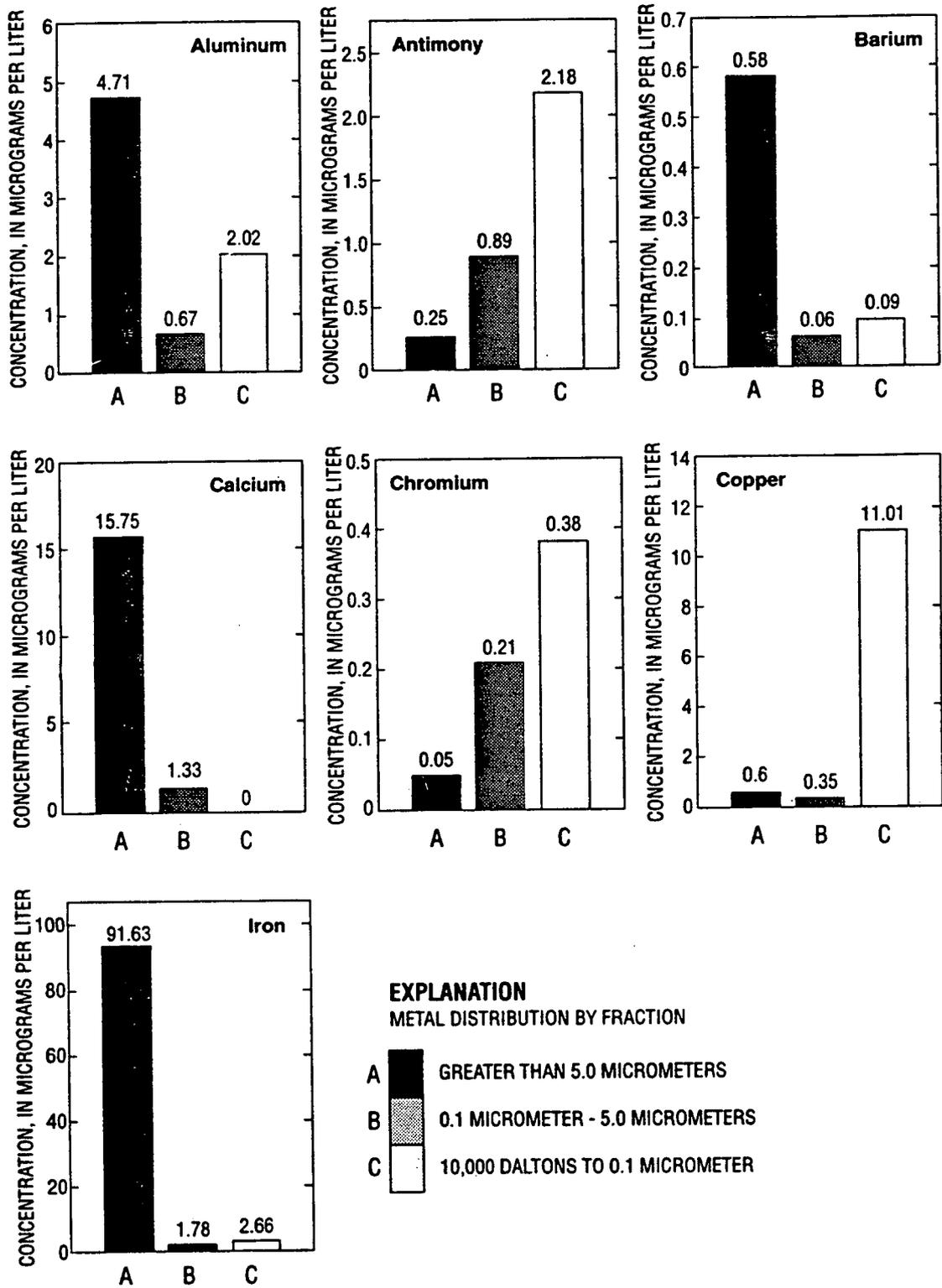
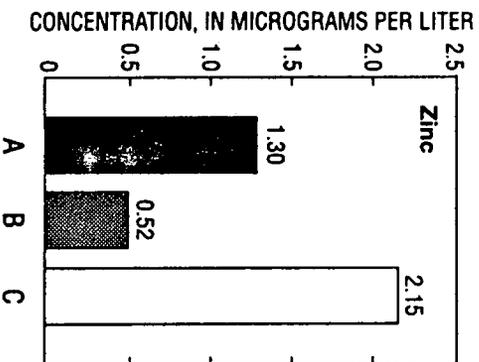
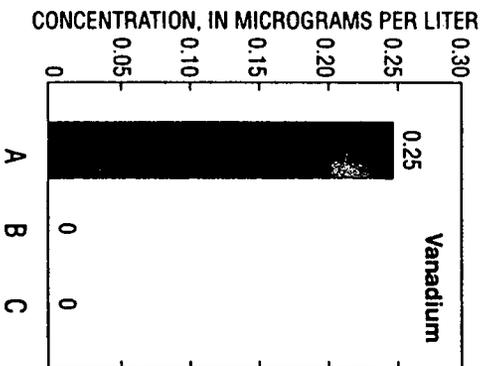
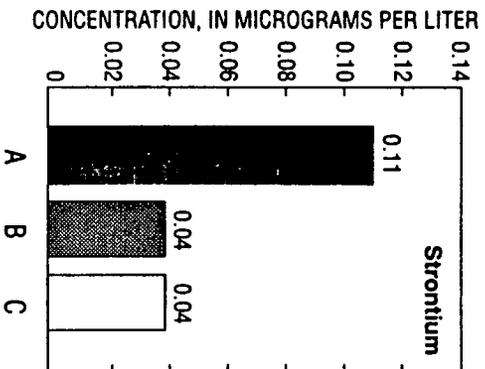
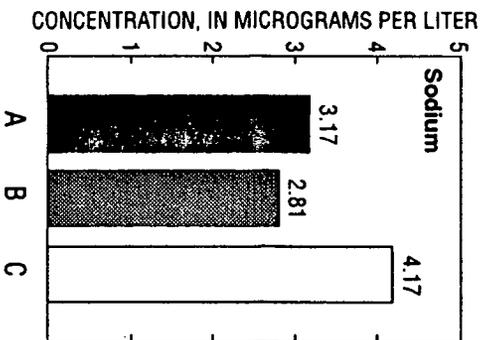
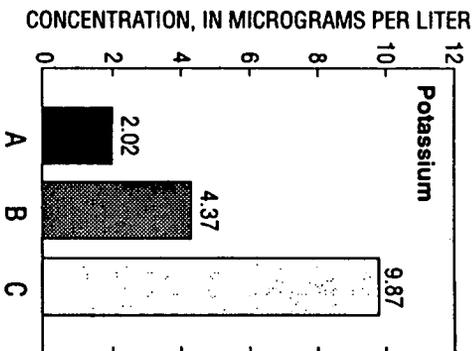
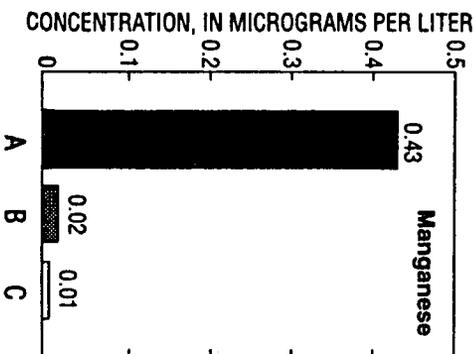
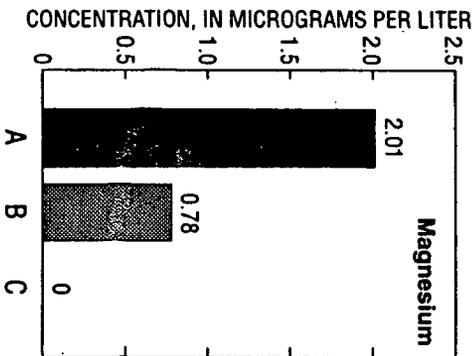


Figure 18a. Mass distribution and percent mass distribution of metals in the particulate and colloidal phase.

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EXPLANATION
METAL DISTRIBUTION BY FRACTION

- A GREATER THAN 5.0 MICROMETERS
- B 0.1 MICROMETER - 5.0 MICROMETERS
- C 10,000 DALTONS TO 0.1 MICROMETER

Figure 18b. Mass distribution and percent mass distribution of metals in the particulate and colloidal phase.

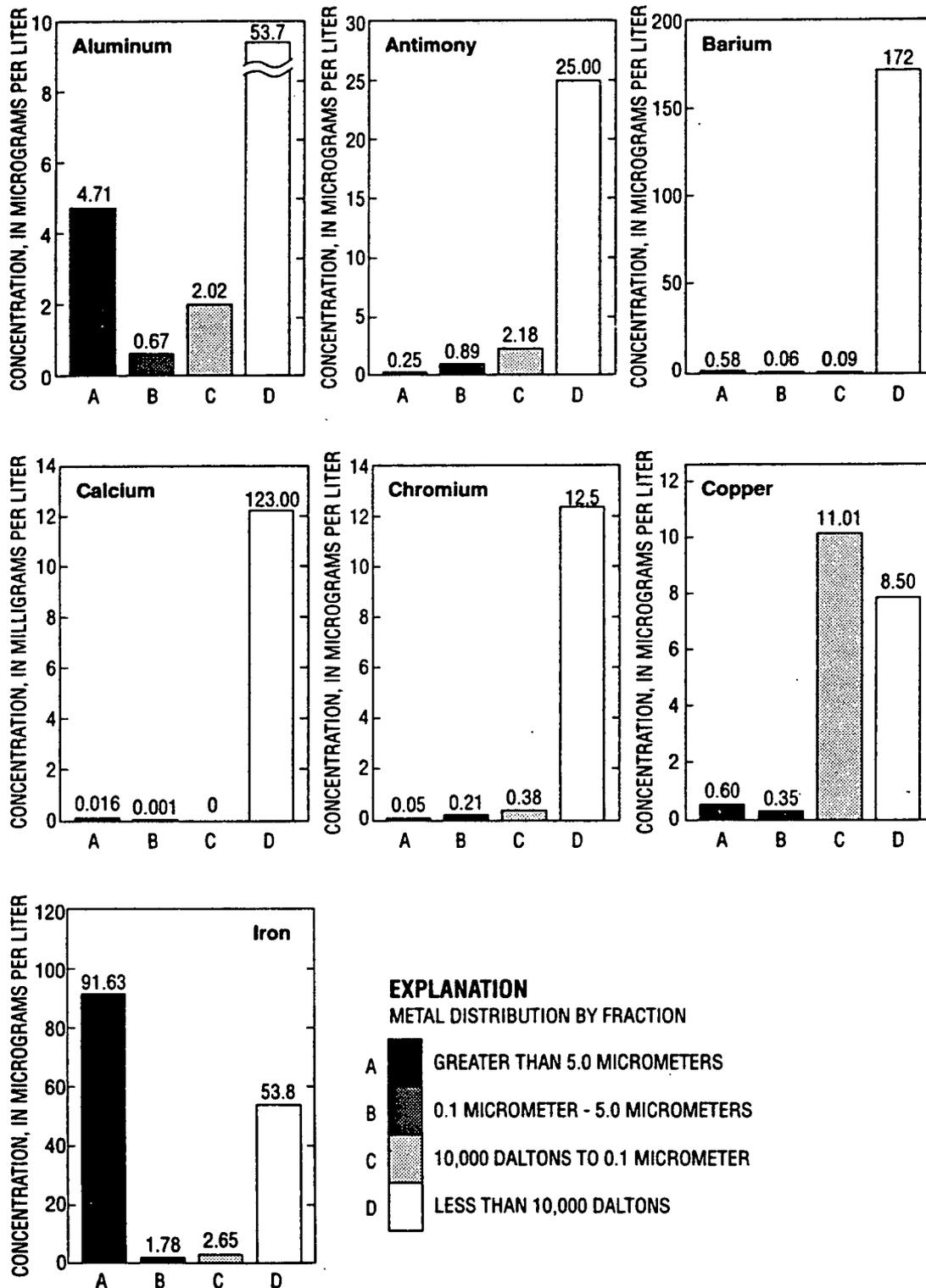


Figure 19a. Mass distribution and percent mass distribution of metals in the unfiltered water sample.

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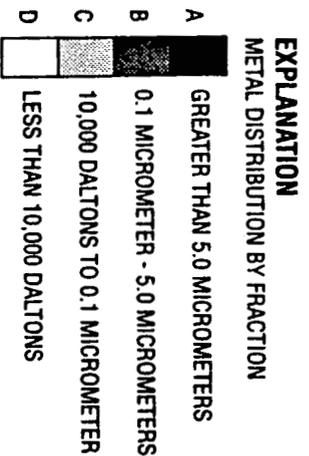
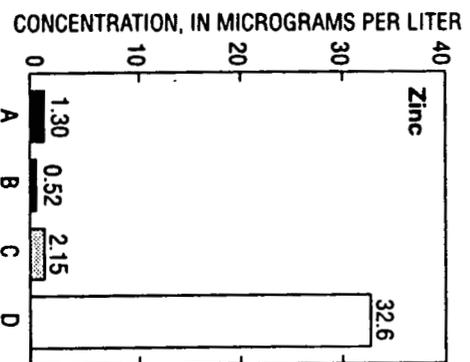
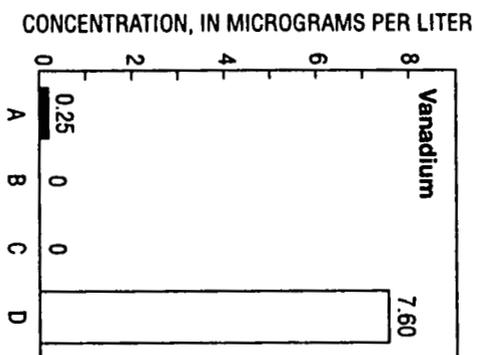
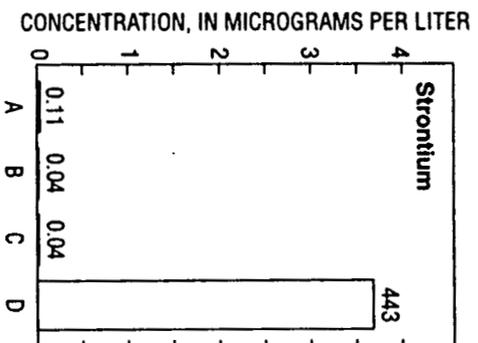
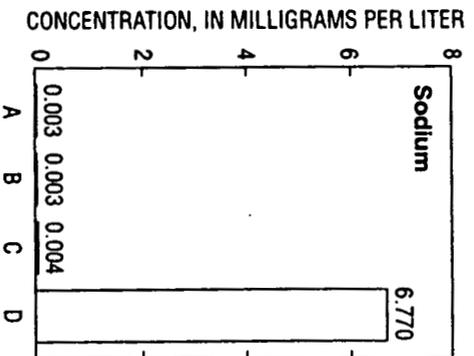
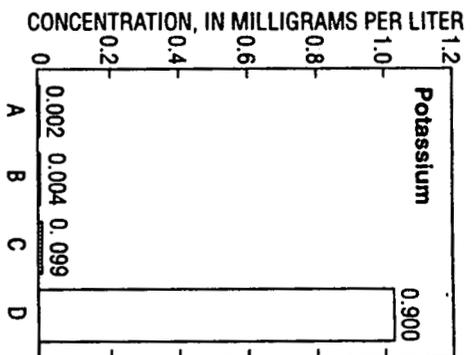
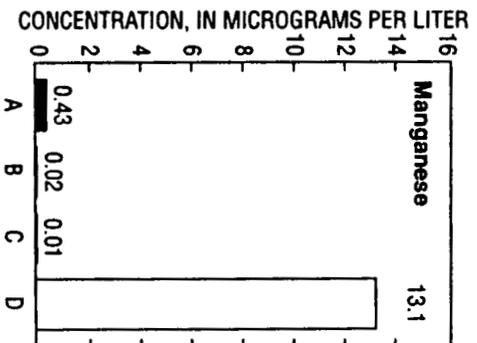
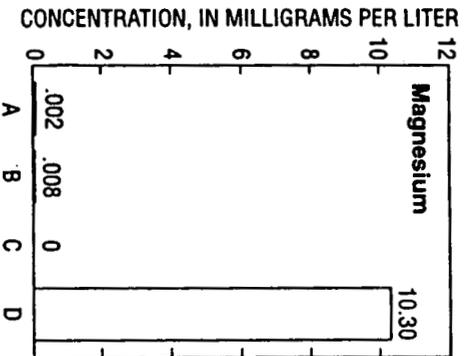


Figure 19b. Mass distribution and percent mass distribution of metals in the unfiltered water sample.

water sample from well 1587 was associated with particulate or colloidal phases; the remaining 35 percent of plutonium activity was associated with the dissolved phase. Particulate (22 percent) and colloidal (43 percent) fractions were determined to have significant activities in relation to the whole-water plutonium activity; therefore, particulate and colloidal forms could have the potential for facilitating the transport of plutonium in this ground water. Americium 241 and plutonium 238 isotope concentrations were too low to permit analysis.

2. Inorganic analysis, equilibrium modeling, and direct examination of the particulate/colloidal phases indicate the presence of mineral species that could facilitate the transport of radionuclides by reason of physiochemical characteristics that predict enhanced mobility in the aquifer and surface activity for potential sorption of actinides. In particular, iron oxyhydroxides predominant in the greater than 5- μm fraction are expected to be extremely surface-active and potential substrates for facilitated transport of actinides (elements with atomic numbers 89–103). In the 0.1- μm to 5- μm fraction, clay minerals or clay mineral particles become more evident; and in the 10,000-Dalton to 0.1- μm fraction, organic matter could be an important component of the colloidal pool. Clay minerals and natural organic substances contain surface-active groups that could affect actinide transport.

3. The high concentrations of copper and zinc in the smallest colloid fraction indicate a potential for organic (for example humic and fulvic acid) complexation of transition metals, and potentially of actinides, in this size fraction. Additional study is needed to determine if organic complexation occurs.

4. Colloid concentrations in the water at well 1587 are extremely low; estimated total concentrations are much less than 1 mg/L. This condition poses challenges to the isolation of sufficient quantities of colloids required for extensive characterization of the colloidal phase. Filtration of much larger volumes would be needed, and sample-integrity issues related to the necessarily long sampling and sample-storage times required would need to be addressed.

5. If transport of colloidal-associated actinides is episodic in response to leaching, the isolation of these colloids, to some extent, will depend on timing future sampling to coincide with rain or snowmelt infiltration.

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