FINAL REPORT ON PHASE SPECIATION OF PU AND AM FOR 'ACTINIDE MIGRATION STUDIES AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE'

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Significant Findings in FY98:

1) Pu and Am concentrations in filterpassing (^{2} 0.45µm) and filter-retained fractions in Walnut Creek at GS03, sampled in August 26-27, 1998, were very low (^{2}1 fCi/L), similar to concentrations of bomb fallout ^{239,240}Pu in natural aquatic systems all over the northern hemisphere.
2) ^{239,240}Pu/^{241}Am activity ratios in both particles as in the ^{2}0.45 µm filter-passing phases were low (i.e., ~1 or below), significantly lower than those in RFETS soils (~5-7).
3) Most Pu and Am (i.e., ~80-90 %) was associated with particles and colloids.
4) However, Pu activity concentrations in the particle-bound fraction was low (i.e., 0.1-0.3 pCi/g), considerably lower than was found in other creeks at RFETS (i.e., ~ 1pCi/g).
5) Significant fractions of Pu and Am which passed a 0.45µm filter were filtered out by 0.1µm and 100kDa ultrafilters, with, on average, about 50% of Pu passing a 100kDa ultrafilter.

Objectives:
1. **To examine the partitioning of Pu and Am between: 1) particulate, colloidal and ^{2}0.45 µm filter-passing phases.**
2. **To determine the chemical nature of the carrier phase (e.g., Fe, Mn, Al, C, etc.).**
3. **To provide data needed for to meet the DQO of the watershed erosion modeling efforts.**

Justification:
The phase speciation of Pu and Am during surface water exceedances is unknown. It is possible that Pu and Am during periods of elevated activity could have been in the colloidal state (i.e., <0.45 µm), which is not considered to be very bioavailable and where metal species complexed by functional groups of microparticles and macromolecules might have a limited lifetime (Wen et al., 1997a). Therefore, experimental determination of phase speciation of Am and Pu, and chemical characterization of the respective carrier phases, will provide the needed information for modeling surface water fate, transport and speciation of Pu and Am. Cross-flow ultrafiltration, CFUF, provides the means to extend our understanding of Pu and Am size distribution to the size realm of microparticles and macromolecules, and to confirm that the truly dissolved fraction of Pu and Am (<100 kDa), is small relative to other size fractions.

Analytical Methods:
The laboratory and analytical aspects include the following:
1) Cross Flow Ultrafiltration (CFUF) evaluation, and
2) surface water sampling.
CFUF evaluation consisted of ultrafiltering 20L of Galveston tap water containing added ^{240}Pu(IV or VI), and ^{242}Pu(IV) used as a yield tracer after two ultrafiltration systems (see below and appendix III). Surface water sampling was conducted only at the end of August 1998 due to delays in contract awards, and low rainfall during the summer months. The protocols of Guo and Santschi (1996,1997) and Wen *et al*. (1996, 1998) were followed for isolating colloidal and particulate phases of metals such as Pu, Am from surface waters by CFUF and ordinary filtration techniques. Chemical parameters which were measured

included total organic carbon (TOC), dissolved organic carbon (DOC), particulate organic carbon (POC), particulate organic nitrogen (PON), anions (fluoride, chloride, nitrate, phosphate, silicate, …), pH, Al, Fe and Mn of the water, and % organic carbon, in the particulate phases, according to Benoit *et al*. (1994), Guo and Santschi (1997) and Wen *et al*. (1998). In order to have enough water for actinide analysis, different collection and filtration methods were applied to different volumes of water (see Table IA).

A note on terminology: We denote all fractions with either the upper or lower size or nominal molecular weight cutoff limit, or both. The terms "dissolved", "filtrate" are ambigous, and the terms "retentate" and "permeate" are reserved for fractions which were retained by or had permeated an ultrafilter.

On August 26, 1998 140 liters of water was collected in several 25 liter carboys just above the little pond at GS03. The water was transported to the Colorado School of Mines (CSM) where the aliquots of whole water (two 2-liter bottles), were used for SPM, POC sampling and for total (particulate and ^{2}0.45 µm filter-passing) Pu and Am analyses. The remaining containers were emptied and mixed into a 55 gallon plastic drum. On August 27, 1998, 220 liters of water was collected in two types of carboys right at the GS03 site. The water was then transported to CSM and the two 2-liter whole water bottles were used for the determination of SPM and POC concentrations. Two 10-liter carboys were set aside for the determination of total Pu and Am, and the remaining water was emptied in the 55 gallon plastic drum. For both samples the water in the drum was mixed and pumped through 20 µm and 0.45 µm 10 inch MSI Calyx filter cartridges in series. The ^{2}0.45 µm filtered water was collected in 20 liter plastic containers. Forty liters of filtered water was set aside for ultrafiltration through a 0.1 micron (Amicon hollow fiber polysulfone) and a 100kDa (Amicon spiral wound regenerated cellulose) ultrafilter cartridges in parallel. Permeate (or ultra-filtrate) and retentate (colloids) fractions were collected for Pu and Am analysis. 20 liters of filtered water was also measured for Pu and Am and to serve as a ^{2}0.45µm fraction.

The methods for isotope separation were adapted from EPA Method 908.0 (1980), USDOE (1979), USEPA (1979), and Yamato (1982) as described briefly here. Each sample was acidified with concentrated nitric acid to pH <2 and allowed to sit for at least 16 hours. For each sample concentrated hydrochloric acid was added at 5ml/L and ^{243}Am and ^{242}Pu yield tracers were added. The samples were placed on a stir plate and 0.5 ml of 40 mg/ml Fe (III) carrier was added. The pH was measured and concentrated hydrochloric acid added until pH is <1. The sample was covered and stirred for 30 minutes and the pH measured again.

Once the pH was <1, concentrated ammonium hydroxide was added until turbidity remained constant then an additional 50 mls was added. The sample was again covered and stirred. After 30 minutes, the sample was removed from the stir plate, the stir bar removed and the precipitate was allowed to settle. The supernate was decanted until the precipitate slurry could be transferred to 250 ml centrifuge tubes. The samples were centrifuged for 30 minutes at 3000 rpm. Once the supernate was decanted, the precipitate was dissolved in 3 N HCl , transferred to Nalgene bottles and shipped to Texas A&M University. Once at Texas A&M University, the samples were evaporated and redissolved in concentrated HCl to which 75 mls of 9 N HCl and 2 ml saturated sodium nitrite were added. The samples were then run through a series of three anion exchange columns. The first separated the Am from the Pu fractions. The Pu was then microprecipitated on a filter, mounted on a stainless steel planchet and alpha counted. The Am fraction was carried through a methanolic anion exchange column followed by a TEVA resin column. The Am fraction was microprecipitated, mounted on a stainless steel planchette and alpha counted.

<u>Results and Discussion:</u>

Pu and Am results are shown in Table 1, Figures 1 and 2, and Appendix I, and ancillary parameter results in Tables 2-4 and Appendix II. Particulate, colloidal and ^{2}100kDa activities of both nuclides are extremely low. As a consequence, not all the measured activity concentrations are statistically significant, requiring that only a subset of the results reported in Table 1 will be useful for activity balance and phase speciation assessment (Figures 1 and 2). As is evident from Figure 1, most Pu was found in the particulate fraction, with only about 10% passing a 0.45 µm filter. A significant fraction of this "dissolved", ^{2}0.45 µm, fraction, however, was associated with colloids (Figure 2). The exact percentage in these fractions is not as certain as one might wish, as it critically depends on counting statistics and sample size, i.e., how representative each sample was. However, it was possible to take the best and largest samples from the second day, and come up with an approximate phase speciation scheme, shown in Figure 2 and Table 5, which includes the error limits. They indicate that even at these low concentrations, about half of the Pu is in different colloidal fractions, and half of Pu is found in the truly dissolved (^{2}100kDa) fraction.

Table 1. Results of ^{239,240}Pu and ^{241}Am analysis of surface water samples.

		^{239,240} Pu		^{241}Am		Pu/Am ratio	
Sample	**Size Fraction**	**pCi/L**	**±1 s**	**pCi/L**	**±1 s**	**pCi/L**	**±1 s**
8/26/96							
RF1	Total	LR	LR	0.0040	0.0009	ND	ND
RF3	>20μm	0.0037	0.0003	0.0051	0.0007	0.73	0.12
RF4	>5μm	0.0067	0.0003	0.0033	0.0001	2.01	0.11
RF5	0.45 – 20μm	0.0030	0.0016	0.0019	0.0020	1.59	1.89
RF6	0.45 – 5μm	0.0003	0.00003	0.0002	0.00002	1.76	0.31
RF2	<0.45μm	0.0009	0.0002	0.0006	0.0002	1.60	.56
RF8	0.1 – 0.45μm	0.0009	0.0002	0.0002	0.0002	5.05	6.24
RF7	<0.1μm	0.0005	0.0001	BD	BD	ND	ND
RF10	100kDa – 0.45μm	0.0003	0.0001	LR	LR	ND	ND
RF9	<100kDa	LR	LR	0.0002	0.0001	ND	ND
8/27/98A							
RF13	Total	0.0035	0.0004	HR	HR	ND	ND
RF15	>20μm	0.0043	0.0004	0.0024	0.0003	1.80	0.30
RF16	>5μm	0.0035	0.0001	0.0016	0.0001	2.17	0.11
RF17	0.45 – 20μm	0.0042	0.0007	0.0010	0.0004	4.38	2.02
RF18	0.45 – 5μm	0.0003	0.00003	LR	LR	ND	ND
RF14	<0.45μm (20L)	0.0004	0.0001	0.0002	0.0001	1.85	1.23
RF23	<0.45μm (5L)	0.0008	0.0004	0.0011	0.0006	0.70	0.51
RF21&22	<0.45μm (40L)	0.0004	0.0001	0.0003	0.0001	1.40	0.72
RF20	0.1 – 0.45μm	0.0003	0.0001	LR	LR	ND	ND
RF19	<0.1μm	0.0003	0.0001	0.0001	0.0001	2.53	3.43
8/27/98B							
RF24	Total	0.0015	0.0005	0.0005	0.0003	3.33	2.70
RF26	>20μm	0.0022	0.0003	0.0013	0.0003	1.74	0.46
RF28	0.1 – 0.45μm	0.0002	0.0001	0.0007	0.0002	0.23	0.15
RF27	<0.1μm	0.0002	0.0001	0.00004	0.0001	4.79	13.57
RF30	100kDa – 0.45μm	0.0001	0.0001	0.0001	0.0001	0.98	0.66
RF29	<100kDa	0.0004	0.0001	0.0006	0.0001	0.56	0.24

*) LR = Recovery too low; HR = Recovery to high; ND = No data; BD = below detection limit

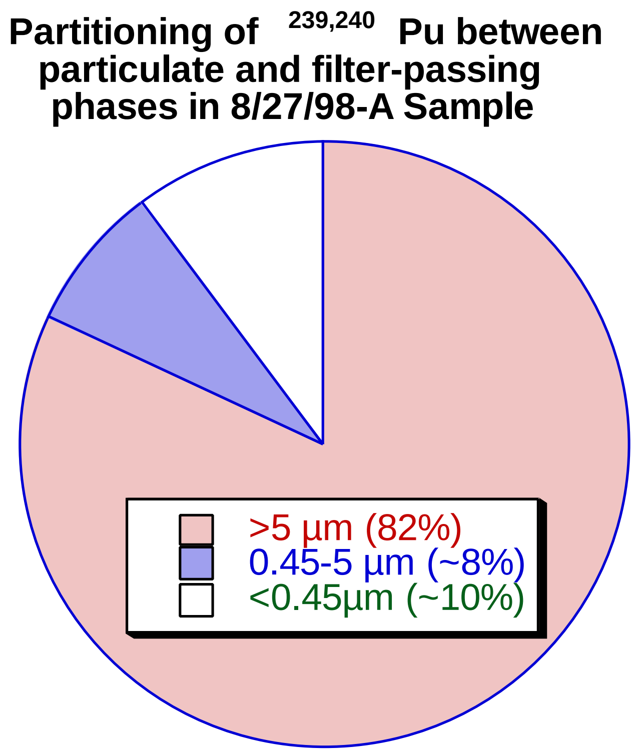

Figure 1. Representative partitioning of $^{239,240}Pu$ into different size fractions in waters from the Aug. 27 sampling expedition calculated from fractions with significant activities only.

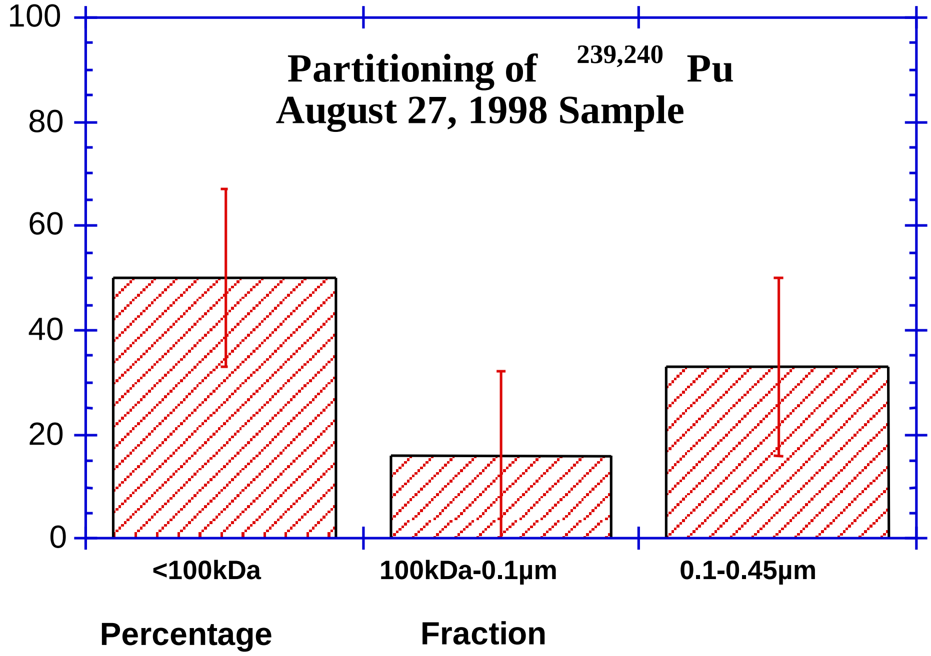

Figure 2. Representative partitioning of ^{239,240}Pu of the < 0.45mm fraction in waters from the Aug. 27 sampling expedition.

It is also noteworthy that these concentrations are 1-2 orders of magnitude below average ^{2}0.45 µm filter-passing Pu concentrations in shallow ground water samples reported by Harnish et al (1994, 1996) and RMRS (1996, 1997). The Pu concentrations we report here are of the same magnitude as those measured from bomb fallout in the 1970's and 1980's in different aquatic environments (Table 6). Furthermore, in our samples, Pu/Am ratios are consistently lower than the average soil ratio of 5-7. Low Pu/Am activity ratios for colloidal and ultrafilter-passing concentrations have also been reported by Harnish et al. (1993, 1996). In many cases, the Pu/Am ratios are not statistically significant, however, due to the fact that many values are close to the detection limit of the method as defined as three times the standard deviation at low values (i.e., ~ 0.0003 pCi/L for both actinides in ^{2}100 L of water). Low activity concentrations coincided with relatively high concentrations of algae and organic carbon in these waters, as is evident from the high organic carbon content of these suspended particles of 18% on 8/26/98 and 30% on 8/27/98. These high biomass and relatively low suspended particulate matter (SPM) concentrations (Table 2) were likely the result of the relatively high nutrient concentrations observed on Aug. 26. The next day, nutrient concentrations were, however, low again (Table 4), but pH remained high (i.e., pH~9.5-9.8, Table 3). Water temperature, Al, Fe and Mn concentrations (Table 3) were significantly lower the second day as well, demonstrating the differences between initial discharge water from just above the small pond (98826) and water sampled 24 hours after discharge began just below the small pond (98827). The August 26 sample was sampled on the first flush from the channel upstream from the small pond at GS03. The August 27 sample was collected on the second day of discharge, below the pond. The pond likely removed the suspended solids and associated constituents (e.g. metals) in the creek flow by settling. Nutrients and organic carbon transformations by aerobic processes might explain the subtle differences in the values measured between the two consecutive days.

Given the actinide concentrations in Table 1, and SPM concentrations given in Table 2, particle-water partition coefficients, Kd, of actinides can be calculated. Pu concentrations in suspended particulate matter were about 0.1 pCi/g on 8/26, and 0.3 pCi/g on 8/27, while "dissolved" (^{2}0.45 µm) Pu concentrations were 0.008 pCi/L on 8/26, and 0.004 pCi/L on 8/27. Kd values for Pu, calculated as the ratio of particulate Pu to "^{2}0.45 µm filter-passing" Pu were therefore 1.3×10^4 on 8/26/98, and 7.3×10^4 (L/Kg, or ml/g)on 8/27/98.

The Pu(IV), Pu(VI) and Am spike recovery experiments with tap water, shown in Figure 3, revealed loss rates of 5-10 %, and a prevalence (~70%) of Pu and Am, regardless of oxidation state, in the ^{2}100 kDa fraction (Figure 3). However, even in tap water, a significant fraction of actinide spike activity was found to be associated with colloids in the 0.1 - 0.45 µm and 100 kDa - 0.1 µm size fractions (Figure 3). While it would have been better to carry out this experiment with Walnut Creek water, it was not possible due to logistical reasons.

Table 2. Summary of data of dissolved organic carbon (DOC), particulate organic carbon (POC) and nitrogen (PON), and suspended particulate matter (SPM) concentrations in water samples collected on August 26-27, 1998.

Sample ID	DOC (mg-C/l)	POC (mg-C/L)	PON (mg-N/L)	POC (mg-C/g)	PON (mg-N/g)	SPM-1 (mg/l)*	SPM-2 (mg/L)*
8-26-98	12.3±0.6	13.1±1.8	1.92±0.24	177±17	26±3	87.7±15	75±18
8-27-98A	14.0±1.0	10.1±0.3	1.37±0.01	304±5	41±1	27.1±5.6	33.2±0.6
8-27-98B	11.9±1.5	9.8±0.5	1.31±0.08	296±11	40±1	34.5±0.3	33.0±3.1

*) Data of SPM-1 and SPM-2 are derived from Nuclepore filters ^{2}0.45 µm and GF/F filters, respectively.

Table 3. Data of trace metal (Fe, Al, and Mn) concentrations (in µg/L) and water temperature (°C), dissolved oxygen, DO (mg/l), and pH in ^{2}0.45 µm water samples collected on August 26-27, 1998.

Sample ID	Fe (µg/L)	Al (µg/L)	Mn (µg/L)	Water Temp (°C)	DO (mg/L)	pH	Alk (meq/l)
8-26-98	43.4±1.4	85.2±3.9	43.3±1.8	25	7.7	9.5	-
8-27-98-A	13.8±1.3	21.7±5.7	22.0±5.8	19.6	8.2	9.8	2
8-27-98-B	12.1±4.1	22.2±5.9	20.9±5.4	19.4	7.9	9.8	2

Table 4. Data of anion (F, Cl, NO_3, HPO_4, Si) concentrations in ^{2}0.45 µm water samples collected on August 26-27, 1998.

Sample ID	F (mg/l)	Cl (mg/L)	NO_3 (mg/L)	HPO_4 (mg/L)	Si (mg/L)
8-26-98	0.42±0.005	91.4±3.2	4.29±0.005	0.79±0.01	5.5±0.05
8-27-98-A	0.34±0.002	92.7±1.2	<0.1	<0.1	5.2±0.07
8-27-98-B	0.33±0.009	93.3±0.4	<0.1	<0.1	4.9±0.01

Table 5: Example of Mass Balance for ^{239,240}Pu sampled on 8/27/98.

		^{239,240}Pu	
		pCi/L	±1σ
	Total (measured)	0.0035	0.0004
Particulate	>5µm	0.0035	0.0001
	0.45 – 5µm	0.0003	0.0000
	>0.45µm (calculated)	0.0038	0.0001
^{2}0.45 µm filter-passing	<0.45µm cartridge	0.0004	0.0001
	Total (calculated)	0.0042	0.0001

Table 6. Comparison of ^{2}0.45 µm filter-passing concentrations of $^{239,240}Pu$ in different water bodies.

^{2}**0.45 µm filter-passing Plutonium (fCi/L)**	**Water Body**	**Reference**
O.4-0.8	^{2}0.45 µm, Walnut Creek at GS03	This work
0.3-0.4	^{2}0.1µm, Walnut Creek at GS03	"
37	Average total concentration at Walnut Creek at GS03	RF/RMRS-97-131.UN
0.1-0.5	^{2}0.45 µm, Great Lakes, 1977	Wahlgren et al., 1980
0.3-1.1	^{2}0.45 µm, Narragansett Bay, 1976-1978	Santschi et al., 1980
0.3-1.1	New York Bight, 1976-1977	Santschi et al., 1980
0.1-0.3	^{2}0.7 µm, Ob and Yenissey Rivers, 1995	Schwantes, Baskaran and Santschi, 1998, in prep.

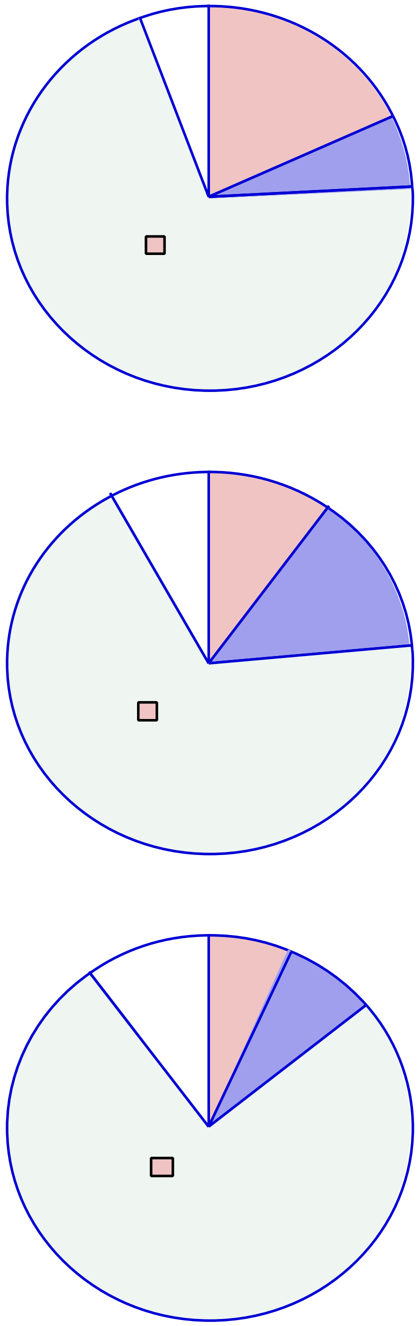

Figure 3. Partitioning of ^{240}Pu and ^{241}Am between different size fractions in tap water passing a 0.45µm filter during spiked laboratory experiments.

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Appendices

Appendix I: Rocky Flats Sample Identification, and Details of Actinide and Ancillary Parameter Results.

Table I-A. Sample Identification

TAMU ID	Sample	Size Fraction	Type	Volume (L)
RF1	98826	Total	Precipitate	4
RF2		<0.45μm	Precipitate	20
RF3		>20μm	Filter paper	10
RF4		>5μm	Cartridge	122
RF5		0.45 – 20μm	Filter paper	
RF6		0.45 – 5μm	Cartridge	122
RF7		<0.1μm	Precipitate	20
RF8		0.1 – 0.45μm	Precipitate	20
RF9		<100kDa	Precipitate	40
RF10		100kDa – 0.45μm	Precipitate	40
RF11		Blank	Precipitate	
RF13	98827A	Total	Precipitate	10
RF14		<0.45μm	Precipitate	20
RF15		>20μm	Filter paper	12
RF16		>5μm	Cartridge	180
RF17		0.45 – 20μm	Filter paper	6
RF18		0.45 – 5μm	Cartridge	180
RF19		<0.1μm	Precipitate	20
RF20		0.1 – 0.45μm	Precipitate	20
RF21+RF22		<0.45μm	Precipitate	40
RF23		<0.45μm	Precipitate	5
RF24	98827B	Total	Precipitate	10
RF25		<0.45μm	Precipitate	20
RF26		>20μm	Filter paper	12
RF27		<0.1μm	Precipitate	20
RF28		0.1 – 0.45μm	Precipitate	20
RF29		<100kDa	Precipitate	40
RF30		100kDa – 0.45μm	Precipitate	40
RF31		Blank	Precipitate	

Table I-B: ^{239,240}Pu and ^{241}Am activities in different size fractions

^{239,240}Pu														
PARTICULATE														
Sample	whole water		> 20 micron		0.45-20 micron		>0.45 (filters)*		> 5 micron		0.45-5micron		>0.45 micron(cartridge)*	
	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-
98826	-	-	0.0037	0.0003	0.0030	0.0016	0.0067	0.0016	0.0067	0.0003	0.0003	0.00003	0.0070	0.0003
98827A	0.0035	0.0004	0.0043	0.0004	0.0042	0.0007	0.0085	0.0009	0.0035	0.0001	0.0003	0.00003	0.0038	0.0001
98827B	0.0015	0.0005	0.0022	0.0003	-	-	-	-	-	-	-	-	-	-
=0.45 µm FRACTION														
Sample	< 0.45 micron		0.1- 0.45 micron*		< 0.45 micron* - 100kDa		0.1 - 0.45 micron		< 0.1 micron		100kDa - 0.45 micron		< 100kDa	
	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-
98826	0.0009	0.0002	0.0014	0.0002	-	-	0.0009	0.0002	0.0005	0.0001	-	-	-	-
98827A	0.0004	0.0001	0.0006	0.0001	0.0004	0.0001	0.0003	0.0001	0.0003	0.0001	-	-	-	-
98827B	-	-	0.0004	0.0001	0.0005	0.0001	0.0001	0.000	0.0001	0.0001	0.0001	0.0001	0.0004	0.0001
^{241}Am														
PARTICULATE														
Sample	whole water		> 20 micron		0.45-20 micron		>0.45 (filters)*		> 5 micron		0.45-5micron		>0.45 micron(cartridge)*	
	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-
98826	0.0040	0.0009	0.0051	0.0007	0.0019	0.0020	0.0070	0.0021	0.0033	0.0001	0.0002	0.00002	0.0035	0.0001
98827A	-	-	0.0024	0.0003	0.0010[m]	0.0004[m]	0.0034	0.0005	0.0016	0.0001	-	-	-	-
98827B	0.0005	0.0003	0.0013	0.0003	-	-	-	-	-	-	-	-	-	-
=0.45 µm FRACTION														
Sample	< 0.45 micron		0.1- 0.45 micron*		< 0.45 micron* - 100kDa		0.1 - 0.45 micron		< 0.1 micron		100kDa - 0.45 micron		< 100kDa	
	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-	pCi/L	+/-
98826	0.0006	0.0002	-	-	-	-	0.0002	0.0002	-	-	-	-	0.0002	0.0001
98827A	0.0002	0.0001	-	-	0.0003[m]	0.0001[m]	-	-	0.0001	0.0001	-	-	-	-
98827B	-	-	0.0007	0.0002	0.0007	0.0001	0.0007	0.0002	0.00004	0.0001	0.0001	0.0001	0.0006	0.0001
* = calculated by addition														
- = no data														
[m] = measured value														
BD = below detection limit														

Table I-C: ^{239,240}Pu and ^{241}Am counting data (+/- signifies ±1 s)

			volume		Detector	Detector Background (cpm)			
Sample	Size Fraction	ID	(L)	Detector	Efficiency	^{242}Pu	+/-	^{240}Pu	+/-
98826	whole water	RF1Pu	4	1	0.20	0.0025	0.0025	0.0003	0.0002
	<0.45 µm	RF2Pu	20	10	0.24	0.0010	0.0010	0.0007	0.0003
	>20 µm	RF3Pu2	10	7	0.20	0.0012	0.0012	0.0003	0.0002
	>5 µm	RF4Pu	122	2	0.20	0.0017	0.0017	0.0002	0.0002
	0.45 – 20 µm	RF5Pu	1	2	0.20	0.0017	0.0017	0.0002	0.0002
	0.45 - 5 µm	RF6Pu	122	3	0.20	0.0010	0.0010	0.0008	0.0004
	<0.1 µm	RF7Pu	20	11	0.22	0.0015	0.0015	0.0005	0.0003
	0.1 - 0.45 µm	RF8Pu	20	8	0.23	0.0008	0.0008	0.0003	0.0002
	<100kDa	RF9Pu	40	12	0.24	0.0006	0.0006	0.0006	0.0003
	100kDa - 0.45 µm	RF10Pu	40	4	0.19	0.0007	0.0007	0.0002	0.0002
	Blank spike	RF11Pu	1	4	0.19	0.0013	0.0013	0.0003	0.0002
98827A	whole water	RF13Pu	10	5	0.25	0.0002	0.0002	0.0002	0.0002
	<0.45 µm	RF14Pu	20	5	0.25	0.0002	0.0002	0.0002	0.0002
	>20 µm	RF15Pu	12	6	0.19	0.0005	0.0005	0.0008	0.0004
	>5 µm	RF16Pu	180	11	0.22	0.0015	0.0015	0.0005	0.0003
	0.45 – 20 µm	RF17Pu	6	7	0.20	0.0010	0.0010	0.0007	0.0003
	0.45 - 5 µm	RF18Pu	180	8	0.23	0.0015	0.0015	0.0005	0.0003
	<0.1 µm n	RF19Pu	20	6	0.19	0.0007	0.0007	0.0013	0.0005
	0.1 - 0.45 µm	RF20Pu	20	5	0.25	0.0002	0.0002	0.0002	0.0002
	<100kDa	RF21Pu	40	6	0.19	0.0008	0.0008	0.0013	0.0005
	100kDa - 0.45 µm	RF22Pu	40	7	0.20	0.0013	0.0013	0.0003	0.0002
	<0.45 µm	RF23Pu	5	8	0.23	0.0000	0.0000	0.0002	0.0002
98827B	whole water	RF24Pu	10	9	0.23	0.0005	0.0005	0.0008	0.0004
	>20 µm	RF26Pu	12	9	0.23	0.0006	0.0006	0.0006	0.0003
	<0.1µm	RF27Pu	20	9	0.23	0.0005	0.0005	0.0008	0.0004
	0.1 - 0.45 µm	RF28Pu	20	10	0.24	0.0010	0.0010	0.0007	0.0003
	<100kDa	RF29Pu	40	11	0.22	0.0015	0.0015	0.0005	0.0003
	100kDa - 0.45 µm	RF30Pu	40	12	0.24	0.0006	0.0006	0.0006	0.0003
	Blank spike	RF31Pu	1	10	0.24	0.0010	0.0010	0.0007	0.0003

Table I-C cont.

			count time	^{242}Pu		Background corrected		^{239,240} Pu		Background corrected		^{242}Pu		^{239,240} Pu	
Sample	Size Fraction	ID	(min)	counts	+/-	counts	+/-	counts	+/-	counts	+/-	cpm	+/-	cpm	+/-
98826	whole water	RF1Pu	6697	77	8.77	60	19.07	121	11	119	11.12	0.0090	0.0028	0.0177	0.0017
	<0.45 µm	RF2Pu	6831	1540	39.22	1533	39.82	49	7	44	7.37	0.2244	0.0058	0.0065	0.0011
	>20 µm	RF3Pu2	16795	2690	51.87	2670	55.53	227	15.07	221	15.59	0.1590	0.0033	0.0132	0.0009
	>5 µm	RF4Pu	6697	1250	35.37	1239	37.13	2250	47.4	2249	47.41	0.1850	0.0055	0.3358	0.0071
	0.45 – 20 µm	RF5Pu	6171	1310	36.19	1300	37.65	14	3.74	13	3.88	0.2106	0.0061	0.0021	0.0006
	0.45 - 5 µm	RF6Pu	6171	1280	35.71	1274	36.25	103	10.15	98	10.41	0.2064	0.0059	0.0158	0.0017
	<0.1 µm	RF7Pu	6831	1253	35.4599	1243	36.94	24	4.8984	21	5.29	0.1819	0.0054	0.0030	0.0008
	0.1 - 0.45 µm	RF8Pu	6697	1200	34.64	1194	35.10	38	6.16	36	6.36	0.1784	0.0052	0.0053	0.0010
	<100kDa	RF9Pu	6831	91	9.5368	87	10.32	12	3.4644	8	3.99	0.0127	0.0015	0.0012	0.0006
	100kDa - 0.45 µm	RF10Pu	6697	2710	52.04	2705	52.24	49	7	48	7.09	0.4040	0.0078	0.0071	0.0011
	Blank spike	RF11Pu	6171	1190	34.45	1182	35.44	2	1.41	0	2.04	0.1915	0.0057	0.0000	0.0003
98827A	whole water	RF13Pu	6699	1250	35.28	1249	35.30	73	8.54	72	8.61	0.1864	0.0053	0.0107	0.0013
	<0.45 µm	RF14Pu	3882	1010	31.72	1009	31.73	14	3.74	13	3.80	0.2600	0.0082	0.0034	0.0010
	>20 µm	RF15Pu	5696	1080	32.79	1077	32.92	129	11.36	124	11.56	0.1891	0.0058	0.0218	0.0020
	>5 µm	RF16Pu	9858	2340	48.37	2325	50.63	3220	56.71	3215	56.78	0.2359	0.0051	0.3261	0.0058
	0.45 – 20 µm	RF17Pu	6163	826	28.74	820	29.41	50	7.07	46	7.37	0.1330	0.0048	0.0074	0.0012
	0.45 - 5 µm	RF18Pu	6163	1320	36.3	1311	37.48	155	12.45	152	12.58	0.2126	0.0061	0.0246	0.0020
	<0.1 µm	RF19Pu	3882	980	31.262	977	31.37	13	3.6062	8	4.05	0.2517	0.0081	0.0020	0.0010
	0.1 - 0.45 µm	RF20Pu	7216	2330	48.22	2329	48.24	26	5.1	25	5.24	0.3227	0.0067	0.0034	0.0007
	<100kDa	RF21Pu	7216	1470	38.33	1464	38.81	42	6.48	32	7.34	0.2029	0.0054	0.0045	0.0010
	100kDa - 0.45 µm	RF22Pu	6699	830	28.81	821	30.19	19	5.0787	4	5.32	0.1226	0.0045	0.0007	0.0008
	<0.45 µm	RF23Pu	6699	1060	32.51	1060	32.51	8	2.83	7	3.05	0.1582	0.0049	0.0010	0.0005
98827B	whole water	RF24Pu	6699	653	25.55	650	25.77	22	4.69	16	5.33	0.0970	0.0038	0.0024	0.0008
	>20 µm	RF26Pu	6165	1320	36.39	1316	36.56	80	8.94	76	9.12	0.2135	0.0059	0.0124	0.0015
	<0.1 µm	RF27Pu	16777	3950	62.87	3942	63.44	42	6.48	28	9.05	0.2349	0.0038	0.0017	0.0005
	0.1 - 0.45 µm	RF28Pu	6699	1700	41.28	1693	41.83	14	3.74	9	4.37	0.2528	0.0062	0.0014	0.0007
	<100kDa	RF29Pu	6699	566	23.79	556	25.87	16	4	13	4.45	0.0830	0.0039	0.0019	0.0007
	100kDa - 0.45 µm	RF30Pu	6699	1610	40.1	1606	40.29	22	4.69	18	5.07	0.2398	0.0060	0.0027	0.0008
	Blank spike	RF31Pu	16779	4870	69.81	4853	71.84	13	3.61	2	6.71	0.2892	0.0043	0.0001	0.0004

Table I-C cont.

			^{242}Pu tracer added		^{239,240} Pu		Blank corrected				^{239,240} Pu			
Sample	Size Fraction	ID	(dpm)	+/-	dpm	+/-	dpm	+/-	pCi	+/-	pCi/l	+/-	% recovery	+/-
98826	whole water	RF1Pu	1.35	0.01	2.6685	0.8830	2.6681	0.8830	1.2128	0.4014	0.3032	0.1003	3.31	1.05
	<0.45 µm	RF2Pu	1.35	0.01	0.0391	0.0066	0.0386	0.0068	0.0176	0.0031	0.0009	0.0002	68.11	1.84
	>20 µm	RF3Pu2	0.996	0.01	0.0826	0.0061	0.0821	0.0063	0.0373	0.0029	0.0037	0.0003	78.00	1.72
	>5 µm	RF4Pu	0.996	0.01	1.8082	0.0663	1.8078	0.0663	0.8217	0.0301	0.0067	0.0002	90.70	2.80
	0.45 – 20 µm	RF5Pu	0.996	0.01	0.0099	0.0030	0.0095	0.0035	0.0043	0.0016	0.0043	0.0016	103.25	3.09
	0.45 - 5 µm	RF6Pu	0.996	0.01	0.0765	0.0084	0.0760	0.0086	0.0345	0.0039	0.0003	0.0000	104.48	3.07
	<0.1 µm	RF7Pu	1.35	0.01	0.0223	0.0058	0.0219	0.0061	0.0099	0.0028	0.0005	0.0001	62.23	1.91
	0.1 - 0.45 µm	RF8Pu	1.35	0.01	0.0404	0.0073	0.0399	0.0075	0.0182	0.0034	0.0009	0.0002	56.80	1.72
	<100kDa	RF9Pu	1.37	0.01	0.1267	0.0645	0.1262	0.0646	0.0574	0.0293	0.0014	0.0007	3.85	0.46
	100kDa - 0.45 µm	RF10Pu	1.35	0.01	0.0239	0.0036	0.0234	0.0040	0.0106	0.0018	0.0003	0.0000	157.73	3.26
	Blank spike	RF11Pu	0.996	0.01	-0.0001	-0.0017	-0.0005	0.0025	-0.0002	0.0011	-0.0002	0.0011	101.32	3.13
98827A	whole water	RF13Pu	1.35	0.01	0.0777	0.0096	0.0772	0.0097	0.0351	0.0044	0.0035	0.0004	54.24	1.58
	<0.45 µm	RF14Pu	1.35	0.01	0.0178	0.0051	0.0174	0.0054	0.0079	0.0025	0.0004	0.0001	75.64	2.44
	>20 µm	RF15Pu	0.996	0.01	0.1148	0.0113	0.1144	0.0114	0.0520	0.0052	0.0043	0.0004	98.37	3.09
	>5 µm	RF16Pu	0.996	0.01	1.3772	0.0386	1.3768	0.0387	0.6258	0.0176	0.0035	0.0001	109.35	2.51
	0.45 – 20 µm	RF17Pu	0.996	0.01	0.0557	0.0092	0.0552	0.0094	0.0251	0.0043	0.0042	0.0007	65.26	2.39
	0.45 - 5 µm	RF18Pu	0.996	0.01	0.1154	0.0101	0.1150	0.0103	0.0523	0.0047	0.0003	0.0000	91.79	2.71
	<0.1 µm	RF19Pu	1.36	0.01	0.0108	0.0057	0.0103	0.0059	0.0047	0.0027	0.0002	0.0001	95.91	3.16
	0.1 - 0.45 µm	RF20Pu	1.35	0.01	0.0144	0.0031	0.0139	0.0036	0.0063	0.0016	0.0003	0.0001	93.89	2.07
	<100kDa	RF21Pu	1.35	0.01	0.0298	0.0068	0.0293	0.0071	0.0133	0.0032	0.0003	0.0001	77.86	2.14
	100kDa - 0.45 µm	RF22Pu	1.35	0.01	0.0072	0.0088	0.0067	0.0090	0.0030	0.0041	0.0001	0.0001	44.36	1.66
	<0.45 µm	RF23Pu	1.34	0.01	0.0087	0.0039	0.0082	0.0043	0.0037	0.0019	0.0007	0.0004	50.77	1.60
98827B	whole water	RF24Pu	1.35	0.01	0.0340	0.0112	0.0335	0.0113	0.0152	0.0051	0.0015	0.0005	30.83	1.24
	>20 mµm	RF26Pu	0.996	0.01	0.0578	0.0071	0.0574	0.0073	0.0261	0.0033	0.0022	0.0003	92.02	2.65
	<0.1 µm	RF27Pu	1.35	0.01	0.0095	0.0031	0.0091	0.0036	0.0041	0.0016	0.0002	0.0001	74.69	1.32
	0.1 - 0.45 µm	RF28Pu	1.34	0.01	0.0075	0.0035	0.0070	0.0039	0.0032	0.0018	0.0002	0.0001	77.28	1.99
	<100kDa	RF29Pu	1.36	0.01	0.0309	0.0110	0.0304	0.0111	0.0138	0.0051	0.0003	0.0001	28.17	1.33
	100kDa - 0.45 µm	RF30Pu	1.34	0.01	0.0151	0.0043	0.0147	0.0046	0.0067	0.0021	0.0002	0.0001	74.03	1.94
	Blank spike	RF31Pu	1.34	0.01	0.0005	0.0019	0.0000	0.0026	0.0000	0.0012	0.0000	0.0012	88.43	1.46

Table I-C cont.

			volume		Detector	Detector Background			
Sample	Size Fraction	ID	(L)	Detector	Efficiency	^{243}Am cpm	+/-	^{241}Am cpm	+/-
98826	whole water	RF1Am	4	1	0.20	0.0002	0.0002	0.0005	0.0003
	<0.45 µm	RF2Am	20	1	0.20	0.0003	0.0002	0.0005	0.0003
	>20 µm	RF3Am	10	1	0.20	0.0002	0.0002	0.0005	0.0003
	>5 µm	RF4Am	122	11	0.22	0.0008	0.0004	0.0008	0.0004
	0.45 – 20 µm	RF5Am	1	2	0.20	0.0003	0.0002	0.0008	0.0004
	0.45 - 5 µm	RF6Am	122	3	0.20	0.0019	0.0006	0.0003	0.0002
	<0.1 µm	RF7Am	20	2	0.20	0.0003	0.0002	0.0008	0.0004
	0.1 - 0.45 µm	RF8Am	20	2	0.20	0.0002	0.0002	0.0008	0.0004
	<100kDa	RF9Am	40	3	0.20	0.0013	0.0005	0.0003	0.0002
	100kDa - 0.45 µm	RF10Am	40	3	0.20	0.0008	0.0004	0.0003	0.0002
	Blank spike	RF11Am	1	7	0.20	0.0003	0.0002	0.0003	0.0002
98827A	whole water	RF13Am	10	4	0.19	0.0000	0.0000	0.0003	0.0002
	<0.45 µm	RF14Am	20	5	0.25	0.0002	0.0002	0.0002	0.0002
	>20 µm	RF15Am	12	9	0.23	0.0015	0.0005	0.0007	0.0003
	>5 µm	RF16Am	180	12	0.24	0.0006	0.0003	0.0007	0.0003
	0.45 – 20 µm	RF17Am	6	10	0.24	0.0015	0.0005	0.0002	0.0002
	0.45 - 5 µm	RF18Am	180	4	0.19	0.0000	0.0000	0.0002	0.0002
	<0.1 µm	RF19Am	20	4	0.19	0.0000	0.0000	0.0002	0.0002
	0.1 - 0.45 µm	RF20Am	20	5	0.25	0.0002	0.0002	0.0002	0.0002
	<100kDa	RF21Am	40	10	0.24	0.0015	0.0005	0.0002	0.0002
	100kDa - 0.45 µm	RF22Am	40	9	0.23	0.0015	0.0005	0.0007	0.0003
	<0.45 µm	RF23Am	5	8	0.23	0.0000	0.0000	0.0002	0.0002
98827B	whole water	RF24Am	10	1	0.20	0.0002	0.0002	0.0005	0.0003
	>20 µm	RF26Am	12	12	0.24	0.0007	0.0003	0.0007	0.0003
	<0.1 µm	RF27Am	20	6	0.19	0.0008	0.0004	0.0013	0.0005
	0.1 - 0.45 µm	RF28Am	20	2	0.20	0.0003	0.0002	0.0008	0.0004
	<100kDa	RF29Am	40	3	0.20	0.0012	0.0004	0.0002	0.0002
	100kDa - 0.45 µm	RF30Am	40	6	0.19	0.0007	0.0003	0.0013	0.0005
	Blank spike	RF31Am	1	7	0.20	0.0005	0.0003	0.0003	0.0002

Table I-C cont.

			count time			Background corrected				Background corrected		^{243}Am		^{241}Am	
Sample	Size Fraction	ID	(min)	^{243}Am counts	+/-	^{243}Am counts	+/-	^{241}Am counts	+/-	^{241}Am counts	+/-	cpm	+/-	cpm	+/-
98826	whole water	RF1Am	5866	1330	36.48	1329.01	36.48	60	7.75	57.03	7.75	0.227	0.006	0.010	0.001
	<0.45 µm	RF2Am	6984	1170	34.22	1167.65	34.22	42	6.48	38.47	6.48	0.167	0.005	0.006	0.001
	>20 µm	RF3Am	16821	696	26.38	693.17	26.38	86	9.27	77.50	9.27	0.041	0.002	0.005	0.001
	>5 µm	RF4Am	6924	1600	39.97	1594.16	39.97	1290	35.86	1284.16	35.86	0.230	0.006	0.185	0.005
	0.45 – 20 µm	RF5Am	16822	3620	60.14	3614.33	60.14	69	8.31	54.82	8.31	0.215	0.004	0.003	0.000
	0.45 - 5 µm	RF6Am	16822	3840	61.98	3808.81	61.98	191	13.82	185.33	13.82	0.226	0.004	0.011	0.001
	<0.1 µm	RF7Am	6985	1130	33.66	1127.65	33.66	18	4.24	12.11	4.24	0.161	0.005	0.002	0.001
	0.1 - 0.45 µm	RF8Am	5468	455	21.33	454.08	21.33	13	3.61	8.39	3.61	0.083	0.004	0.002	0.001
	<100kDa	RF9Am	6986	961	31	951.58	31	27	5.2	24.65	5.20	0.136	0.004	0.004	0.001
	100kDa - 0.45 µm	RF10Am	5866	11	3.32	6.06	3.32	2	1.41	0.02	1.41	0.001	0.001	0.000	0.000
	Blank spike	RF11Am	7216	7	2.65	4.57	2.65	11	3.32	8.57	3.32	0.001	0.000	0.001	0.000
98827A	whole water	RF13Am	5866	1650	40.56	1650.00	40.56	59	7.68	57.02	7.68	0.281	0.007	0.010	0.001
	<0.45 µm	RF14Am	6846	1560	39.5	1558.85	39.5	32	5.66	30.85	5.66	0.228	0.006	0.005	0.001
	>20 µm	RF15Am	7219	1730	41.61	1719.05	41.61	120	10.95	115.13	10.95	0.238	0.006	0.016	0.002
	>5 µm	RF16Am	6925	1610	40.06	1605.99	40.06	924	30.4	918.99	30.40	0.232	0.006	0.133	0.004
	0.45 – 20 µm	RF17Am	7219	1870	43.22	1859.05	43.22	43	6.56	41.78	6.56	0.258	0.006	0.006	0.001
	0.45 - 5 µm	RF18Am	16823	276	16.61	276.00	16.61	240	15.49	237.16	15.49	0.016	0.001	0.014	0.001
	<0.1 µm	RF19Am	6987	821	28.65	821.00	28.65	14	3.74	12.82	3.74	0.118	0.004	0.002	0.001
	0.1 - 0.45 µm	RF20Am	16793	143	11.96	140.17	11.96	17	4.12	14.17	4.12	0.008	0.001	0.001	0.000
	<100kDa	RF21Am	6912	1510	38.85	1499.52	38.85	28	5.29	26.84	5.29	0.217	0.006	0.004	0.001
	100kDa - 0.45 µm	RF22Am	6884	673	25.94	662.56	25.94	23	4.8	18.36	4.80	0.096	0.004	0.003	0.001
	<0.45 µm	RF23Am	6846	1260	35.5	1260.00	35.5	29	5.39	27.85	5.39	0.184	0.005	0.004	0.001
98827B	whole water	RF24Am	6842	763	27.62	761.85	27.62	19	4.36	15.54	4.36	0.111	0.004	0.002	0.001
	>20 µm	RF26Am	7219	1310	36.12	1304.78	36.12	58	7.62	52.78	7.62	0.181	0.005	0.007	0.001
	<0.1 µm	RF27Am	6846	1590	39.91	1584.23	39.91	30	5.48	20.77	5.48	0.231	0.006	0.003	0.001
	0.1 - 0.45 µm	RF28Am	6842	652	25.53	649.69	25.53	31	5.57	25.23	5.57	0.095	0.004	0.004	0.001
	<100kDa	RF29Am	6842	786	28.04	777.93	28.04	47	6.86	45.85	6.86	0.114	0.004	0.007	0.001
	100kDa - 0.45 µm	RF30Am	16794	1540	39.29	1528.68	39.29	57	7.55	34.36	7.55	0.091	0.002	0.002	0.000
	Blank spike	RF31Am	6846	1290	35.97	1286.54	35.97	17	4.12	14.69	4.12	0.188	0.005	0.002	0.001

Table I-C cont.

			^{243}Am		^{241}Am		blank corrected (dpm)				^{241}Am			
Sample	Size Fraction	ID	dpm added	+/-	dpm	+/-	^{241}Am	+/-	pCi	+/-	pCi/l	+/-	%recovery	+/-
98826	whole water	RF1Am	1.12	0.009	0.0481	0.0067	0.0353	0.0076	0.0160	0.0034	0.0040	0.0009	100.73	2.77
	<0.45 µm	RF2Am	1.12	0.009	0.0369	0.0063	0.0241	0.0073	0.0110	0.0033	0.0005	0.0002	74.33	2.18
	>20 µm	RF3Am	1.12	0.009	0.1252	0.0157	0.1124	0.0161	0.0511	0.0073	0.0051	0.0007	18.32	0.70
	>5 µm	RF4Am	1.13	0.009	0.9103	0.0342	0.8975	0.0344	0.4079	0.0156	0.0033	0.0001	94.08	2.36
	0.45 – 20 µm	RF5Am	1.12	0.009	0.0170	0.0026	0.0042	0.0044	0.0019	0.0020	0.0019	0.0020	93.68	1.56
	0.45 - 5 µm	RF6Am	1.12	0.009	0.0545	0.0042	0.0417	0.0055	0.0190	0.0025	0.0002	0.0000	101.92	1.66
	<0.1 µm	RF7Am	1.12	0.009	0.0120	0.0042	-0.0008	0.0056	-0.0003	0.0025	0.0000	0.0001	70.40	2.11
	0.1 - 0.45 µm	RF8Am	1.12	0.009	0.0207	0.0090	0.0079	0.0097	0.0036	0.0044	0.0002	0.0002	36.21	1.70
	<100kDa	RF9Am	1.13	0.009	0.0293	0.0062	0.0165	0.0072	0.0075	0.0033	0.0002	0.0001	60.78	1.98
	100kDa - 0.45 µm	RF10Am	1.12	0.009	0.0042	0.2607	-0.0086	0.2607	-0.0039	0.1185	-0.0001	0.0030	0.46	0.25
	Blank spike	RF11Am	1.12	0.009	2.1008	1.4656	2.0880	1.4657	0.9491	0.6662	0.9491	0.6662	0.28	0.16
98827A	whole water	RF13Am	1.12	0.009	0.0387	0.0053	0.0259	0.0064	0.0118	0.0029	0.0012	0.0003	132.38	3.26
	<0.45 µm	RF14Am	1.12	0.009	0.0222	0.0041	0.0094	0.0055	0.0043	0.0025	0.0002	0.0001	79.85	2.03
	>20 µm	RF15Am	1.14	0.009	0.0764	0.0075	0.0636	0.0083	0.0289	0.0038	0.0024	0.0003	89.65	2.18
	>5 µm	RF16Am	1.13	0.009	0.6466	0.0268	0.6338	0.0270	0.2881	0.0123	0.0016	0.0001	84.81	2.12
	0.45 – 20 µm	RF17Am	1.13	0.009	0.0254	0.0040	0.0126	0.0054	0.0057	0.0025	0.0010	0.0004	93.37	2.18
	0.45 - 5 µm	RF18Am	1.13	0.009	0.9710	0.0862	0.9582	0.0863	0.4356	0.0392	0.0024	0.0002	7.65	0.46
	<0.1 µm	RF19Am	1.12	0.009	0.0175	0.0051	0.0047	0.0063	0.0021	0.0029	0.0001	0.0001	55.30	1.93
	0.1 - 0.45 µm	RF20Am	1.12	0.009	0.1132	0.0343	0.1004	0.0345	0.0457	0.0157	0.0023	0.0008	2.93	0.25
	<100kDa	RF21Am	1.13	0.009	0.0202	0.0040	0.0074	0.0054	0.0034	0.0025	0.0001	0.0001	78.66	2.04
	100kDa - 0.45 µm	RF22Am	1.12	0.009	0.0310	0.0082	0.0182	0.0090	0.0083	0.0041	0.0002	0.0001	36.88	1.45
	<0.45 µm	RF23Am	1.11	0.009	0.0245	0.0048	0.0117	0.0060	0.0053	0.0027	0.0011	0.0005	71.28	2.01
98827B	whole water	RF24Am	1.12	0.009	0.0228	0.0065	0.0101	0.0074	0.0046	0.0034	0.0005	0.0003	49.50	1.80
	>20 µm	RF26Am	1.13	0.009	0.0457	0.0067	0.0329	0.0076	0.0150	0.0035	0.0012	0.0003	66.18	1.84
	<0.1µm	RF27Am	1.12	0.009	0.0147	0.0039	0.0019	0.0053	0.0009	0.0024	0.0000	0.0001	107.05	2.70
	0.1 - 0.45 µm	RF28Am	1.11	0.009	0.0431	0.0097	0.0303	0.0103	0.0138	0.0047	0.0007	0.0002	41.78	1.64
	<100kDa	RF29Am	1.13	0.009	0.0666	0.0102	0.0538	0.0109	0.0245	0.0049	0.0006	0.0001	50.73	1.83
	100kDa - 0.45 µm	RF30Am	1.11	0.009	0.0249	0.0055	0.0122	0.0066	0.0055	0.0030	0.0001	0.0001	42.49	1.09
	Blank spike	RF31Am	1.12	0.009	0.0128	0.0036	0.0000	0.0051	0.0000	0.0023	0.0000	0.0023	81.99	2.30

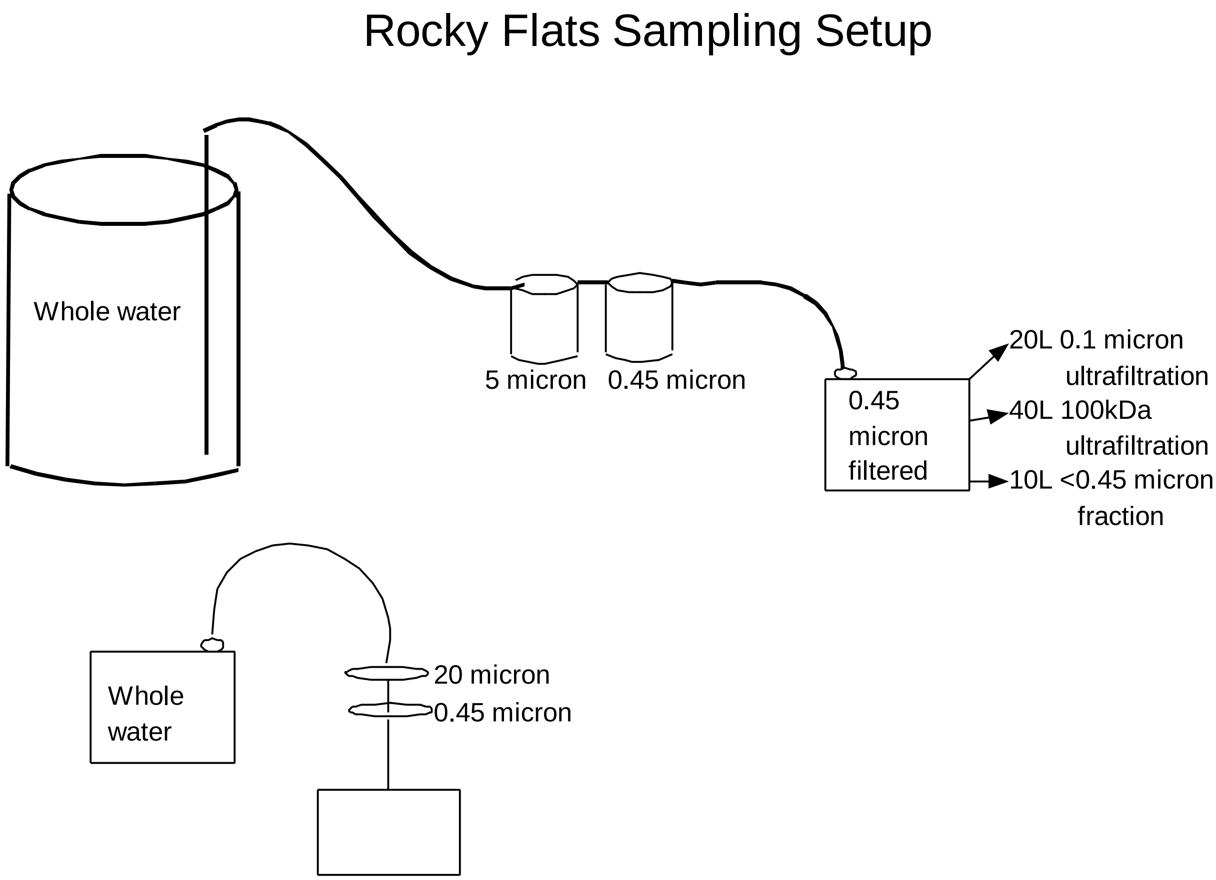

Figure I-A: Rocky Flats Filtration and Ultrafiltration Sampling Setup. Two systems were run in parallel – one for the particulate fractions (>0.45mm and >20mm), and one for CFUF (cross flow ultrafiltration) experiments.

Appendix II. Detailed Data for SPM, POC, PON, DOC, Fe, Al, Mn, and Anion Concentrations.

August 26-27, 1998 Sampling Trip

Table II-A: Suspended particulate matter (SPM) concentration (^{3}0.45µm):

SS#	Sample ID	Sample	SPM (mg/l)	Avg (mg/l)	±1σ	±1σ (%)
1	98826-1	8/26/98	98.6	87.7	15.4	17.5
2	98826-2*	8/26/98	76.9			
3	98827A-1*	8/27/98-A	23.1	27.1	5.6	20.6
4	98827A-2	8/27/98-A	31.1			
5	98827B-1	8/27/98-B	34.3	34.5	0.3	0.9
6	98827B-2	8/27/98-B	34.7			
	SPM data from GF/F filters					
1	98826-1	8/26/98	87.6	75.0	17.9	23.9
2	98826-2	8/26/98	62.3			
3	98827A-1	8/27/98-A	32.8	33.2	0.6	1.7
4	98827A-2	8/27/98-A	33.6			
5	98827B-1	8/27/98-B	35.2	33.0	3.1	9.3
6	98827B-2	8/27/98-B	30.9			

*: particles lost to the filter holder.

Table II-B: POC/PON (in mg-C/liter):

SS#	Sample ID	Sample	POC (mg/l)	POC (mg/l) Average	±1σ (mg/	±1σ (%)	PON (mg/l)	PON (mg/l) Avg	±1σ
1	98826-1	8/26/98	14.4	13.1	1.86	14.2	2.09	1.92	0.24
2	98826-2	8/26/98	11.8				1.75		
3	98827A-1	8/27/98-A	9.8	10.1	0.34	3.4	1.37	1.37	0.00
4	98827A-2	8/27/98-A	10.3				1.37		
5	98827B-1	8/27/98-B	10.1	9.8	0.54	5.5	1.37	1.31	0.08
6	98827B-2	8/27/98-B	9.4				1.26		

Table II-C: POC/PON (in mg-C /g particle):

SS#	Sample ID	Sample	POC (mg/g)	POC (mg/g) Average	±1σ (mg/g)	±1σ (%)	PON (mg/g)	PON (mg/g) Average	±1σ (%)
1	98826-1	8/26/98	164.7	177.0	17.4	9.8	23.92	26.01	2.97
2	98826-2	8/26/98	189.2				28.11		
3	98827A-1	8/27/98-A	300.2	303.8	5.1	1.7	41.66	41.16	0.70
4	98827A-2	8/27/98-A	307.4				40.66		
5	98827B-1	8/27/98-B	288.1	296.1	11.3	3.8	38.82	39.76	1.34
6	98827B-2	8/27/98-B	304.1				40.71		

Table II-D: Concentration of dissolved organic carbon (DOC) in ²0.45 µm fraction

SS#	Sample ID	Bottle #	DOC (ppm)	±1σ (ppm)	DOC (ppm) Avg	±1σ (ppm) Avg	
1	98826-1	B275	12.43	0.3	12.3	0.6	
2	98826-2	B062	12.71	0.2			
3	98826-3	C068	11.62	0.2			
4	98827-1	B153	14.69	0.3	14.0	1.0	
5	98827-2		A185	13.22		1.0	
98827-1	Trace Metal		13	18		18	
98827-2	Trace Metal		15	26		26	
98827-1	Nutrient		18	29		26	3.
98827-2	Nutrient		12	26		25	1
98827-3	Nutrient		10	18		17	4.
98827-4	Nutrient		8	16		16	

Table II-F: Concentrations of Anions in ²0.45 µm Fraction:

Sample I.D.	F (mg/L)	Cl (mg/L)	NO_3 (mg/L)	HPO_4 (mg/L)	SO_4 (mg/L)	Si (mg/L)	
98827-1	0.33	93.5	<0.1	<0.2	30.7	5.3	T
98827-2	0.34	91.8	<0.1	<0.2	30.3	5.2	(µ
98827-3	0.33	93.0	<0.1	<0.2	30.2	4.9	(µ
98827-4	0.34	93.6	<0.1	<0.2	30.9	4.9	1.
98826-1	0.42	87.8	4.3	0.8	32.7	5.5	M
98826-2	0.41	92.6	4.3	0.8	34.4	5.6	8
98826-3	0.41	94.0	4.3	0.8	34.7	5.5	M

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Appendix III. Detailed Results from Ultrafiltration Test Experiments

Prior to field sampling, several laboratory tests were completed in the laboratory to determine efficiency of chemical (Tests 1-3) and ultrafiltration (EXP 1-3) procedures (A&B). In all cases deionized or tap water was used and samples were done in duplicate. Approximately 2.7 pCi of tracers ^{240}Pu and ^{242}Pu were added. A brief description of each experiment is as follows:

In Test 1 Pu determination was made using precipitation of $CaPO_4$. ^{240}Pu was added to each sample and the yield tracer ^{242}Pu was added at different stages (before precipitation, before columns, before microprecipitation) of the chemical procedure to determine efficiencies of each step. The percent recoveries were calculated based on the activity of ^{240}Pu measured /activity of ^{240}Pu added. Test 2 was a repeat of test 1 with ^{241}Am also measured. Test 3 was a repeat of test 2 but FeOH was precipitated instead of $CaPO_4$.

For EXP 1 tap water was collected in a large volume container (~100L). The water was filtered to a 0.45μm filter and collected in 20 liter carboys. To each carboy, ^{240}Pu, ^{242}Pu, ^{241}Am and ^{243}Am were added. A 20 liter carboy was used for each ultrafiltration setup 0.1μm, 1kDa, 100kDa. After completion of the ultrafiltration, two washes of the ultrafiltration setups collected processed and counted. The 0.1μm filter was washed with dilute HCl followed by EDTA and the 1kDa and 100kDA filters were washed with dilute HNO3 followed by oxalic acid. EXP 2 was a repeat of EXP 1. EXP 3 was the same as 1 and 2 but Pu (VI) was used instead of Pu (IV).

Table III-A: Percent recovery of 2.7 pCi added during precipitation experiments

	Am-241		Pu-240	
Test 1 ($CaPO_4$)			87.72	+/-2.20
			79.57	+/-1.95
Test 1 ($CaPO_4$)			77.23	+/-1.76
(heated)			66.61	+/-1.22
Test 2 ($CaPO_4$)	82.65	+/-4.47	84.77	+/-2.86
	89.31	+/-3.96	89.50	+/-2.95
Test 2 ($CaPO_4$)	92.65	+/-3.29	82.79	+/-3.15
(heated)	93.39	+/-3.37	75.74	+/-2.88
Test 3 (FeOH)	88.56	+/-3.79	97.79	+/-4.25
	99.21	+/-4.16	96.77	+/-3.16

Table III-B: Mass Balance of ultrafiltration experiments. Values represent % recovery of the 2.7 pCi of analyte (Am-241 or Pu-240) added.

				Am-241							Pu-240			
		0.1µ		1kDa		100kDa			0.1µ		1kDa		100kDa	
		%	±	%	±	%	±		%	±	%	±	%	±
EXP1	Permeate	24.84	1.27	13.68	.67	27.7	1.85		56.96	2.78	35.76	2.25	57.12	2.89
	Retentate	30.66	1.44	35.22	1.77	11.8	0.71		12.81	1.11	30.15	2.13	11.37	1.91
	Wash(%)	-		-		-			-		-		-	
	Sum(%)	55.5	1.92	48.9	1.89	39.5	1.98		69.77	2.99	65.91	3.10	68.49	3.46
EXP2	Permeate (%)	76.33	5.28	31.12	1.45	70.15	4.78		81.85	4.19	28.85	1.87	68.28	3.10
	Retentate(%)	18.08	.98	44.69	1.33	10.44	0.44		10.06	0.68	38.56	2.17	8.27	0.80
	Wash(%)	13.64	3.76	3.9	.36	8.08	1.31		6.83	0.82	1.83	1.54	0.63	0.44
	Sum(%)	108.05	5.37	79.71	1.97	88.67	4.80		98.74	4.32	69.24	3.25	77.18	3.23
EXP3	Permeate (%)								83.58	3.65	77.97	3.04	75.88	3.68
	Retentate(%)								6.54	0.66	10.08	0.80	7.35	0.62
	Wash(%)								-		-		-	
	Sum(%)								90.12	3.71	88.05	3.04	83.23	3.73

Appendix IV. QA/QC

A total of 56 samples were run. The duplicate error ratio (DER) for the 8 samples run in duplicate ranged from 1.5 ± 1.0 to 3.3 ± 1.6. The alpha spectrometers were energy and efficiency calibrated prior to counting of samples. During alpha counting, energy calibration checks were performed once a week. No recalibrations were performed as the peaks remained within 40keV of the expected energy.

The breakdown of tracer recoveries is as follows: two >100%, twenty eight between 75-100%, eighteen <75%, 2<30% and 6<10%. All samples greater than 100% and less than 10% recovery were considered unusable and labeled HR (high recovery) or LR (low recovery). During sample processing one sample was combined with another by mistake, accounting for its high recovery. An explanation for the low recoveries is the difficult matrix, primarily algae, which caused problems with digestion and columns. Matrix spikes were not run as all water collected was processed to a variable degree, and time for processing did not allow it. Laboratory control samples were the basis for the laboratory test samples prior to sampling. Tracer recoveries were 100%. Two blanks were run and all samples were blank and background corrected. The blank values were 0.0058 ± 0.0016 pCi for ^{241}Am and 0.0013 ± 0.0009pCi for ^{239,240} Pu.