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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 FY00:

1) Actinide Phase Speciation: Total ^{239,240}Pu and ^{241}Am concentrations in the storm runoff and pond discharge samples were below the discharge limit of 0.15 pCi/L. Both Pu and Am activity concentrations in storm runoff (GS10 on 5/8/00) and pond discharge (GS03 on 4/27/00) samples were also comparable to those determined in 1999 samples from Walnut Creek at GS03. Both storm runoff and pond discharge sampled this year showed again a substantial fraction of Pu and Am in the 0.5 µm filter-passing fraction. The majority of Pu and Am in the 0.5µm filter-passing fraction was colloidal, i.e., it was filtered out by 100kDa or 3kDa ultrafilters using cross-flow ultrafiltration (CFUF), with only a small fraction of the 0.5µm filter-passing Pu and Am passing a 3 kDa ultrafilter. Pu activity concentrations in the particulate fractions (>20 µm & 0.5-20 µm) amounted to about 90% of the total.
2) Colloidal Pu remobilization during soil erosion: Pu concentrations above the action level (≥ 0.15 pCi/L) occur predominantly during storm runoffs in early spring and summer. During these events, elevated concentrations of colloidal Pu, accompanying those in the particle phase, are being observed (Santschi et al., 1999). We hypothesized that colloidal Pu, as a pseudo-colloid (i.e., Pu associated with a different carrier phase), is generated by soil erosion and transport, and that remobilization of colloid-bound Pu during soil erosion events is likely aided by elevated concentrations of humic acids in soil waters. During this funding year, we tested this hypothesis in soil resuspension experiments with RFETS soils, and demonstrated that there is a close relationship between the remobilization of colloidal Pu (≤0.4 µm) and the dissolved organic matter (DOM) concentration (i.e., humic acids) in the water at environmentally relevant concentrations. We furthermore observed a kinetic effect, i.e., an increasing colloidal Pu concentration at increasing resuspension time, and a particle-concentration effect, i.e., a decreasing phase partition coefficient of Pu (Rp {in L/kg} = ratio of particulate concentration {in pCi/kg} to the filter-passing concentration {in pCi/L}) with increasing particle concentration (Cp, in kg/L), as $\log(Rp) = 3.2 - 0.74 \log(Cp)$. Which of these factors is dominant in the field would require further analysis of field data.
3) Colloid Composition: Isoelectric focusing of ^{14}C radiolabelled colloids showed again that they contain strong acid functional groups which impart the organic fraction a negative charge at natural pH's. Bulk colloidal matter contains also strong basic functional groups, as was evident from ^{59}Fe labeling experiments. Colloids isolated from the pond discharge and storm runoff samples contained about 3 to 12 % organic carbon, 0.02 to 1 % Fe, and 0.02 to 1.3 % Al, with the higher value for the GS10 sample from 5/8/00. These values agree with their predominantly inorganic nature. The clay- and organic carbon - rich nature of the colloids was corroborated by additional Scanning Transmission Electron Microscopy (TEM) and Energy Dispersive Spectroscopy (EDAX) images, which also suggest that GS10 colloids contain larger amounts of Fe-rich colloids.

4) Particle and Th(IV) residence times in Pond B-5: Measurements of Th(IV), as an analogue of Pu(IV), in total, dissolved and particulate fractions of Pond B-5 water resulted in estimates of how long, on the average, it would take for a four-valent ion to be scavenged and removed from the water. Th(IV)-scavenging and particle residence times, derived from measurements of $^{238}U/^{234}Th$ disequilibria in the water, were of the order of a few hours to one day, respectively, supporting the effectiveness of the ponds in reducing particle and Pu loads. From these results, we would predict that at water residence times in excess of one to a few days, most of the Pu and particle load entering a pond would be removed to the sediments. Water residence times are currently more than a month. If holding ponds were to remain, they should be designed with water residence times of more than a few days to maximize their effectiveness in reducing particle and actinide loads to down stream sites.

1. Overview, Questions and Hypothesis

The goal of the Actinide Migration Evaluation (AME) is to provide data in support of Site closure, including: 1) soil action levels vis-à-vis surface water quality; 2) long-term disposition of the ponds; 3) 'far-field' actinide behavior during long-term closure.

Four broad areas of investigation emerged from group discussions with AME members and site personnel for consideration for FY 2000 work:

1) Phase speciation studies of actinides in surface waters;
2) Colloid characterization studies;
3) Studies of colloid generation during simulated soil erosion (i.e., soil resuspension) events;
4) Determination of in-situ removal time studies for particles and colloids in ponds.

The following question was addressed in the FY2000 research: What causes exceedances of Pu and Am concentrations in RFETS surface waters, which often are associated with storm runoff?

The following Hypothesis was tested in the FY2000 research: The elevated concentrations are related, besides hydrodynamic conditions (i.e., storm surface runoff vs base flow), to the nature and types of organic matter present in soils, stormwater and pond water at the time of sampling. Organic matter can modify the surface charge and characteristics of particle and colloid aggregates. On the one hand, humic and fulvic acids can impart a negative surface charge to particles and colloids, which can promote disaggregation and dispersion of aggregates, and thus, increased mobility and concentrations of colloidal species in surface waters. On the other hand, large, surface-active, organic molecules such as exopolymeric acid polysaccharides from bacteria and algae, act to bind colloidal and particulate species together, and thus, cause their removal and lower their concentrations in surface waters.

2. Justification

A wide range of work at Rocky Flats has demonstrated that 'particulate' and 'colloidal' forms of Pu and Am make up a significant fraction of actinides in surface waters and suggests that surface water transport of Pu and Am is dominated by aggregation and disaggregation processes of particles and colloids during soil erosion events. A clear understanding of the phase speciation of Pu is a prerequisite for the development of defensible closure strategies. Questions concerning the environmental form of Pu and Am have direct bearing on evaluating the importance of various migration pathways and in the development and refinement of Data Quality Objectives (DQO: i.e., model parameters and uncertainty levels).

3. Tasks

3.1 Task 1: Colloid and particulate associations of Pu and Am

In FY2000 we continued the phase speciation work of Pu and Am in stream water, using the sites already investigated in 1998 and 1999.

3.1.1 Objectives:

1. To determine the association of Pu and Am with: 1) particulate, 2) colloidal and 3) dissolved phases for field samples from a selected pond, pond release waters and a future compliance point.
2. To determine if natural organic matter plays a role in the formation of colloids that facilitate transport of actinides from contaminated soil source terms.
3. To determine the chemical nature of the colloidal carrier phase (e.g., Fe, Mn, C, Al, etc.).

3.1.2 Justification:

FY 98&99 work has demonstrated that 0.5 µm filter-passing Pu is predominantly in a colloidal form. The issue of the speciation of filter-passing forms of Pu is crucial for understanding of transport, i.e., the relationship between surface water quality and soil action levels, as well as to evaluate the scientific defensibility of RESRAD model simulations. The distribution of Pu and Am among different particles sizes and colloid molecular weights is important for developing management controls on surface water quality.

3.1.3 Analytical Plan:

Phase association studies of Pu and Am investigations involved the following subtasks:

a) Surface water sampling by CFUF,
b) Colloid isolation and characterization,

The selected sampling sites were Pond B-5, pond release waters at compliance point GS03: Walnut Creek at Indiana Street, and stormwater runoff at GS10, South Walnut Creek at the B-series bypass (Figure 1). Grab samples were collected by bailing water from the stream using a small container followed by compositing the water into clean 15 to 20 Liter Nalgene carboys for processing and analysis in the laboratory (Figure 2). In addition, soil samples were collected for extraction of natural organic matter fractions and used in the soil erosion/colloid stability experiments at Texas A&M University at Galveston (TAMUG).

The methods for isotope separation were adapted from EPA Method 908.0 (1980), USDOE (1979), USEPA (1979), and Yamato (1982) as described also in (Appendix 2). 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 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. The supernate was decanted and the precipitate was shipped to TAMUG. At TAMUG, the samples were dissolved in concentrated HCl to which 75 ml 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 (Appendix 2). The first column 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.

The protocols of Guo and Santschi (1996,1997) and Wen *et al*. (1996, 1999) were followed for isolating colloidal and particulate phases of metals such as Pu, Am from surface waters by CFUF. Chemical parameters measured in the aqueous sample fractions were:

- total organic carbon (TOC),
- dissolved organic carbon (DOC),
- colloidal organic carbon (COC),
- particulate organic carbon (POC),
- pH,
- alkalinity, and
- Al, Fe and Mn.

Chemical parameters measured in the filtered solids (colloid and particulate phases) were:

- % organic carbon,
- Al, Fe and Mn

Aliquots of colloid and particle samples were kept for TEM imaging analyses. These analyses were used to visually identify the gross composition of the particulate and colloidal phases. In selected cases, electrophoretic focusing experiments (Quigley, 2000) determined isoelectric points of isolated colloids. *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.

3.1.4 Strategy:

Surface water samples from the field were taken in an extended sampling expedition in April/May 2000. Laboratory processing of samples was carried out in the following months.

3.2 Task 2: Resuspension/Colloidal Pu release experiments

The laboratory experiments involved treatment of actinide-contaminated soil with commercial and extracted, natural organic matter to investigate natural mechanisms of colloid formation during storm water runoff.

3.2.1 Objectives:

1. To determine if natural organic matter plays a role in the formation of colloids that facilitate transport of actinides from contaminated soil source terms.
2. To experimentally determine what natural processes favor particle and colloid stabilization or destabilization using experimental approaches given in Wilkinson et al. (1997).

3.2.2 Justification:

A better understanding of the process(es) which control the speciation of filter-passing forms of Pu is crucial for evaluating their transport. A major focus of this task is to determine the dominant Pu species generated via resuspension during storm runoff. Some of the major processes which control colloid formation will be studied in controlled laboratory experiments, with the purpose of relating colloid stability to organic matter composition. The type of natural organic matter can control the tendency of particles and colloids to aggregate. For example, small organic molecules such as fulvic acid and other organic acids can increase dispersion of soil colloids through their effect on particle charge, while large, surface-active, organic molecules such as polysaccharides act to bind colloid particles together (e.g., Gu and Doner, 1993; Kretzschmar et al., 1997; Wilkinson et al., 1997). Colloid stability is an important colloid characteristic because it reflects the extent to which colloidal material interacts with particles and immobile soil media. Therefore, colloid stability is a primary parameter for 1) estimating colloid mobility and for 2) development of engineering and management strategies for removing colloidal-associated actinides from surface water.

3.2.3 Analytical Plan:

Colloid formation and associated actinide partitioning was investigated in controlled soil resuspension experiments. The soil was first sieved through a 1mm sieve to reduce inhomogeniety of the Pu activities. This fine soil fraction was added to a teflon jar containing 0.5μm filtered tap water which was rapidly stirred using a stir bar on a stir plate. The resuspension experiments were carried out with variable amounts of commercial organic matter compounds (i.e., Aldrich humic acids, exopolymers of microorganisms, such as Alginic Acids, Xanthen) and soil extracts (e.g., humic acids). Pu -contaminated soils from the Site were resuspended in the presence or absence of these natural organic substances and then 0.45μm filtered. In select cases the filtered fraction was fractionated further using 1kDa stirred cell ultrafiltration to determine if Pu colloid formation occurs(Table 17). The solutions were analyzed for DOC, Pu and Am to determine whether colloid formation also liberated Pu and Am from the soils. A schematic of these experiments is displayed in Figure 3. Pu-contaminated soil in 0.5μm filtered tap water served as a control.

3.2.4 Strategy:

The strategy was to carry out these soil resuspension and colloid remobilization experiments using existing samples. Laboratory processing of these samples was carried out throughout the year.

3.3 Task 3: In-situ particle and colloid residence time determination

We determined in-situ particle and colloid residence times in pond waters using the $^{234}Th/^{238}U$ disequilibrium technique (e.g., Baskaran et al., 1996; Guo et al., 1997; Santschi et al., 1999a).

3.3.1 Objective

To investigate if in-situ particle and Th(IV) residence time determination in pond waters using the $^{234}Th/^{238}U$ disequilibrium technique prove to be feasible.

3.3.2 Justification:

FY 98&99 work has demonstrated that 0.5 μm filter-passing Pu is predominantly in a colloidal form. Colloid stability is a primary parameter for 1) estimating colloid mobility and for 2) development of engineering and management strategies for removing colloidal-associated actinides from surface water. Particle and colloids residence times in ponds are likely short, as actinides are removed to better than 90% in the pond water in about 45 days, according to Site sources. Better knowledge of residence times of particles and colloids would facilitate a future decision about pond operations.

3.3.3 Analytical Plan:

Because ^{238}U concentrations were high enough (i.e., 1 dpm/l, or higher), and relatively constant over time, the $^{234}Th/^{238}U$ disequilibrium technique (e.g., Baskaran et al., 1996; Guo et al., 1997; Santschi et al., 1999a), it can provide estimates of particle and colloid residence times at steady state based on the deficiency of ^{234}Th with respect to its mother nuclide, ^{238}U, in particle, colloid and solution phases. Large volumes (600 to 1100L) of water were pumped from the ponds through a prefilter (changed three times during filtration) and two MnO_2 impregnated fiber filters, which efficiently extract ^{234}Th. The efficiency of extraction is determined from the activity ratio in the two cartridges, and is usually over 90% (Baskaran et al., 1993).

3.3.4 Strategy:

Surface water samples from Pond B-5 were taken in a sampling expedition in April 26-27 by pumping water through extractor cartridges in the field. Laboratory processing of samples was carried out in the month following sampling.

4. RESULTS AND DISCUSSION

We sampled, filtered, ultra-filtered and processed a discharge sample on 4/27/00 and a storm runoff sample on 5/8/00, in duplicate. For the discharge sample, approximately 200 liters of water were taken from GS03 and brought in various containers to CSM, where it was combined in a 55 gallon drum, which had been pre-cleaned with Radiacwash (a detergent). The 200 liters of storm runoff were collected from GS10 and brought to Colorado School of Mines (CSM) in two 55 gallon drums, which had been pre-cleaned with Radiacwash. Both samples were size fractionated in duplicate for a total of 4 samples of approximately 90 liters each. 12 - 18 liters of total water (unfiltered) was set aside for each sample. The remaining water was passed through 20 micron and 0.5 micron filters and collected in 5 gallon carboys. The 5 gallon carboys used to take sub-samples from the drums had been washed with Radiacwash, as well as with acid. One carboy of 0.5 micron filtered water was set aside for later analysis of the 0.5 micron filter passing fraction. The remaining 0.5 micron filtered water was used for the cross flow ultrafiltration (CFUF). Thirty to forty liters were run through a 3 kDa filter membrane and another thirty to forty liters were run through a 100 kDa filter membrane. All size fractions were processed by $Fe(OH)_3$ precipitation at CSM. The resultant precipitate was sent to TAMUG for radiochemical analysis of ^{239,240} Pu and ^{243}Am.

In a another experiment, Pond B-5 was sampled 4/26/00 and 4/27/00 for ^{234}Th and ^{238}U to calculate residence times of the particulate and filter-passing phases. Th-234 was extracted by pumping hundreds of liters of water through a 0.5 µm prefilter and two 0.5 µm MnO_2 impregnated cartridge filters. Filters were then combusted in a muffle furnace and the ash was packed into counting vials and counted on a well Ge gamma detector ^{238}U was determined by ICP-MS.

At TAMUG, soil resuspension experiments were also conducted using Rocky Flats soil (99A3372-002.006) as a source of Pu to the water. Soil concentrations, resuspension times, as well as type and quantity of organic substances were varied to assess colloid formation effects on <0.45 µm filter-passing ^{239,240} Pu activities.

4.1. Pu and Am concentration and phase speciation data from pond and storm discharge samples:

^{239,240}Pu and ^{241}Am phase speciation results are shown in Tables 1a, 1b and 2, and ancillary results in Tables 3-11. Unfortunately, in the first pond discharge sample (A), Pu and Am activities in the 3kDa ultrafiltration samples were anomalous. The problem was attributed to the tracer not having equilibrated with the sample, likely due to interference by organics. To alleviate the organic matter problem, all subsequent samples were treated with strong acids (HCl and HNO_3) first, and evaporated after tracer addition prior to the column chemistry. The second pond discharge sample (B), gave more reliable results, as judged from the Pu activity balance.

The data used for Figues 4-6 are summarized in Table 2. Sample 'B' was used for the particulate values with the total being the sum of the measured fractions for Pu and the >20μm being calculated by the difference between the measured total and the other fractions. For the <0.5μm fractions, average values were used where feasible. For the 100kDa – 0.5μm fraction, the measured 100kDa container and cartridge acid wash was added to the measured 100kDa – 0.5μm fraction. The 3-100kDa was calculated by difference (sum of <0.5μm fractions - <3kDa – 100kDa-0.5μm).

As had been observed in previous years, most of the Pu and Am in the water (Tables 1a & 1b, Figures 4-6) was found in the particulate phase, with most of the remainder in the colloidal phase. Pu/Am ratios (Table 12) of the different phases and samples indicate that the storm runoff samples generally had lower ratios than the pond discharge samples. Values of phase partition coefficients, Rp, range from 4.8 to 8.0 10^4 L/kg (Table 13).

4.2. Colloid characterization:

ICP-MS, CHN, isoelectric focusing and TEM were used for characterizing freeze-dried colloids from the April and May sampling expeditions. As in previous years, colloids were primarily composed of clay and organic matter colloids (0.02-1 % Fe, 0.02-1.3 % Al, 3.1-11.9 **%** OC, see Tables 8, 9). Colloids from the storm discharge contained higher Al, Fe, Mn (Table 9), and %OC (Table 8). The same was true for % OC in suspended particulate matter (Table 4).

Colloids were radioactively tagged with ^{14}C and ^{59}Fe (^{14}C –Dimethyl sulfate on the -OH sites of sugars ^{59}Fe on –OH sites of Fe oxyhydroxides and clay minerals), using protocols given Wolfinbarger and Crosby (1983). The labeling is a methylation reaction and labels mainly the hydroxyl groups of both neutral and amino sugars (Wolfinbarger and Crosby, 1983). The ^{14}C radiolabeled organic matter was stored in dH_2O in a sterilized bottle at 4°C. Generally, both isotopes were added to a small aliquot of colloids in a batch reactor. Each isotope were measured in the different filter fractions separately using liquid scintillation counting, LSC. Small volumes (100µl with approximately 20 nCi ^{14}C) of ^{14}C or ^{59}Fe radiolabeled colloids were then used in isoelectric focusing gel electrophoresis experiments (Figure 7, Table 14). The charged molecules migrate through the gel toward one of the electrophoresis electrodes until protonation or deprotonation within the pH gradient results in a net neutral charge for the molecule. A Multiphor II system, purchased from Amersham Pharmacia Biotech, was used. Typically, sample detection within the gel was made using liquid scintillation counting. The gel was sectioned into 1cm sections and each section was put in a glass liquid scintillation vial with 3 ml 0.1% sodium dodecylsulfate (SDS). The SDS solution worked as a detergent to wash the molecules out of the gel matrix which greatly increased counting recovery of the radiolabels. The gel sections were allowed to soak in the SDS solution for 24 hrs. before liquid scintillation fluid was added and the vials counted.

Isoelectric focusing results are given in Table 14, and Figure 7. These results reveal that ^{14}C-labeled colloidal organic matter rich in carbohydrate moieties exhibits strong functional groups with pKa of 3.5 or less, imparting these colloids a negative charge, and therefore, allowing the majority of organic matter to migrate to the anode (pH≤3.5). Furthermore, ^{59}Fe-labeled iron hydroxides and clay minerals were apparently still positively charged, and thus, were able to migrate to the cathode at pH≥9.5. In summary, the gel electrophoresis results support our contention that the majority of the colloids is composed of organic matter (^{14}C-labelled) with strong acid, and inorganic clay and iron oxide minerals (^{59}Fe-labelled) with basic functional groups. Results from 2000 are in agreement with those from 1999, and any differences between the two sets of measurements are small.

Transmission Electron Microscopy (TEM) combined with Energy Dispersive X-Ray Microprobe Analysis (EDAX) were applied to obtain information on composition and forms of colloid samples. Aliquots of freeze-dried colloids (RF137 and RF157) were analyzed by Marcia West and Gary Leppard at McMaster University. Major findings: The main colloids in both samples were clays (some of which contained Fe), microbes (mainly bacteria), microbe parts, debris, biogenic silicates, membranous structures (probably biological), titanium microcrystals and extremely small colloids aggregated into complex structures. These latter aggregates of extremely small colloids could not be properly analyzed morphologically because the samples had been processed as dehydrated powders. Thus the fractal arrangements and long range associations would almost certainly have been artifactually altered (see earlier work of Buffle and Leppard).

The freeze-drying method that was employed can clump the colloids severely, making fine distinctions between the two samples more difficult to ascertain. Likely, sample RF157 was more mineralized than sample RF137, which is corroborated by the metal analysis of the colloid species (Table 9). Sample RF157 was richer in Fe-containing clays than was RF137 (Appendix 4), in agreement with the analytical data (Table 9).

4.3. ^{238}U/^{234}Th disequilibrium to derive thorium scavenging and particle residence times in Pond B-5:

Measurements of Th(IV), as an analogue of Pu(IV), in total, dissolved and particulate fractions of Pond B-5 water resulted in estimates of how long, on the average, it would take for a four-valent ion to be scavenged and removed from the water. Results of these measurements indicate that the ^{238}U concentrations in Pond B-5 were 2.28±0.04 µg/L (0.76±0.01 pCi/L) in unfiltered water, 2.11±0.11 µg/L (0.71±0.04 pCi/L in 0.5 µm filtered water; the ^{234}Th concentration in the 0.5 µm filter-passing fraction was equal to or less than 0.005 pCi/L, three times our detection limit, while the particulate (≥0.5 µm) $^{234}Th_p$ was about 0.04 pCi/L or less (Table 15).

From these activity determinations, the scavenging and particle residence times (τ_i) can be determined, as follows:

$$\tau_i = \tau_{234} \text{ x } ([^{234}Th_i]/\{[^{238}U]- [^{234}Th_I]\}), \quad (1)$$

where i = total, dissolved, or particulate (Baskaran et al., 1996; Santschi et al., 1999).

With $\tau_{234} = \lambda_{234}^{-1}$, λ_{234} = decay constant of ^{234}Th = 0.03 d^{-1}, $[^{234}Th_i]/[^{238}U_I]$ = activity ratio.

Removal residence times of total, dissolved and particulate ^{234}Th from the water and of ≤ 1 days, ≤ 6 hours and 12 hours, respectively (Table 16), were calculated from our measured ^{238}U and ^{234}Th activities in the water given in Table 15. These short residence times confirm the high efficiency of the holding ponds for particles, and particle-reactive elements such as Pu. .

From these results, we would predict that at water residence times in excess of one to a few days, most of the Pu and particle load entering a pond would be removed to the sediments. Water residence times are currently more than a month. If holding ponds were to remain, they should be designed with water residence times of more than a few days to maximize their effectiveness in reducing particle and actinide loads to down stream sites.

4.4. Colloidal Pu release potential from soil resuspension experiments:

Soil resuspension experiments were done with 0.5g of the fine fraction (<1mm) of RFETS soil (726 ± 68 pCi/g ^{239,240}Pu, 4.9% OC) in 190 ml of filtered tap water. These experiments demonstrate that Pu that is released during these experiments is mostly of colloidal nature (Table 17). Colloidal Pu is occurring as a pseudo-colloid, i.e., Pu associated with a different carrier phase. As shown in Figures 8 and 9, colloidal Pu concentrations increase over a resuspension time of 1-7 days, in the presence and absence of additional humic acid (at 59.2 mg DOM/L = 24.3 mg-C/L of DOC). Thus, there is a significant kinetic effect during the release of colloidal Pu. When the results from the control experiment are compared with those with increasing Aldrich humic acid (41% OC) concentrations, it becomes clear that colloidal Pu concentrations significantly increase with increasing humic acid concentrations. Soil humic acids extracted from uncontaminated Rocky Flats soils (using the procedure given in appendix 3) contained 16% OC. The soil extracted humic acids turned out to be considerably more effective than the commercial Aldrich humic acids in remobilizing Pu from contaminated soils into the water, given that concentrations are expressed in dissolved organic carbon, DOC, rather than dissolved organic matter (Figures 10 and 11). DOC concentrations in soil solutions (but not in streams), and organic carbon of suspended matter can be of similar magnitude as the concentrations that were used in these lab experiments. Also, pH values during soil resuspension experiments did not change by more than a few tenths of a pH unit, i.e., pH increased from 8.2 to 8.6 in the Controls and 8.5 to 8.6 in the presence of commercial and extracted humic acids (120ppm), after three days of resuspension.

There was only a small difference between the control and alginic acid (Figure 12), regardless of the concentration range, when similar resuspension times were compared. Soil resuspension experiments with the bacteria-derived acid polysaccharide Xanthen, revealed that it acts as a particle-glue, i.e., it exhibited strong particle aggregation effects (e.g., strong filter clogging), but showed insignificant effects on the colloid production potential of resuspended soils, which is consistent with its particle aggregating effects. Its strongest effects is thus on increasing the cohesiveness of particles.

When results at different suspended particulate matter concentrations, after 4-5 days of resuspension, are compared, it is apparent that Pu remobilization is inversely proportional to the soil particle-water ratio (Figure 13). The ratio of resuspended soil to water is thus an important parameter in predicting Pu colloid formation. The phase partitioning coefficient (Rp {in L/kg} = ratio of particulate concentration {in pCi/kg} to the filter-passing concentration {in pCi/L}) for Pu was inversely proportional to suspended particulate matter concentration (Cp, in kg/L), with $Rp = 10^{3.2}\ Cp^{-0.74}$, or $\log(Rp) = 3.2 - 0.74 \log(Cp)$ (Figure 13). The observation of Rp being a function of particle concentration is consistent with the presence of colloidal Pu in the filter-passing fraction, and is called "particle concentration effect". This phenomenon was previously ascribed to the presence of colloidal species of the analyte, e.g., for Th(IV) and other particle-reactive metals (e.g., Honeyman and Santschi, 1988, 1989). It suggests that higher colloidal Pu concentrations are also found at times of greater amounts of resuspended soils in the water. Even though such an effect might be caused by an organic matter sub-fraction, this particle-concentration effect is much smaller for organic matter itself (Figure 14).

The particle-concentration-effect is, however, also evident from the field samples we collected in1998 to 2000 (including data from Santschi et al., 1998, 1999b, see Figure 13). Field-derived Rp values for Pu in GS10 and GS03 waters are, however, lower at equivalent suspended matter concentrations (Cp), possibly due to differences in organic matter content and/or physical forms of Pu (particle size distribution). It suggests that Pu in soils further away from source areas might be in a more “weathered form”, i.e., they are associated with finer particles and colloids. As a consequence, colloidal fractions of Pu in field samples amount to 30-70 % of the total Pu in the field samples, at suspended matter concentrations of 30 - 120 mg/L, with the remainder in the particulate fraction. Despite the lowering of the Rp value with increasing Cp concentration, only 0.1 - 1 % of the Pu was remobilized from the soil collected near the 903 Pad(Figure 15). Similar proportions of Pu were remobilized with increased DOC concentrations.

While it is true that most of the relatively high Pu concentrations are coincident with high suspended matter concentrations, which contain 0.1-1 pCi/g of Pu, a much greater proportion of Pu in samples from GS03 and GS10 is in a colloidal rather than particulate form. As mentioned before, this might be due to more weathered Pu further away from sources, and more particulate forms of PuO_2 near the 903 Pad, the site where the soil for the laboratory experiments was taken. It is also likely that in the field, the quality (i.e., composition, e.g., humic acids vs aquagenic organic matter such as acid polysaccharides), in addition to the quantity of DOC, will play an important role in remobilizing Pu from soils.

Regardless, soil resuspension experiments suggest that colloidal Pu release during soil erosion events increases as a function of

a) suspended particulate matter concentration,
b) resuspension time, and
c) humic acid (DOC) concentrations.

Which one of these is the dominant factor in the field will require further analysis of field data.

Pu release from resuspended particles slightly decreases as a function of alginic acid concentrations in the water. Alginic acid, an excretion product by both algae and bacteria in soil and water, has an aggregating effect on particles, while humic acids, the main degradation product of plant matter in soils, generally has a dispersing effect.

These results thus suggest that the addition of colloidal Pu from sediment resuspension and soil erosion to stream water during pond discharge and storm run-off could increase the total Pu concentration, over that of particulate Pu alone, by a factor of 2-4. The increase depends on the amount of eroded soil particles in suspension, the length of time the soil is resuspended, as well as the humic acid concentration of the soil. This conclusion is in agreement with all the field data, which show a good correlation between total Pu concentrations and TSS (total suspended solids, Cp) in the water at most measuring stations. Scatter around the least squares line is approximately around a factor of 2-4, which can be produced by having colloidal Pu, in addition to particulate Pu, in the water rather than only variable Pu concentrations in suspended matter.

4.5. Implications of soil erosion studies: Importance of organic matter for Pu remobilization:

One of the conclusions of the soil resuspension experiments is that DOC concentrations have a major effect in remobilizing colloidal Pu into solution. DOC concentrations measured in 1998, 1999 and 2000 stream samples varied from 4 to13 mg C/L, with no obvious correlation with Pu concentrations. This could, however, have been due to compositional differences of DOM between spring and summer, aquagenic vs pedogenic organic matter. Furthermore, both quantity as well as the quality of DOC in soil solutions could indirectly affect the extent of soil aggregation/disaggregation, and thus, of Pu remobilization.

5. Conclusions

1. Pond and storm discharge samples collected in spring 2000 showed low Pu concentrations, with a large fraction of the Pu (~90 %) associated with suspended particulate matter, and the 0.5 µm filter-passing fraction of Pu being mostly in colloidal forms. Clay- and organic matter - rich colloids are present in different sizes, as was evident from elemental analyses and TEM and EDAX images.
2. Scavenging and particle residence times in Pond B-5, derived from measurements of $^{238}U/^{234}Th$ disequilibria in the water, were of the order of a less than 4 hours to 1 day or less, respectively, supporting the effectiveness of the ponds in reducing particle and Pu loads. From these results, we would predict that at water residence times in excess of one to a few days, most of the Pu and particle load entering a pond would be removed to the sediments. Water residence times are currently more than a month. If holding ponds were to remain, they should be designed with water residence times of more than a few days to maximize their effectiveness in reducing particle and actinide loads to down stream sites. While these short Th(IV)-derived residence times are typical for aquatic systems with suspended particulate matter concentrations of 1-100 mg/L, more measurements are needed to relate Th(IV)-derived residence times to prevailing suspended particulate matter concentrations.
3. Soil resuspension experiments suggest that Pu release during soil erosion events increases as a function of a) suspended particulate matter concentration, b) resuspension time, and c) humic acid (DOC) concentrations. Pu release from resuspended particles slightly decreases as a function of alginic acid (an excretion product of algae and bacteria) concentrations in the water. Soil resuspension experiments were conducted at DOC concentrations typical of soil solutions, but elevated compared to typical stream concentrations. Humic acids extracted from Rocky Flats soils were considerably more effective than commercially available Aldrich Humic Acid in remobilizing colloidal Pu from Rocky Flats soils into the water. These results suggest that higher colloidal Pu found in stream water during pond discharge and storm run-off could be regulated by the length of time the soil is resuspended, as well as the humic acid concentration of the soil. Soil resuspension experiments with the bacteria-derived acid polysaccharide Xanthen may act as a particle-glue, i.e., exhibited strong particle aggregation effects (e.g., strong filter clogging), but showed only insignificant effects on the colloid production potential of resuspended soils, consistent with its more aggregating nature. Its strongest effects is thus in increasing the effective particle size of aggregates, thus reducing soil erosion, rather than in reducing the colloidal Pu production of already resuspended soil.
4. The ratio of resuspended soil to water is also important in regulating Pu colloid formation. The phase partitioning coefficient, Rp (in L/kg), was inversely proportional to suspended particulate matter concentration (Cp, in kg/L), with $Rp = 10^{3.2}\ Cp^{-0.74}$, or $\log(Rp) = 3.2 - 0.74 \log(Cp)$. A comparison between these soil samples with elevated Pu concentrations and field data from streams at GS3 and GS10 reveals parallel relationships, with lower Rp values in field samples than observed during the soil resuspension experiments. Inverse relationships between Rp and Cp have been described before for Th(IV) (e.g., Honeyman and Santschi, 1988, 1989), and are a consequence of the presence of colloidal forms of Pu in the filter-passing fraction.

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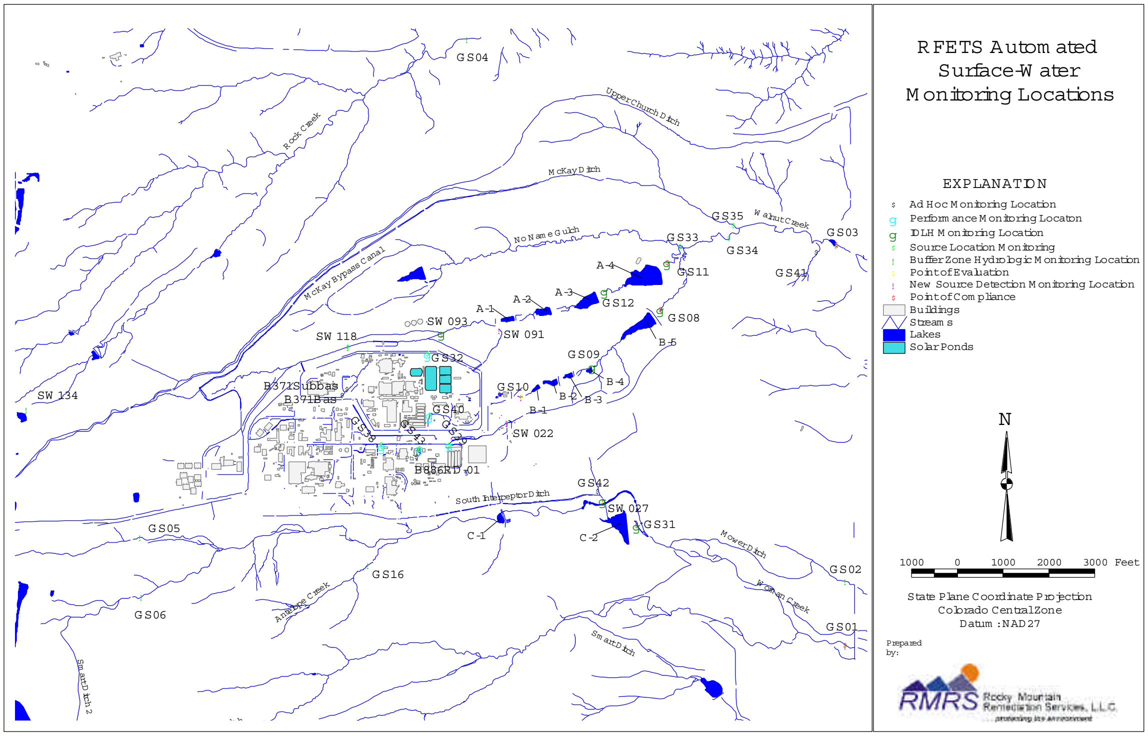


Figure 1. Map of sampling locations, with GS10 at the upstream end of the B Series ponds, and GS03 on Walnut Creek near Indiana Street.

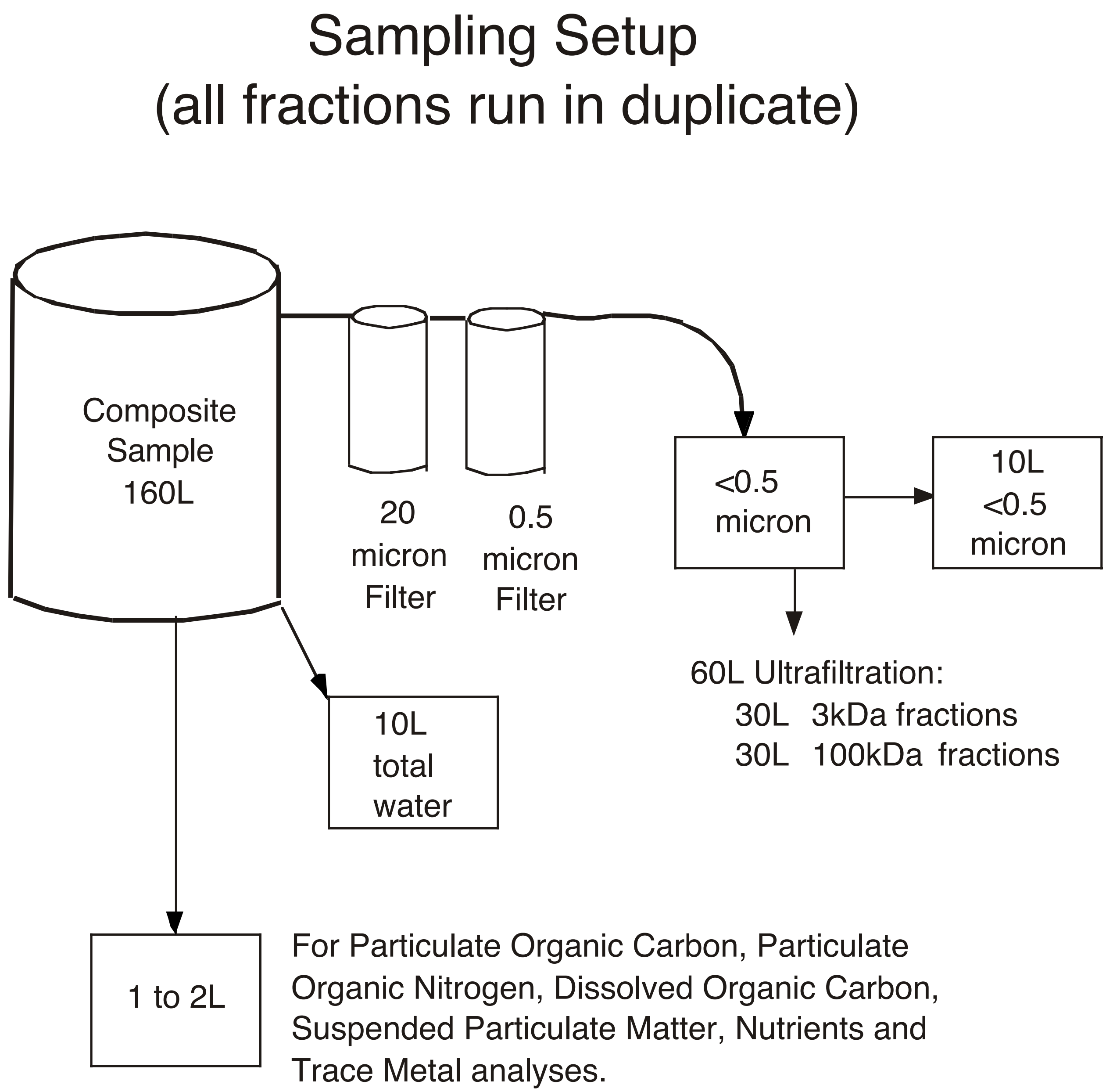


Figure 2. Schematic of field sampling.

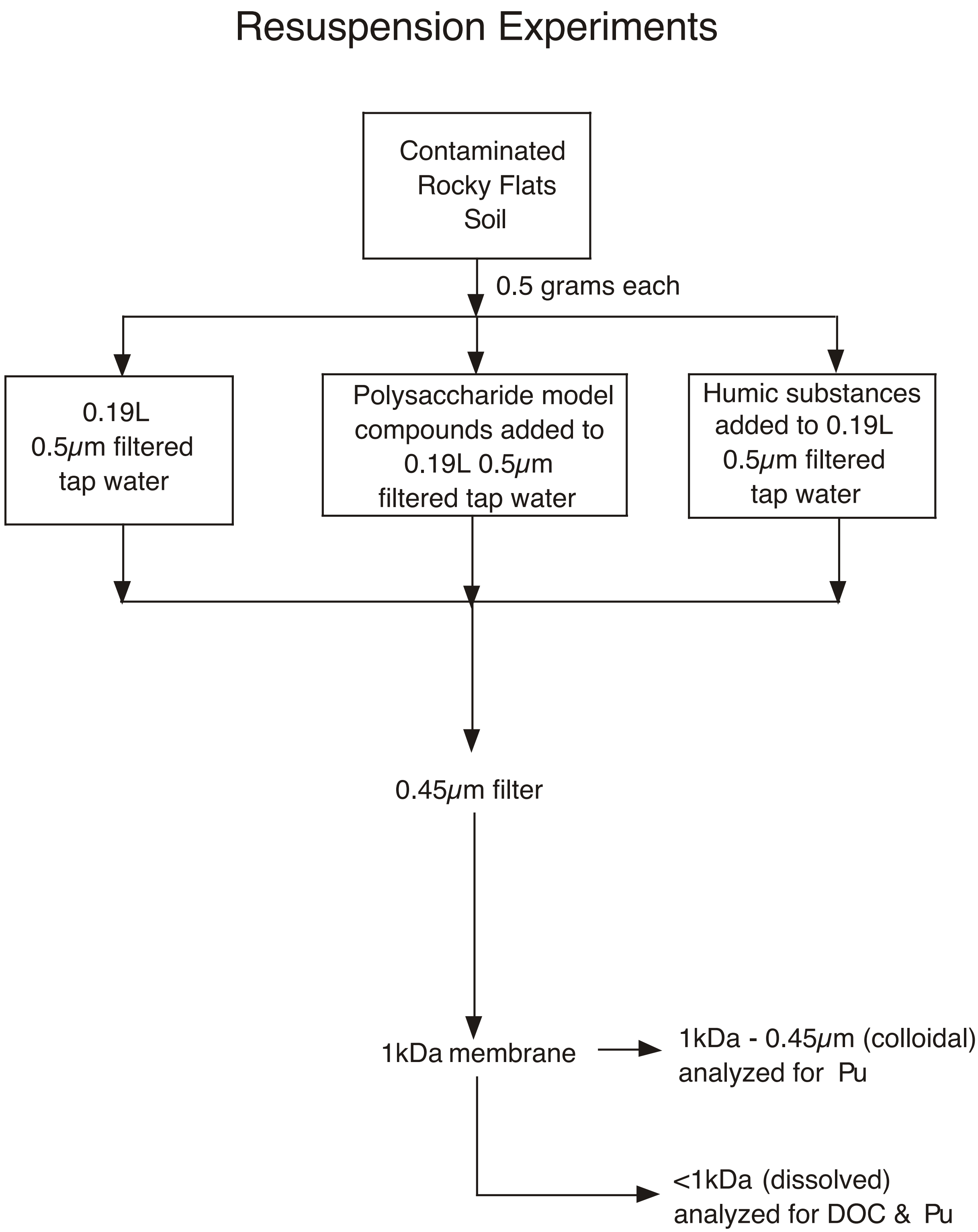
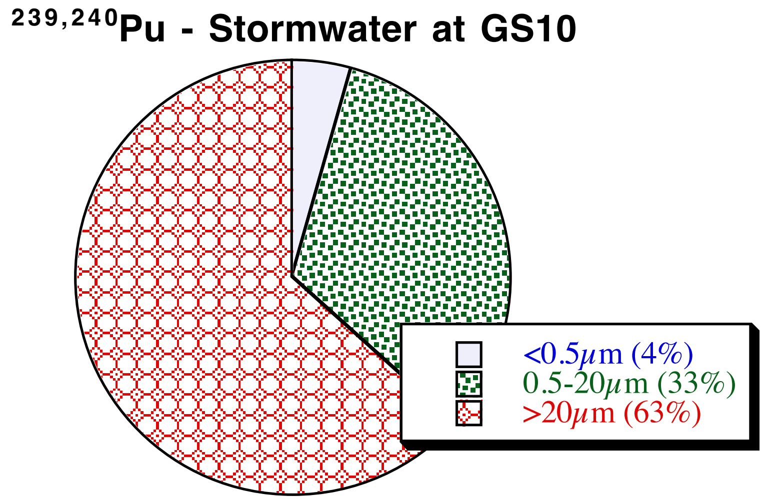


Figure 3. Schematic of soil resuspension experiments. Stirred cell ultrafiltration was only carried out in selected samples.



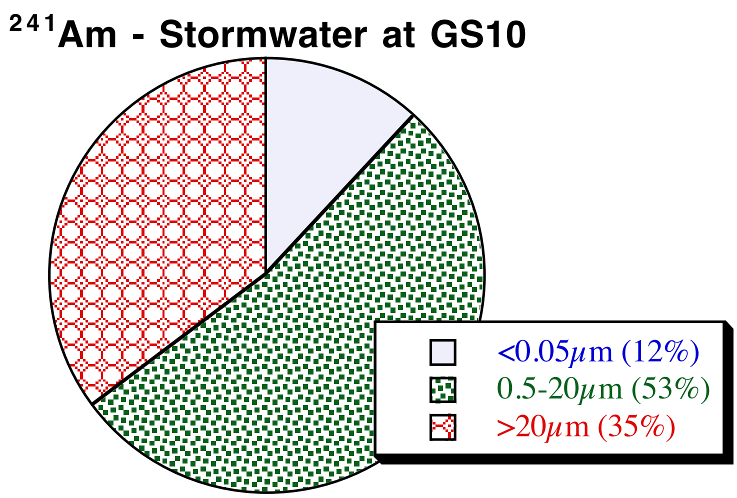
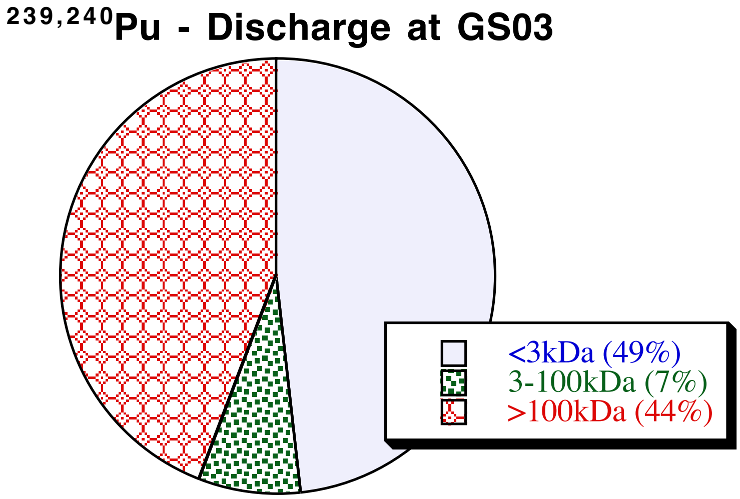


Figure 4. Partitioning of ^{241}Am and ^{239,240} Pu between 0.5μm filter-passing and particulate phases.



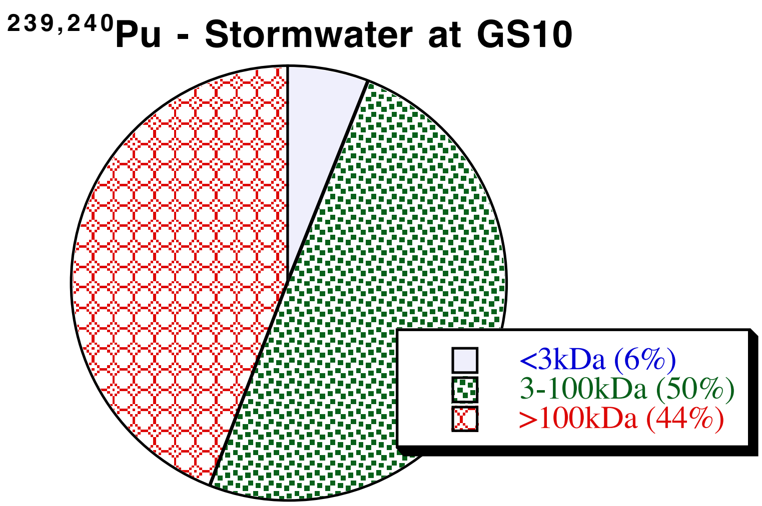
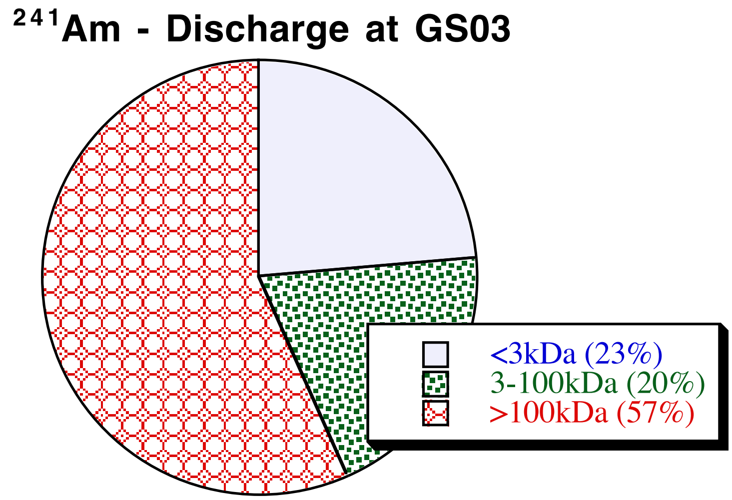


Figure 5. Partitioning of ^{239,240}Pu between ultrafilter-size fractions within the 0.5μm filter-passing fraction where <3kDa = measured value, >100kDa = measured 100kDa – 0.5μm + 100kDa wash, and 3-100kDa = Total – (<3kDa + >100kDa).



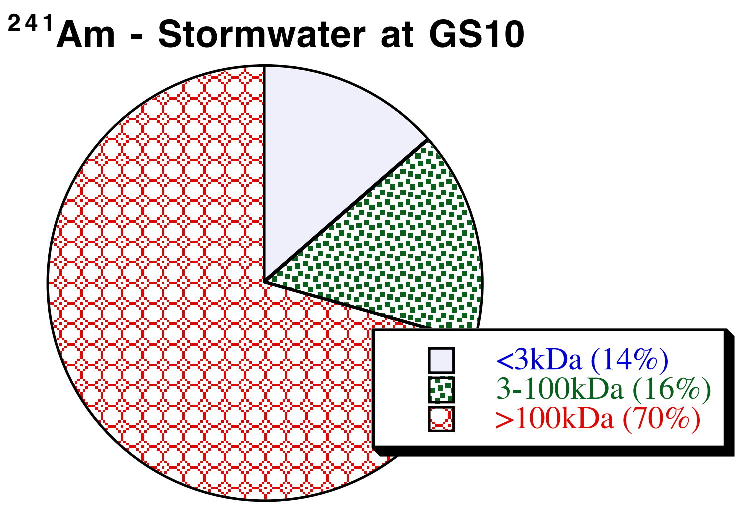


Figure 6. Partitioning of ^{241}Am between ultrafilter-size fractions within the 0.5μm filter-passing fraction where <3kDa = measured value, >100kDa = measured 100kDa – 0.5μm + 100kDa wash, and 3-100kDa = Total – (<3kDa + >100kDa).

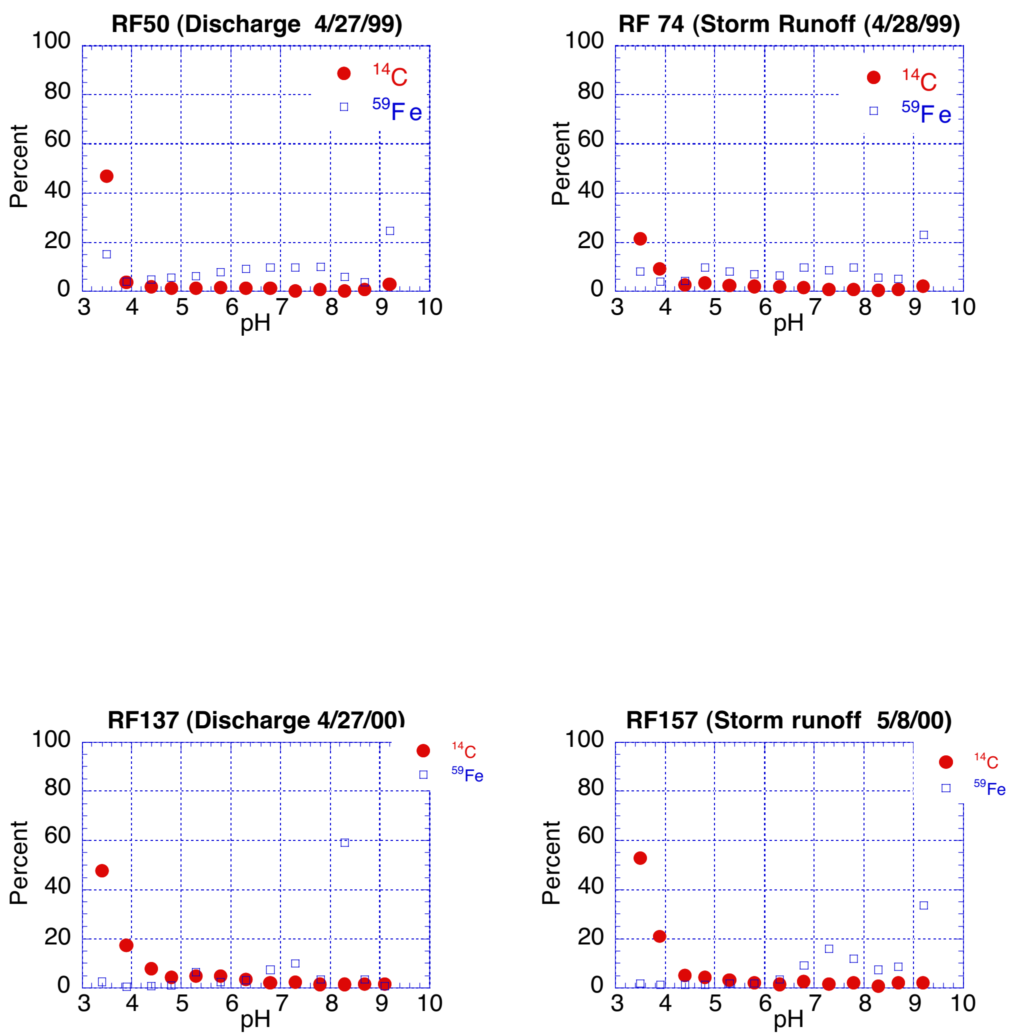


Figure 7. Comparison of gel electrophoresis results for colloids sampled in 1999 and 2000. ^{14}C labels the –OH sites of sugars which are negatively charged showing strong functional groups with pKa≤3.5. ^{59}Fe labels the –OH sites of Fe oxyhydroxides and clay minerals which are positively charged with pKa≥9.5.

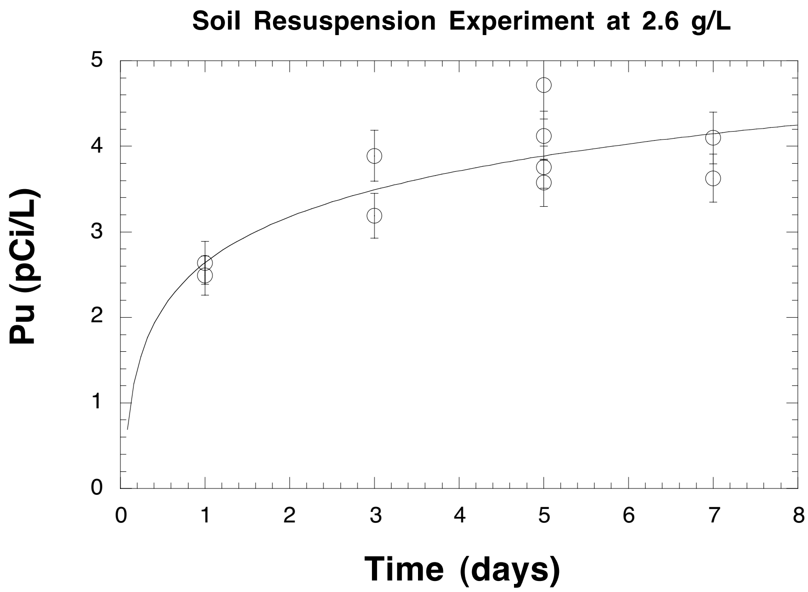


Figure 8. Kinetics of colloidal Pu remobilization during soil resuspension, with no humic acids added. DOC concentrations in solution, resulting from soil resuspension alone, ranged from 2.5 to 2.7 mg/L DOC (0.5 g/190 ml).

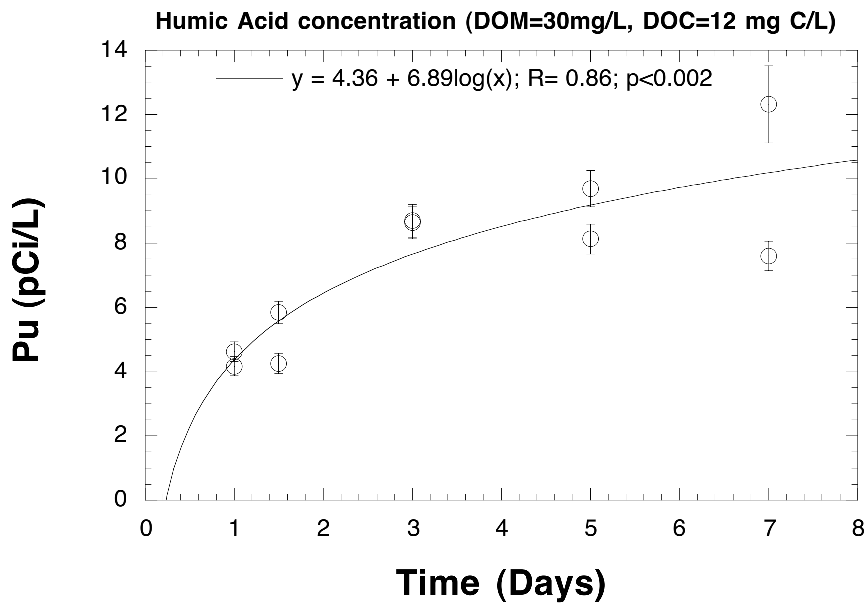


Figure 9. Kinetics of colloidal Pu remobilization during soil resuspension, with a constant amount of Aldrich humic acid added to the experimental solution (0.5 g/190 ml).

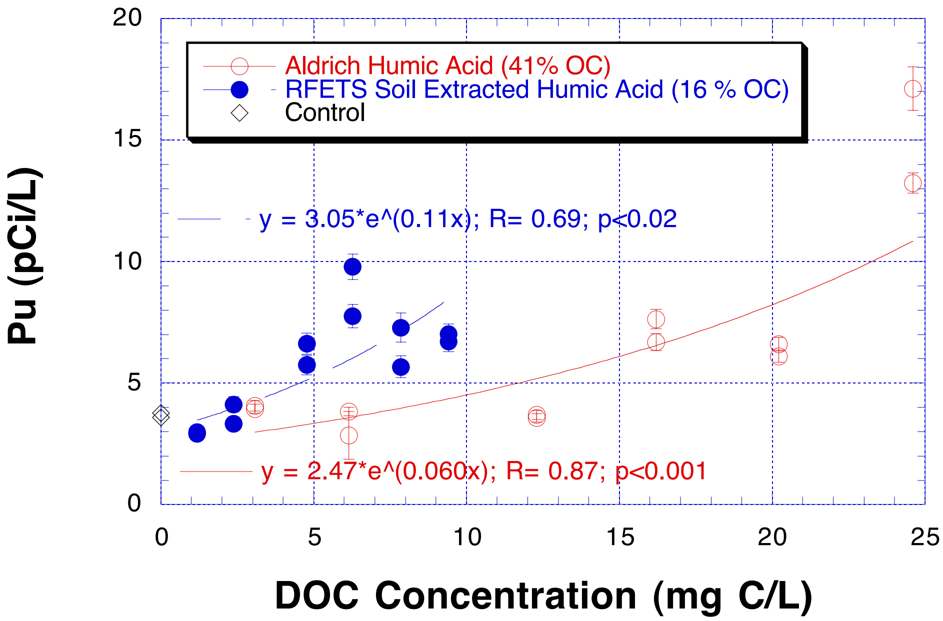


Figure 10. Comparison between Aldrich (41 % OC) and RFETS soil extracted (16 % OC) humic acid additions to the experimental solutions at resuspension times ranging from 3-7 days (0.5 g/190 ml). DOC concentration shown is concentration added to the solution.

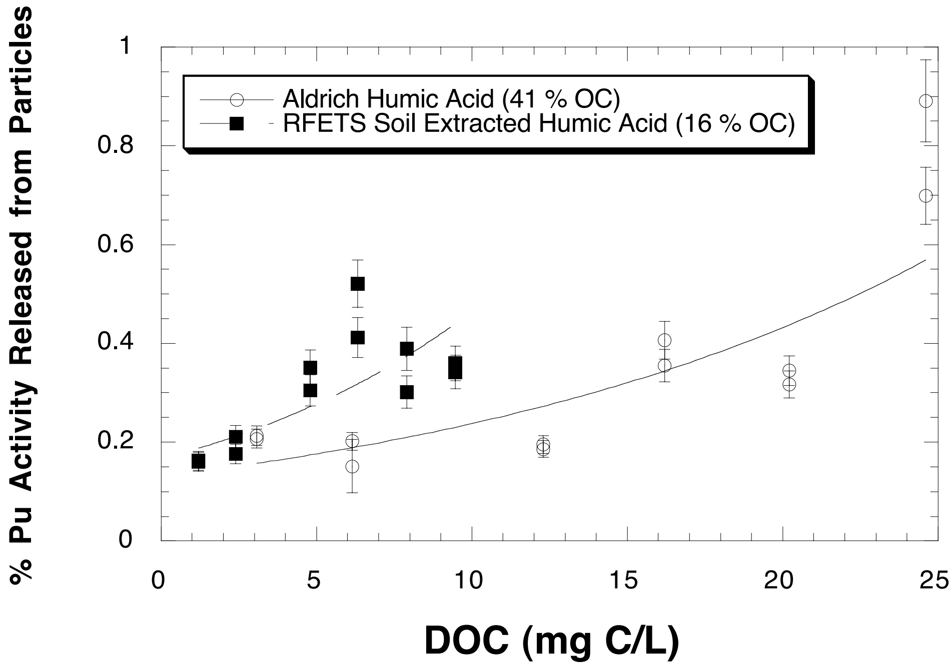


Figure 11. Percent Pu released from the application of different types of humic acids during 3-7 days of soil resuspension (0.5 g/190 ml).

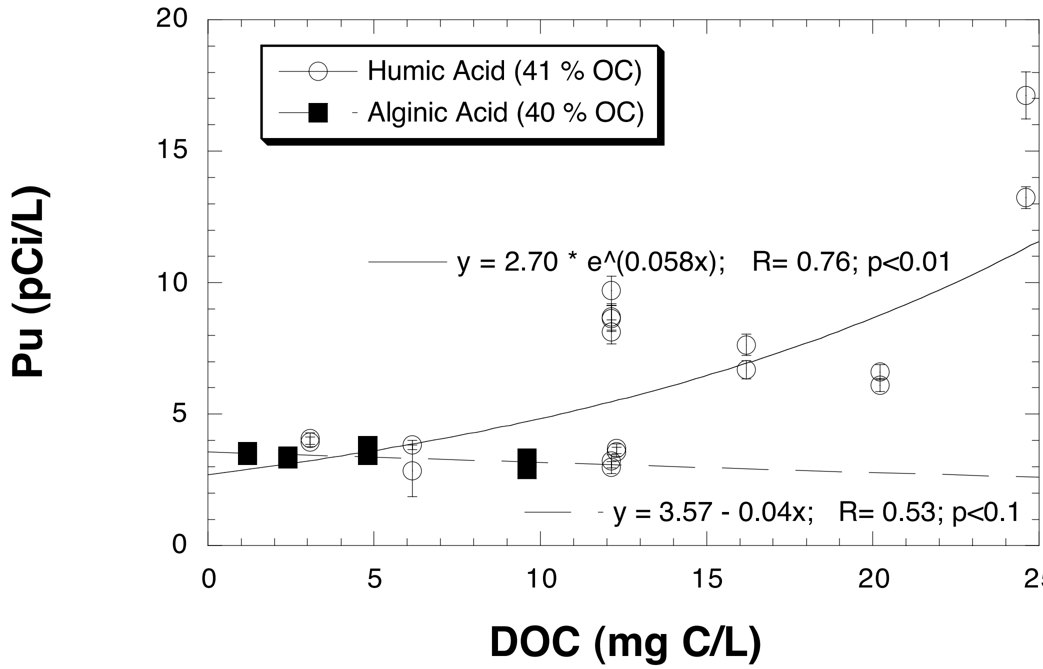


Figure 12. Comparison between Aldrich Humic Acid treated and Alginic Acid treated Pu remobilization experiments by soil resuspension (0.5 g/190 ml).

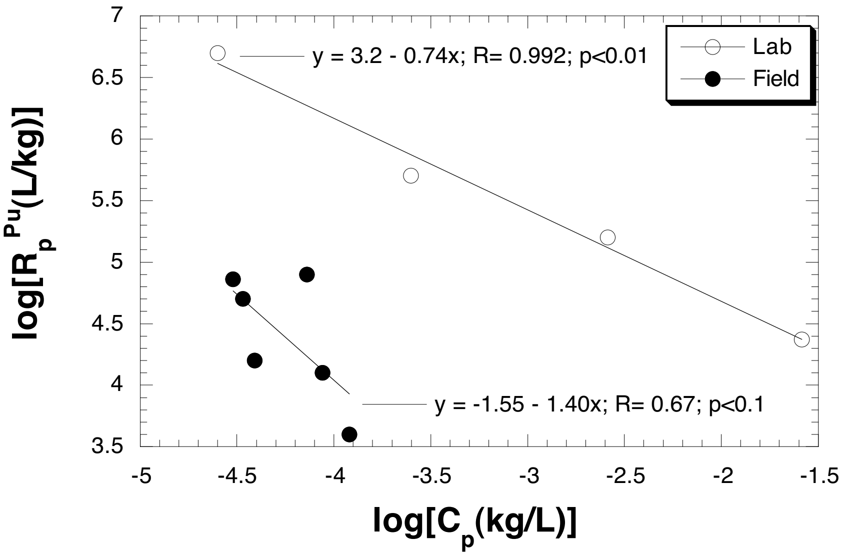


Figure 13. Relationship between total suspended particulate matter (or: solids) concentration (Cp) and phase partition coefficient (Rp) of Pu during soil resuspension experiments, after 4-5 days of particle resuspension, with no additional humic acid added. As Figures 8 and 9 demonstrate, differences between 4-5 days of resuspension are not significant. Laboratory results with RFETS soil taken near 903 Pad are also compared with field results from our water samples taken from GS03 and GS10 during 1998, 1999, and 2000.

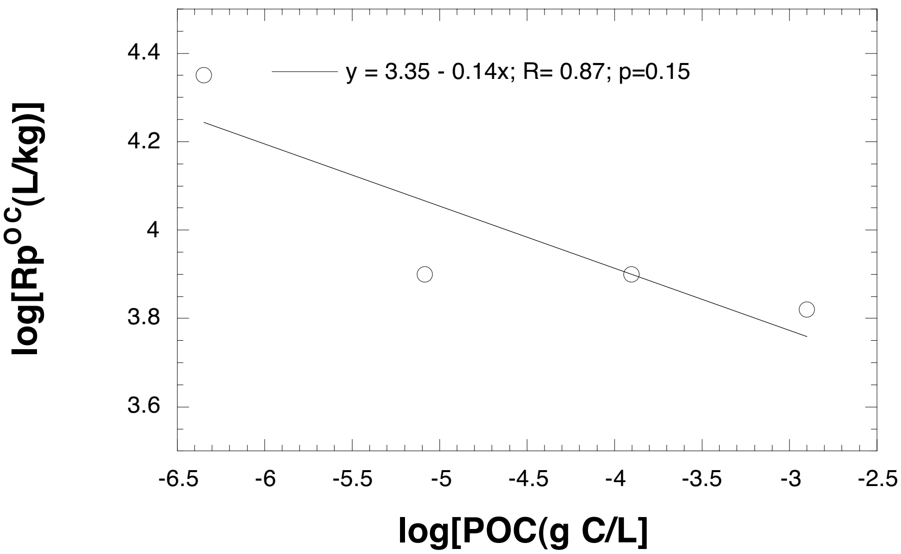


Figure 14. Relationship between total suspended particulate matter (or: solids) concentration (Cp) and phase partition coefficient (Rp) of organic carbon during soil resuspension experiments, after 4-5 days of particle resuspension, with no additional humic acid added.

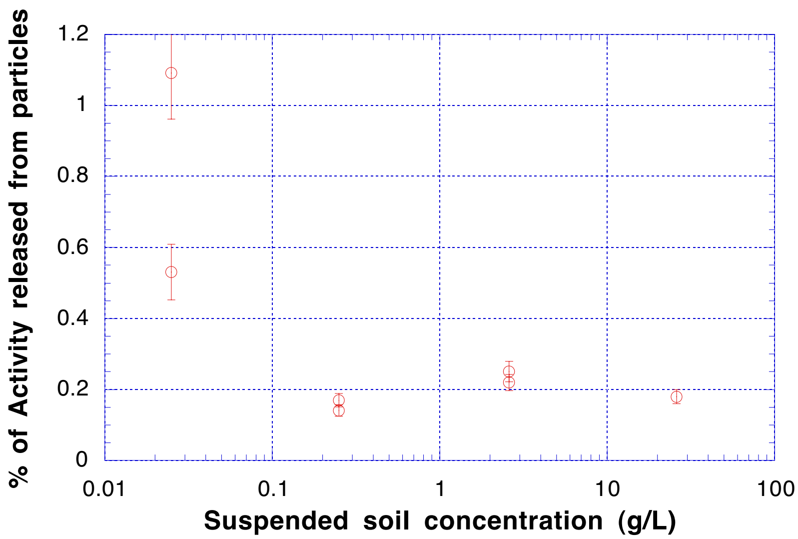


Figure 15. Fraction of soil-bound Pu which is released as a pseudo-colloid during simulated soil resuspension experiments, after 4-5 days of resuspension.

Table 1a. Summary of ^{239,240}Pu in duplicate samples A & B (SD = propagated 1 standard deviation based on α-counting data).

Sample ID	Fraction	A (pCi/l)	SD	B (pCi/l)	SD	Avg	SD
4/27/00	Total	0.0571	0.0018	-	-	-	-
(Discharge)	<0.5µm	0.0207	0.0014	-	-	-	-
	>20µm	-	-	-	-	-	-
	0.5-20µm	-	-	-	-	-	-
	100kDa–0.5µm	0.0068	0.0007	0.0006	0.0005	0.0037	0.0009
	<100kDa	0.0038	0.0006	0.0022	0.0008	0.0030	0.0010
	100kDa-Wash	0.0163	0.0006	0.0001	0.0005	0.0082	0.0078
	Sum of Fractions	0.0269	0.0011	0.0029	0.0011	0.0149	0.0016
	3kDa – 0.5µm	-	-	0.0100	0.0008	-	-
	<3kDa	-	-	0.0131	0.0012	-	-
	3kDa -Wash	-	-	0.0016	0.0006	-	-
	Sum of Fractions	-	-	0.0247	0.0016	-	-

Sample ID	Fraction	A (pCi/l)	SD	B (pCi/l)	SD	Avg	SD
5/8/00	Total	0.0726	0.0043	0.1056	0.0043	0.0891	0.0006
(Storm)	<0.5µm	0.0124	0.0013	0.0053	0.0012	0.0885	0.0018
	>20µm	-	-	0.0773	0.0065	-	-
	0.5-20µm	0.0828	0.0021	0.0396	0.0016	0.0612	0.0026
	100kDa–0.5µm	0.0070	0.0006	0.0058	0.0007	0.0064	0.0009
	<100kDa	0.0033	0.0007	0.0041	0.0012	0.0037	0.0014
	100K-Wash	0.0014	0.0005	0.0001	0.0005	0.0008	0.0007
	Sum of Fractions	0.0117	0.0010	0.0100	0.0015	0.0109	0.0018
	3kDa – 0.5µm	0.0148	0.0012	0.0158	0.0014	0.0153	0.0018
	<3kDa	0.0012	0.0006	0.0007	0.0007	0.0010	0.0009
	3K-Wash	-	-	-	-	-	-
	Sum of Fractions	0.0160	0.0013	0.0165	0.0016	0.0163	0.0021

- = tracer recovery <10%

Table 1b. Summary of ^{241}Am data in duplicate samples A & B (SD = propagated 1 standard deviation based on α-counting data).

Sample ID	Fraction	A(pCi/l)	SD	B(pCi/l)	SD	Avg	SD
4/27/00	Total	0.0011	0.0009	0.0026	0.0008	0.0019	0.0012
(Discharge)	<0.5µm	0.0004	0.0007	0.0003	0.0007	0.0004	0.0010
	>20µm	-	-	-	-	-	-
	0.5-20µm	-	-	-	-	-	-
	100kDa–0.5µm	0.0007	0.0004	-	-	-	-
	<100kDa	0.0022	0.0004	-	-	-	-
	100K-Wash	0.0065	0.0005	-	-	-	-
	Sum of Fractions	0.0094	0.0008	-	-	-	-
	3kDa – 0.5µm	0.0030	0.0008	-	-	-	-
	<3kDa	0.0030	0.0005	-	-	-	-
	3K-Wash	0.0067	0.0006	0.0029	0.0006	0.0048	0.0008
	Sum of Fractions	0.0127	0.0011	-	-	-	-

Sample ID	Fraction	A(pCi/l)	SD	B(pCi/l)	SD	Avg	SD
5/8/00	Total	0.0602	0.0040	0.0768	0.0033	0.0685	0.0051
(Storm)	<0.5µm	0.0017	0.0005	0.0092	0.0009	0.0055	0.0010
	>20µm	-	-	-	-	-	-
	0.5-20µm	0.1327	0.0028	0.0406	0.0022	0.0867	0.0036
	100kDa–0.5µm	0.0030	0.0005	0.0041	0.0004	0.0036	0.0006
	<100kDa	0.0023	0.0005	0.0030	0.0004	0.0027	0.0006
	100K-Wash	0.0031	0.0006	0.0020	0.0004	0.0026	0.0007
	Sum of Fractions	0.0084	0.0009	0.0091	0.0007	0.0089	0.0011
	3kDa – 0.5µm	0.0106	0.0009	0.0050	0.0004	0.0078	0.0010
	<3kDa	0.0018	0.0006	0.0006	0.0004	0.0012	0.0007
	3K-Wash	0.0009	0.0004	0.0008	0.0004	0.0009	0.0006
	Sum of Fractions	0.0133	0.0012	0.0064	0.0007	0.0099	0.0014

- = tracer recovery <10%

Table 2. Summary of Pu and Am data used in Figures 4-6.

Sample ID	Size Fraction	^{239,240}Pu Activity (pCi/L)	Note	%	^{241}Am Activity (pCi/L)	Note	%
4/27/00 Discharge	<3kDa	0.0131	B	49	0.0030	A	23
	3-100kDa	0.0020	#	7	0.0025	#	20
	>100kDa	0.0119	Avg	44	0.0072	A	57
	Total	0.0269	A	100	0.0127	A	100
5/8/00 Stormwater	<0.5µm	0.0053	B	4	0.0092	B	12
	0.5-20µm	0.0396	B	33	0.0406	B	53
	>20µm	0.0773	B	63	0.0270#	#	35
	Total	0.1222	*	100	0.0768	B	100
	<3kDa	0.0010	Avg	6	0.0012	Avg	14
	3-100kDa	0.0081	#	50	0.0014	#	16
	>100kDa	0.0072	Avg	44	0.0062	Avg	70
	Total	0.0163	Avg	100	0.0088	Avg	100

Note: A = from sample 'A', B= from sample 'B', Avg = average of samples 'A' & 'B', # = calculated by difference (total – other fractions), * = sum of fractions. The size fractions are defined as follows: <3kDa = measured, 3-100kDa = Total – (<3kDa +>100kDa), > 100kDa = measured 100kDa – 0.5μm + 100kDa wash.

Table 3 . Suspended Particulate Matter (SPM, ≥0.45µm) Concentration.

Sample ID	Filter #	SPM (mg/l)	Average (mg/l)	SD	SD (%)
4/27/00-A	8	36.4	34.4	2.8	8.2
"	10	32.4			
4/27/00-B	5	28.8	27.5	1.9	7.2
“	6	25.2			
"	7	28.4			
5/8/00	1	53.6	72.4	16.5	22.8
"	2	84.4			
"	4	79.2			

Table 4. Particulate Organic Carbon (POC, ≥0.7μm) Concentration.

Sample ID	Filter #	POC (mg-C/l)	Avg(mg/l)	SD	SD(%)
4/27/00	RF021	2.93			
"	RF022	3.29			
"	RF023	3.29	3.17	0.21	6.65
5/8/00	RF015	4.07			
"	RF016	3.96			
"	RF018	4.25	4.09	0.15	3.61

Detection limit = 0.003mg based on three times the SD of the blank values

Table 5. Particulate Organic Nitrogen (PON, ≥0.7µm) Concentration.

Sample ID	Filter #	PON (mg-N/l)	Avg(mg/l)	SD	SD(%)
4/27/00	RF021	0.52			
"	RF022	0.57			
"	RF023	0.63	0.57	0.05	8.98
5/8/00	RF015	0.43			
"	RF016	0.43			
"	RF018	0.43	0.43	0.00	0.00

Detection limit = 0.007mg based on three times the SD of the blank values

Table 6. C/N atomic Ratio of suspended particulate matter (≥0.7µm).

Sample ID	Filter #	POC(mg/l)	PON(mg/l)	C/N Ratio	Avg	SD	%OC	SD
4/27/00	RF021	2.93	0.52	6.54				
"	RF022	3.29	0.57	6.69				
"	RF023	3.29	0.63	6.14	6.46	0.29	10	1.1
5/8/00	RF015	4.07	0.43	11.13				
"	RF016	3.96	0.43	10.83				
"	RF018	4.25	0.43	11.63	11.19	0.40	5.7	1.3

Table 7. Dissolved organic carbon (DOC, <0.5μm) concentration.

Sample ID	Container #	DOC (ppm)	Avg (ppm)	SD (ppm)
4/27/00	A-1	4.232		
"	A-2	3.648	3.94	0.41
"	B-1	3.733		
"	B-2	3.453	3.59	0.20
5/8/00	A-1	5.094		
"	A-2	5.492		
"	A-3	11.92		
"	A-4	5.384		
"	A-5	5.413	6.66	2.94
"	B-1	5.431		
"	B-2	5.341		
"	B-3	5.082		
"	B-4	5.325		
"	B-5	5.237	5.28	0.13

Detection limit = 0.04ppm based on three times the SD of the blank values

Table 8. Carbon and Nitrogen (%) in colloids, as average of duplicate measurements.

Sample ID	Description	Size	%C	%N	C/N
RF137	Discharge	3kDa–0.5μm	3.10	0.21	14.81
RF147	Discharge	3kDa–0.5μm	3.87	0.24	16.52
RF157	Storm	3kDa–0.5μm	11.94	0.600	19.88
RF167	Storm	3kDa–0.5μm	10.68	0.60	19.88

Table 9. Trace Metal Concentrations in aqueous (4/27/00 and 5/8/00) and colloidal (RF137, RF147, RF157 and RF167) samples.

Sample ID	Subsample	Fe (ppb)	Al (ppb)	Mn (ppb)
Pond B5	total	45.55	35.24	70.72
	<0.5	26.13	23.34	38.50
	<0.5	26.92	24.55	42.37
	<0.5	26.57	25.84	41.21
4/27/00	A	40.31	15.81	35.14
	B	34.94	14.81	32.30
5/8/00	A	281.41	291.67	19.43
	B	279.67	290.43	21.87

Sample ID	Colloid size	Fe (mg/g)	Al (mg/g)	Mn (µg/g)
RF137	3kDa-0.5µm	0.15	0.07	86.05
RF147	3kDa-0.5µm	0.20	0.16	60.52
RF157	3kDa-0.5µm	10.30	13.32	131.41
RF167	3kDa-0.5µm	8.69	11.91	148.54

Detection limit = 0.030ppb (Fe), 0.34ppb (Al) and 0.05ppb (Mn) based on three times the SD of blank values

Table 10. Inorganic anion (Cl, NO_3, HPO_4, and SO_4) concentrations.

Sample ID	Subsample	Cl (ppm)	NO_3 (ppm)	HPO_4 (ppm)	SO_4 (ppm)
POND B5	4/26-1	319.03	86.08	0.88	46.74
	4/26-2	293.27	85.67	1.75	42.50
	4/26-3	308.55	85.55	1.0	36.06
	4/26-4	322.89	85.45	1.19	43.03
	4/27-5	314.26	83.47	1.06	37.63
	4/27-6	305.50			29.61
4/27/00	1	>250	7.83	-	61.78
	2	>250	7.71	-	
	3	>250	6.80	-	56.63
	4	>250	7.04	-	57.51
5/8/00	1	>250	28.20	-	7.87
	2	>250	28.35	-	7.93
	3	>250	28.25	-	7.77
	4	>250	28.04	-	7.74

Detection limits = 0.60ppm (Cl), 0.41ppm (NO_2), 0.17ppm (HPO_4), and 0.04 (SO_4) based on three times the SD of the 0.5ppm standard and 0.41ppm ($N0_2$) based on three times the SD of the 1ppm standard Note: NO_2 Concentrations are below detection limit (BD)

Table 11. Ancillary data taken at time of sampling by site personel.

Parameter	B5 4/26/00 10:45	B5 4/26/00 15:25	B5 4/27/00 9:10	GS03 4/27/00
pH	9.84	9.95	9.76	8.62
Temperature (˚C)	15.3	16.5	13.8	17.6
SC (mS/cm)	1.444	1.391	1.264	1.172
DO(mg/L)	14.47	15.98	14.34	7.62
Alkalinity(mg/L Ca)	110	112.5	100	131

Table 12. $^{239,240}Pu/^{241}Am$ ratios in dissolved, colloidal and particulate samples.

Date	Sample Name	Fraction	Pu/Am-1	SD	Pu/Am-2	SD	Pu/Am-Avg
4/27/00	Discharge	Total	51.91	1.34	-	-	-
		<0.5µm	51.75	6.13	-	-	-
		>20µm	-	-	-	-	-
		0.5-20µm	-	-	-	-	-
		100kDa–0.5µm	9.71	0.57	-	-	-
		<100kDa	1.73	0.05	-	-	-
		100K-Wash	2.51	0.01	-	-	-
		3kDa – 0.5µm	-	-	-	-	-
		<3kDa	-	-	-	-	-
		3K-Wash	-	-	0.55	0.24	-
5/8/00	Storm event	Total	1.21	0.00	1.38	0.08	2.58
		<0.5µm	7.29	0.22	0.58	0.14	7.87
		>20µm	-	-	-	-	-
		0.5-20µm	0.62	0.00	0.98	0.07	1.60
		100kDa–0.5µm	2.33	0.03	1.41	0.22	3.75
		<100kDa	1.43	0.07	1.37	0.44	2.80
		100K-Wash	0.45	0.03	0.05	0.25	0.50
		3kDa – 0.5µm	1.40	0.01	3.16	0.38	4.56
		<3kDa	0.67	0.11	1.17	1.40	1.83
		3K-Wash	-	-	-	-	-

Table 13. Phase distribution coefficients (R_p, l/kg or ml/g) of $^{239,240}Pu$ and ^{241}Am.

Isotope	Sampling Date	Sample Name	C_p (mg/l)	R_p (ml/g)
$^{239,240}Pu$	4/27/00	Discharge	34	$4.8\ 10^4$
$^{239,240}Pu$	5/8/00	Storm event		
^{241}Am	4/27/00	Discharge	72	$8.0\ 10^4$
^{241}Am	5/8/00	Storm event		

C_p (mg/l) is the concentration of suspended particulate matter.

Table 14. Percent of total activity recovered during isoelectric focusing. ^{14}C labels –OH sugars and ^{59}Fe labels –OH sites of Fe oxyhydroxides and clays.

pH	^{14}C RF137	^{14}C RF 157	^{59}Fe RF137	^{59}Fe RF 157
<3.9	47.79	52.90	2.50	1.96
3.9	17.15	20.84	0.45	1.26
4.4	7.67	5.08	0.70	1.29
4.8	4.05	4.09	0.86	1.29
5.3	4.79	3.01	6.49	1.90
5.8	4.79	2.19	2.22	1.97
6.3	3.52	1.34	2.95	3.42
6.8	2.04	2.48	7.29	9.29
7.3	2.44	1.55	9.95	15.99
7.8	1.31	2.17	3.40	11.86
8.3	1.31	0.62	59.06	7.35
8.7	1.64	1.88	3.55	8.74
>8.7	1.51	1.88	0.57	33.69

Table 15. Results of ^{238}U (ICP-MS) and ^{234}Th (Gamma Spectrometry) measurements of pond B5 water.

Day	#	[$^{234}Th_d$]*	[$^{234}Th_p$]	[$^{234}Th_t$] (dpm/L)	[$^{238}U_t$]	[$^{238}U_d$]	[$^{238}U_p$]
4/26	1	≤0.01	0.04±0.008	0.05	1.69±0.03	1.56±0.08	0.13±0.09
	2	≤0.003	0.01±0.002	0.013	"	"	"
4/27	1	≤0.02	0.03±0.004	0.05	"	"	"
	2	≤0.01	0.02±0.002	0.03	"	"	"
Average		≤0.01	0.025	0.036	1.69±0.03	1.56±0.08	0.13±0.09

*) Calculated from 3 times base line concentration of ^{234}Th gamma region (63 keV). Value also equal to MDA (minimum detectable activity).

Table 16. Results of total, dissolved and particulate ^{234}Th residence times in pond B5 water, calculated using eq. 1 (see text).

Day	#	τ_t	τ_d (days)	τ_p
4/26	1	≤1.1	≤0.22	0.84
	2	≤0.3	≤0.07	0.21
4/27	1	≤1.1	≤0.45	0.62
	2	≤0.6	≤0.22	0.42
Average		≤0.8	≤0.24	0.52

Table 17: Phase speciation results using a 1kDa stirred cell ultrafiltration after resuspending 0.5g RFETS soil in 190ml of filtered tap water.

	Fraction	pCi/L	SD	%	pCi/L	SD	%
Control	<0.45μm	2.812	0.253		5.547	0.409	
	0.45-1kDa	2.649	0.324	94.21	3.572	0.378	64.40
	<1kDa	1.011	0.209	35.97	0.896	0.153	16.15
	Filter leach	0.004	0.003	0.14	0.013	0.006	0.23
	Sum	3.665	0.386	130.33	4.481	0.408	80.77
Aldrich Humic Acid (59 mg/L)	<0.45μm	7.141	0.507		6.428	0.444	
	0.45-1kDa	4.565	0.430	63.94	4.819	0.506	74.98
	<1kDa	0.152	0.071	2.13	0.368	0.110	5.72
	Filter leach	0.043	0.010	0.60	0.074	0.019	1.15
	Sum	4.760	0.436	66.66	5.261	0.518	81.85

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

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September 29, 2000

Appendices

Appendix 1: Sample Identification Numbers

Table A1a: Summary of Field Samples.

ID#	LOCATION	COC#	DESCRIPTION	ANALYSIS
RF130	GS03	TAMU42700B	Total	A,B
RF131	GS03	TAMU42700B	>20 micron	A,B
RF132	GS03	TAMU42700B	0.5-20 micron	A,B
RF133	GS03	TAMU42700B	<0.5 micron	A,B
RF134	GS03	TAMU42700B	100kDa – 0.5 micron	A,B
RF135	GS03	TAMU42700B	<100kDa	A,B
RF136	GS03	TAMU42700B	100kDa wash	A,B
RF137	GS03	TAMU42700B	3kDa – 0.5 micron	A,B, C,D
RF138	GS03	TAMU42700B	<3kDa	A,B
RF139	GS03	TAMU42700B	3kDa wash	A,B,
RF140	GS03	TAMU42700B	Total	A,B
RF141	GS03	TAMU42700B	>20 micron	A,B
RF142	GS03	TAMU42700B	0.5-20 micron	A,B
RF143	GS03	TAMU42700B	<0.5 micron	A,B
RF144	GS03	TAMU42700B	100kDa – 0.5 micron	A,B
RF145	GS03	TAMU42700B	<100kDa	A,B
RF146	GS03	TAMU42700B	100kDa wash	A,B
RF147	GS03	TAMU42700B	3kDa – 0.5 micron	A,B
RF148	GS03	TAMU42700B	<3kDa	A,B
RF149	GS03	TAMU42700B	3kDa wash	A,B,
RF150	GS10	TAMU050800	Total	A,B
RF151	GS10	TAMU050800	>20 micron	A,B
RF152	GS10	TAMU050800	0.5-20 micron	A,B
RF153	GS10	TAMU050800	<0.5 micron	A,B
RF154	GS10	TAMU050800	100kDa – 0.5 micron	A,B
RF155	GS10	TAMU050800	<100kDa	A,B
RF156	GS10	TAMU050800	100kDa wash	A,B
RF157	GS10	TAMU050800	3kDa – 0.5 micron	A,B,C,D
RF158	GS10	TAMU050800	<3kDa	A,B
RF159	GS10	TAMU050800	3kDa wash	A,B
RF160	GS10	TAMU050800	Total	A,B
RF161	GS10	TAMU050800	>20 micron	A,B

ID#	LOCATION	COC#	DESCRIPTION	ANALYSIS
RF162	GS10	TAMU050800	0.5-20 micron	A,B
RF163	GS10	TAMU050800	<0.5 micron	A,B
RF164	GS10	TAMU050800	100kDa – 0.5 micron	A,B
RF165	GS10	TAMU050800	<100kDa	A,B
RF166	GS10	TAMU050800	100kDa wash	A,B
RF167	GS10	TAMU050800	3kDa – 0.5 micron	A,B
RF168	GS10	TAMU050800	<3kDa	A,B
RF169	GS10	TAMU050800	3kDa wash	A,B
RF170			Blank	A,B
RF171			Blank spike	A,B
RF172			Blank	A,B
RF173			Blank spike	A,B
RF174			Blank	A,B
RF175			Blank spike	A,B
RF176			Blank	A,B
RF177			Blank spike	A,B

A= ^{241}Am activity; B= ^{239,240} Pu activity, C=IEF, D=TEM

Table A1b: Summary of Resuspension Data

ID	RIN	Type	Sed wt/vol (g/ml)	Duration Time(d)	Conc (mg/L)	Analysis
P336	99A3372	Control	0.5/20	4	0	B,E
P337	99A3372	Control	0.5/20	4	0	B,E
P319	99A3372	Control	0.5/100	4	0	B,E
P321	99A3372	Control	0.5/100	4	0	B,E
P265	99A3372	Control	0.5/190	5	0	B,E
P267	99A3372	Control	0.5/190	5	0	B,E
P279	99A3372	Control	0.1/400	4	0	B,E
P281	99A3372	Control	0.1/400	4	0	B,E
P315	99A3372	Control	0.5/2000	4	0	B,E
P317	99A3372	Control	0.5/2000	4	0	B,E
P291	99A3372	Control	0.5/190	1	0	B,E
P293	99A3372	Control	0.5/190	1	0	B,E
P295	99A3372	Control	0.5/190	3	0	B,E
P297	99A3372	Control	0.5/190	3	0	B,E
P299	99A3372	Control	0.5/190	5	0	B,E
P301	99A3372	Control	0.5/190	5	0	B,E
P303	99A3372	Control	0.5/190	7	0	B,E
P305	99A3372	Control	0.5/190	7	0	B,E
P178	99A3372	Control	0.5/190	5	0	B,E
P180	99A3372	Control	0.5/190	5	0	B,E
P233	99A3372	Aldrich Humic Acid	0.5/190	4	7.5	B,E
P235	99A3372	Aldrich Humic Acid	0.5/190	4	7.5	B,E
P237	99A3372	Aldrich Humic Acid	0.5/190	4	15	B,E
P239	99A3372	Aldrich Humic Acid	0.5/190	4	15	B,E
P241	99A3372	Aldrich Humic Acid	0.5/190	4	30	B,E
P243	99A3372	Aldrich Humic Acid	0.5/190	4	30	B,E
P269	99A3372	Aldrich Humic Acid	0.5/190	5	39.5	B,E
P271	99A3372	Aldrich Humic Acid	0.5/190	5	39.5	B,E
P273	99A3372	Aldrich Humic Acid	0.5/190	5	49.3	B,E
P275	99A3372	Aldrich Humic Acid	0.5/190	5	49.3	B,E
P245	99A3372	Aldrich Humic Acid	0.5/190	4	60	B,E
P247	99A3372	Aldrich Humic Acid	0.5/190	4	60	B,E
P283	99A3372	Aldrich Humic Acid	0.1/400	4	30	B,E
P285	99A3372	Aldrich Humic Acid	0.1/400	4	30	B,E
P307	99A3372	Aldrich Humic Acid	0.5/190	1	29.6	B,E
P309	99A3372	Aldrich Humic Acid	0.5/190	1	29.6	B,E
P311	99A3372	Aldrich Humic Acid	0.5/190	3	29.6	B,E
P313	99A3372	Aldrich Humic Acid	0.5/190	3	29.6	B,E
P323	99A3372	Aldrich Humic Acid	0.5/190	5	29.6	B,E
P325	99A3372	Aldrich Humic Acid	0.5/190	5	29.6	B,E

ID	RIN	Type	Sed wt/vol (g/ml)	Duration Time(d)	Conc (mg/L)	Analysis
P331	99A3372	Aldrich Humic Acid	0.5/190	7	29.6	B,E
P333	99A3372	Aldrich Humic Acid	0.5/190	7	29.6	B,E
P182	99A3372	Aldrich Humic Acid	0.5/190	5	29.6	B,E
P184	99A3372	Aldrich Humic Acid	0.5/190	5	29.6	B,E
P338	99A3372	Aldrich Humic Acid	0.5/190	4	7.5	B,E
P339	99A3372	Aldrich Humic Acid	0.5/190	4	7.5	B,E
P340	99A3372	Aldrich Humic Acid	0.5/190	4	15	B,E
P341	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	15	B,E
P341	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	15	B,E
P342	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	30	B,E
P343	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	30	B,E
P344	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	39.5	B,E
P345	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	39.5	B,E
P346	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	49.3	B,E
P347	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	49.3	B,E
P348	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	59.2	B,E
P349	99A3372 & 00R0972	Extracted Humic Acid	0.5/190	4	59.2	B,E
P249	99A3372	Aldrich Alginic Acid	0.5/190	3	7.5	B,E
P251	99A3372	Aldrich Alginic Acid	0.5/190	3	7.5	B,E
P253	99A3372	Aldrich Alginic Acid	0.5/190	3	15	B,E
P255	99A3372	Aldrich Alginic Acid	0.5/190	3	15	B,E
P257	99A3372	Aldrich Alginic Acid	0.5/190	3	30	B,E
P259	99A3372	Aldrich Alginic Acid	0.5/190	3	30	B,E
P261	99A3372	Aldrich Alginic Acid	0.5/190	3	60	B,E
P263	99A3372	Aldrich Alginic Acid	0.5/190	3	60	B,E
P287	99A3372	Aldrich Alginic Acid	0.1/400	4	30	B,E
P289	99A3372	Aldrich Alginic Acid	0.1/400	4	30	B,E

B= ^{239,240}Pu activity, E=DOC

Appendix 2: Procedure used for ^{241}Am and ^{239,240}Pu-activity determinations.

<u>Coprecipitation</u>

1. To 10L of sample add of conc. HNO_3 until pH <2. Let sample sit for 16 hours after acidification before processing.
2. Add 50 ml conc HCl
3. Add tracers (^{243}Am and ^{242}Pu).
4. Add 5 ml ferric chloride carrier (40 mg Fe3+/ml)
5. Mix sample and measure pH with pH paper; if pH >1 add 12N HCl until pH <1. Cover and stir for 30 min.
6. Check pH again. If <1, gently add conc NH4OH; add until turbidity persists than add additional 50 ml.
7. Stir for 30 minutes then let settle overnight..
8. Siphon ~9L of solution; discard
9. Transfer remaining ~1L into 250ml centrifuge tubes, rinsing bucket and transfer beaker (if used) with minimum DI.
10. Centrifuge for 15 min. at 3500 rpm
11. Decant and discard supernate
12. To dissolve precipitate add 3 times ppt volume of conc HCl and mix by vortexing.*
13. Add 75 ml of 9 N HCl
14. Add 2 ml of saturated NaNO2 to samples, mix well and set aside for 15 minutes.

*Note: For samples containing material that did not dissolve in HCL and complete digestion was done as follows:
concentrated nitric, hydrochloric and hydrofluoric acids drynes
concentrated, hydrochloric and nitric (hydrogen peroxide if necessary) dryness
hyrdrochloric dryness. After initial samples showed a problem with tracer equilibration, the remaining samples were treated with concentrated, hydrochloric and nitric dryness followed by hydrochloric acid dryness. As described in

Santschi, P.H., Allison, M.A., Asbill, S., Perlet, A.B., Cappellino, S., Dobbs, C., and McShea, L. 1999. Sediment transport and Hg recovery in Lavaca Bay, as evaluated from radionuclide and Hg distributions. Eviron. Sci. Technol. 33, 378-391.

Pu column

1. Fill disposable plastic column with 7 cm AG1x8 resin (by resin/DI slurry)
2. Place funnel on top of column with Whatman filter paper
3. Wet filter paper with 9N HCl
4. Condition resin with 50 ml 9 N HCl
5. Load sample through filter; rinse 2x with 20 ml 9N HCl (= Am fraction)
6. Rinse 2x with 20 ml 9N HCl, discard
7. Elute Pu with 20ml 9N HCl + 1.5 ml HI
8. Add 1 ml conc HNO_3, evaporate to dryness

Pu Microprecipitation

1. Add 1 ml conc HCl; mix well to resolubilize
2. Add 14 ml DI; mix well
3. Add 1.0 ml lanthanum carrier + 0.5 ml H_20_2 mix well
4. Add 5 ml 3N HF
5. Let sample sit for min. 15-20 minutes
6. Set up filtration apparatus: place 25 mm filter membrane on support screen and lock
7. Apply vacuum; rinse filter with 1-2 ml methanol, rinse filter with DI
8. Transfer sample, rinsing beaker once with 5 ml DI
9. Rinse filter with 10-15 ml DI
10. Turn off vacuum, use "sharpie" and place dot on outside edge of filter marking which side is up. Place filter in pyrex beaker and put in drying oven (90-100˚C) for ~ 1min.
11. Mount filter on stainless steel planchet with double sided adhesive tape.

Am Methanolic Anion Exchange Column

1. Mix anion exchange resin with twice the volume of 1N HNO_3/93% methanol solution (for 160 ml: 10ml conc HNO_3, 150 ml methanol) overnight.
2. Add 5-10 ml conc HNO3;
3. Add 100-125 ml DI;
4. Add 1.0 ml Fe carrier.
5. Transfer to cent. tube and ppt with conc NH4OH. Cent.@3500 rpm for 15 min/
6. Add conc. HNO_3 and evaporate; Repeat
7. Add conc HNO_3 until dissolved. Add methanol (15 ml of methanol for each 1 ml conc HNO_3).
8. Pour resin slurry into disposable column and let resin settle to 7cm, place a layer of silica or glass beads on top of resin
9. Place funnel on top of column with Whatman filter paper, wet filter with 1N HNO_3 /93% methanol solution.
10. Condition column with 40 ml of 1N HNO_3 /93% methanol solution.
11. Load the sample onto column through filter; rinse with 25 ml 1N HNO_3 /93% methanol solution.
12. Remove and discard filter and rinse twice with 25 ml 1N HNO_3 / 93% methanol solution. Discard into methanolic waste receptacle.
13. Strip Am by passing three 20 ml volumes of 8 N HNO_3 through the column allowing each rinse to pass completely before adding next rinse. Collect eluate in beaker for Teva column
14. Evaporate to dryness.

Am Teva Resin

1. Redisolve sample from above in 10 ml 2M NH_4SCN/ 0.1M formic acid (for 100ml 15.2 g NH_4SCN & 0.35 ml 98% formic acid in 100ml DI). Allow sample to sit for 1 hour to ensure dissolution.
2. Condition a TEVA resin 2 ml column with 5 ml of 2M NH_4SCN/ 0.1M formic acid solution
3. Transfer sample into TEVA column in two portions using disposable polyethylene transfer pipet. Rinse the sample container with 1 ml of 2M NH_4SCN/ 0.1M formic acid and transfer to column. Repeat rinse and add to column.
4. Rinse the TEVA column with two 5 ml volumes of 1M NH_4SCN/ 0.1M formic acid (for 100 ml dissolve 7.6 g NH_4SCN and 0.35 ml 98% formic acid in 100 ml of DI). Allow first wash to pass completely before adding the second wash. (This washes lanthanides from column).
5. Strip Am from column with 15 ml of 2N HCl in three 5ml portions allowing each 5 ml to pass completely..
6. To decompose thiocyanate, add 2.5 ml conc HNO3 and 7.5 ml conc. HCL to the Am solution. Swirl gently. Evaporate until ~1 drop solution is remaining.
7. Add 5 ml conc HNO3. Evaporate until volume is ~1 drop.

Am Micro-precipitation (SOP 780)

1. Add 1 ml conc HCl to sample. Heat for 5 minutes. Add 15 ml DI.
2. Add 0.5 ml lanthanum carrier. mix well. Add 5 ml HF. Mix well.
3. Allow sample to stand for 15-20 minutes minimum.
4. Place 25 mm filter membrane in a filter funnel assembly and turn on vacuum. Rinse with 1-2 ml alcohol.
5. Load sample into filter. Rinse sample beaker once with 5 ml DI and add to funnel.
6. After sample has passed through filter. Rinse filter with 10-15 ml DI
7. Turn off vacuum, use “sharpie” and place dot on outside edge of filter marking which side is up. Place filter in pyrex beaker and put in drying oven (90-100˚C) for ~ 1min.
8. Mount filter on stainless steel planchet with double sided adhesive tape.

Appendix 3 Humic Acid Extraction Procedure

1. RFETS soil (RIN# 00R0972) was analyzed in duplicate for ^{239,240}Pu activity. The average activity for the two measurements was 0.150pCi/g.
2. In duplicate, 10g of dried sediment was added to 100 ml of 0.1M NaOH in a 250ml centrifuge tube and allowed to sit overnight.
3. The extracts were centrifuged at 2000rpm for 15 min. The supernatant was collected in a separate container. The sediment was resuspended in 10ml of 0.1M NaOH and centrifuged at 2000 rpm for 15 min. This was repeated two more times and all supernatants combined.
4. To precipitate the humic acids, the supernatant was acidified with 6N HCl to pH=1 and centrifuged at 2000rpm for 15 min.
5. The supernatant was collected as the fulvic acid fraction and the precipitate was washed three times with 0.1N HCl.
6. The two humic acid fractions were combined and dried. An aliquot was treated with HCl , allowed to dry and analyzed for organic Carbon.

Adopted from:
Schnitzer, M. & Khan. S.U. 1972. Humic Substances in the Envrionment. Marcel Dekkar, New York, 327p.

Appendix 4 Transmission Electron Microscopy (TEM) and Energy dispersive X-Ray Microprobe Analysis (EDAX)

The samples were infiltrated and embedded in Nanoplast FB101 melamine resin, then sectioned. The ultra-thin sections (60-80 nm) were made on a Reichert Ultracut E microtome, then collected on uncoated 200-mesh Cu grids for morphological characterization and on Formvar, Carbon-coated 100-mesh Ni grids for EDS analysis. The sections were viewed in a JEOL JEM 1200 EX TEMSCAN microscope operating at 80kV. The range of primary magnifications used for morphological analyses was 10,000-75,000 X. The microscope is equipped with a Tracor Northern EDS detector (laboratory resolution of 150.7 eV), connected to a computer that runs Iridium System (Version 2.2) software (IXRF Systems, Inc.). The duration for each point of analysis was 100s with parameters such as kV, tilt, spot size and working distance kept the same for all analyses. The spectra obtained are keyed to individual particles on micrographs some of which are shown in the following figures.

The freeze-drying method that was employed can clump the colloids severely, making fine distinctions between the two samples difficult to ascertain. For morphological analyses, in the future we consider preparing the samples in Nanoplast on site with no sample storage whatsoever, possibly as a part of a multi-method protocol employing some conventional methods for TEM sample preparation. While Nanoplast preserves the three-dimensional aspect of colloid aggregates, it does not provide good contrast for cells or organic materials in general. A second protocol used correlatively, could correct for this problem; such a protocol will be applied in future work.

Since this procedure is semi-quantitative there are no calibration procedures.

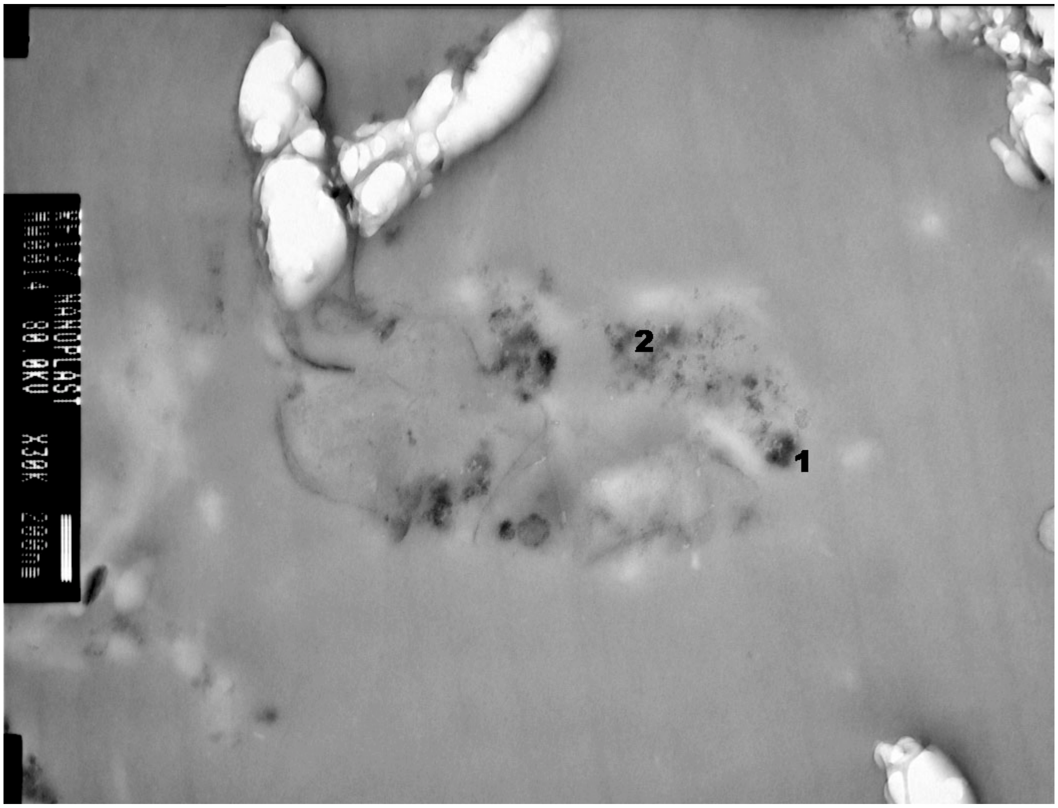


Figure A4a. TEM pictures of colloid samples. RF137N-1

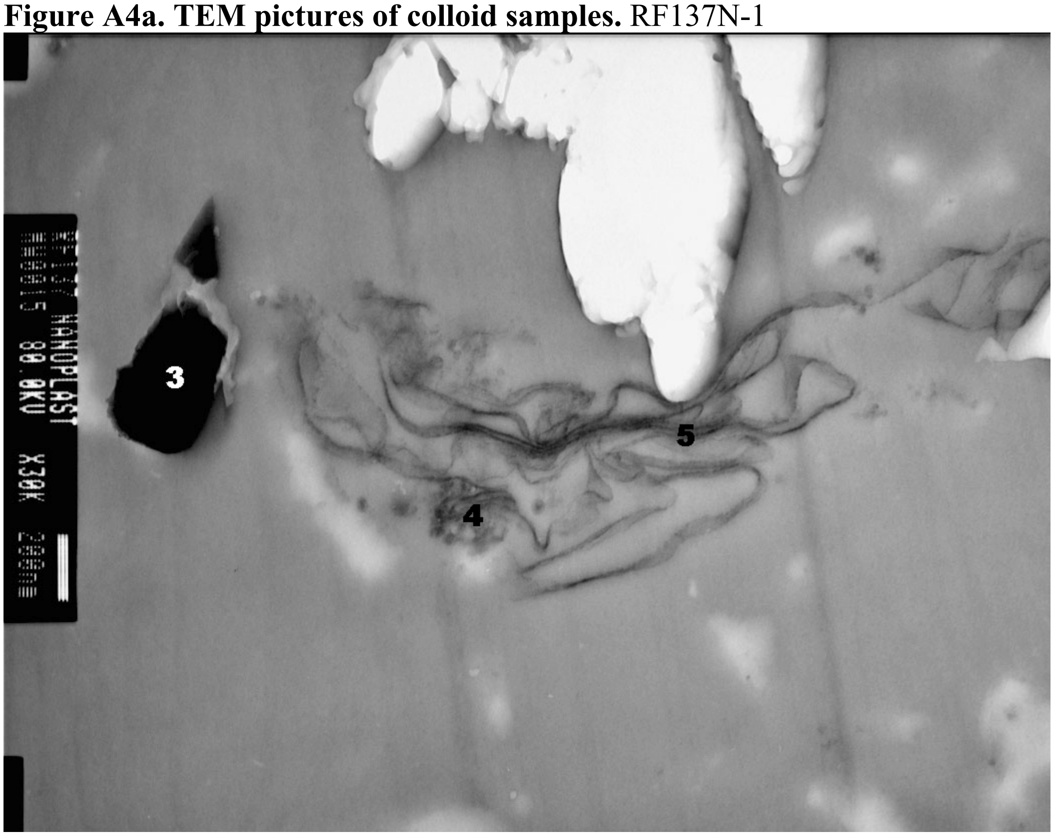


Figure A4b. TEM pictures of colloid samples. RF137N-2

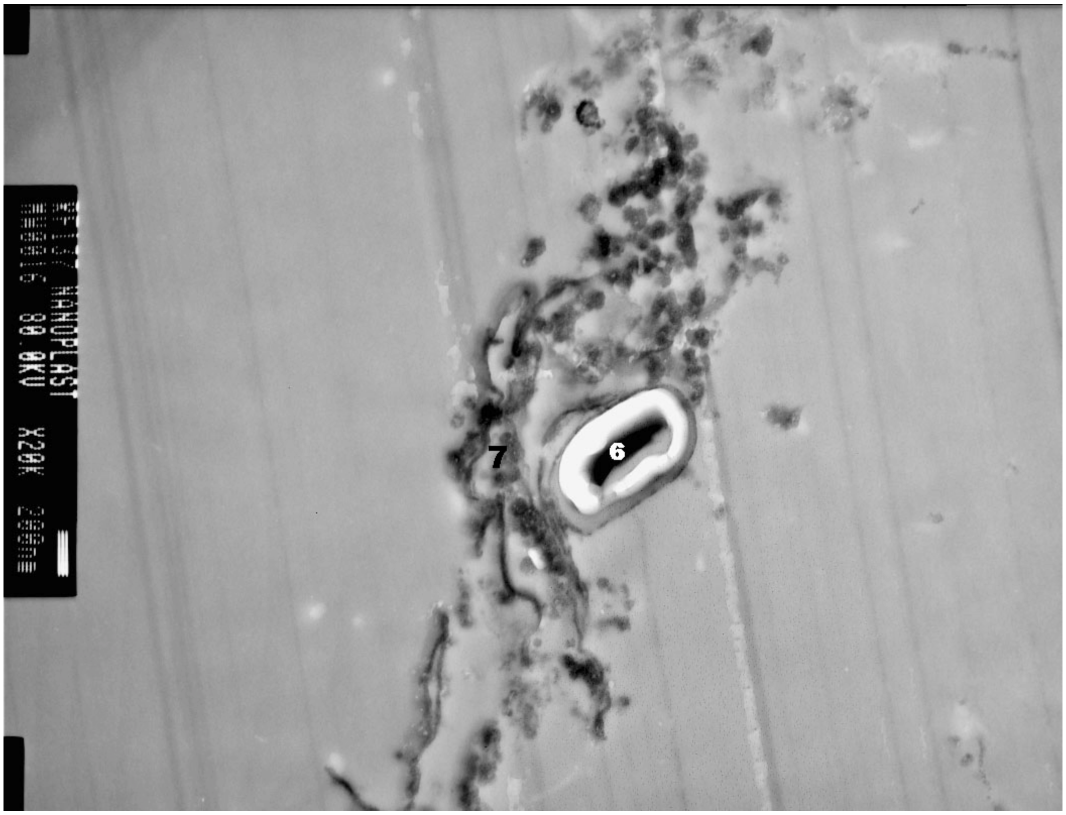


Figure A4c. TEM pictures of colloid samples. RF137N-3

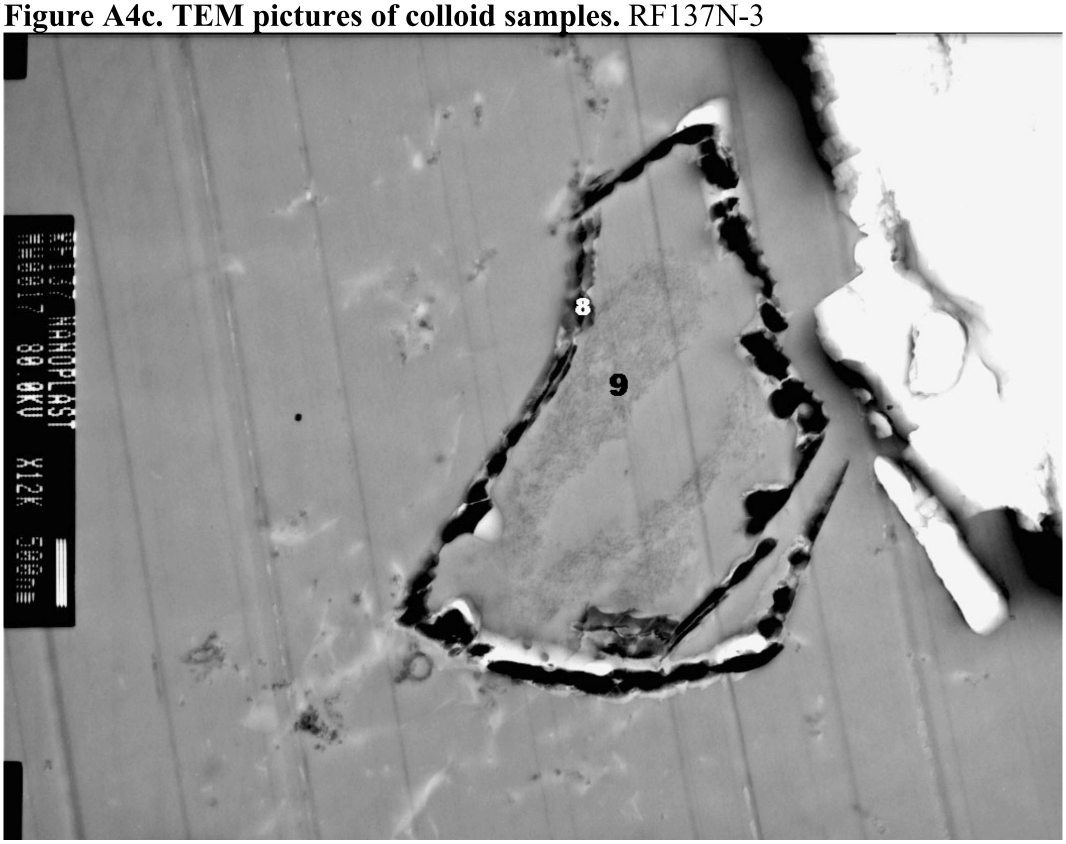


Figure A4d. TEM pictures of colloid samples. RF137N-4

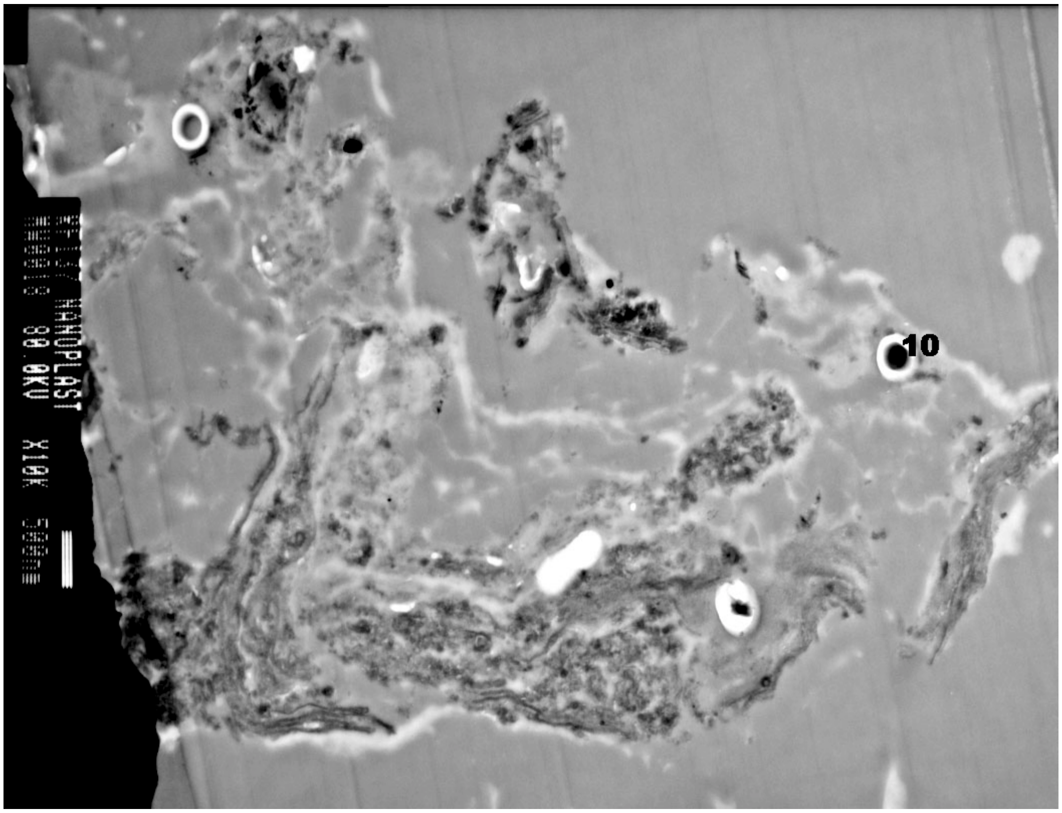


Figure A4e. TEM pictures of colloid samples. RF137N-5

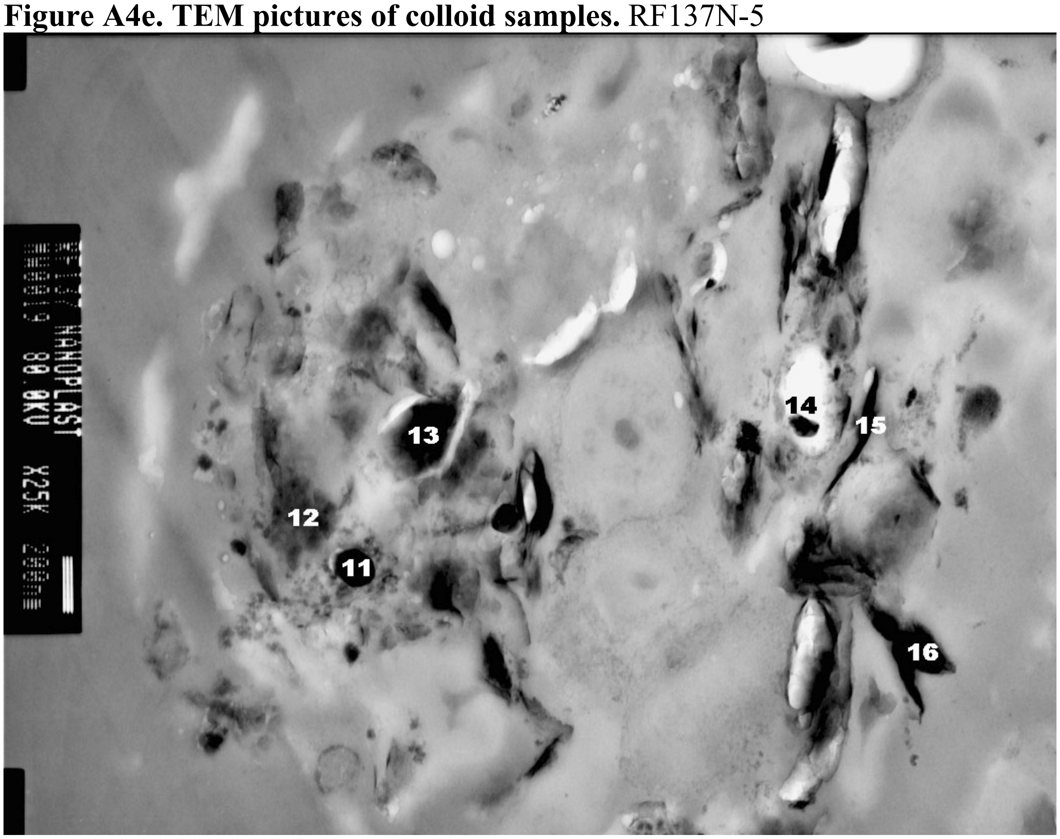


Figure A4f. TEM pictures of colloid samples. RF137N-6

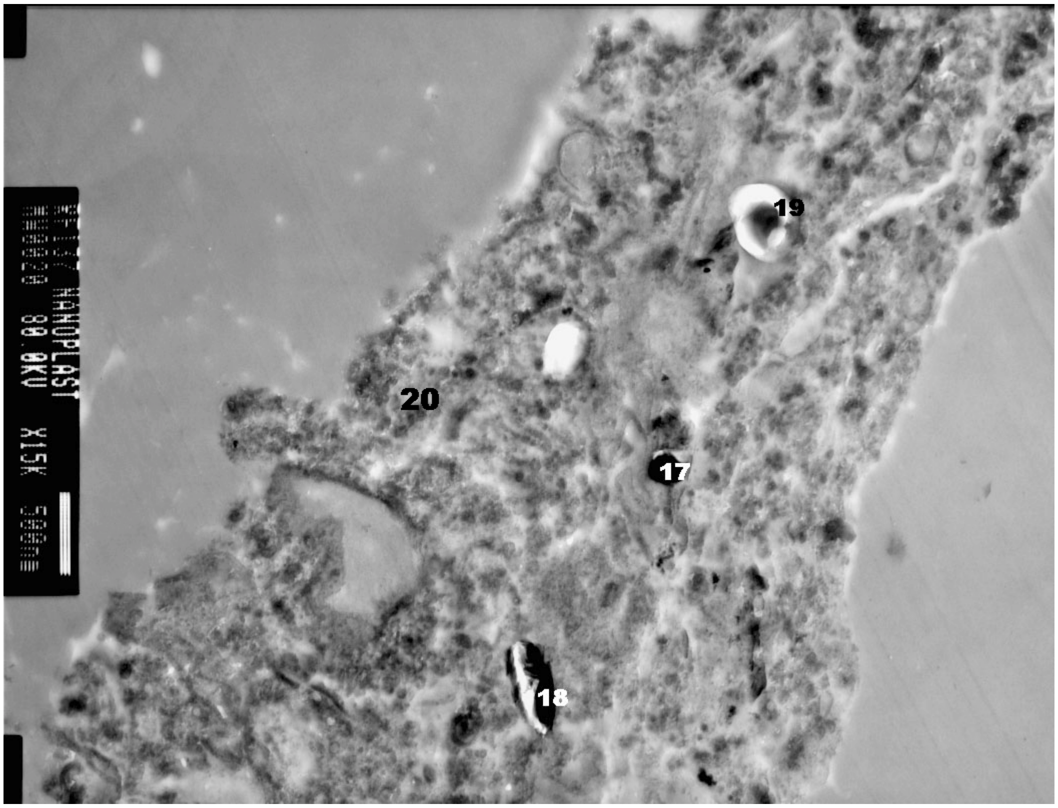


Figure A4g. TEM pictures of colloid samples. RF137N-7

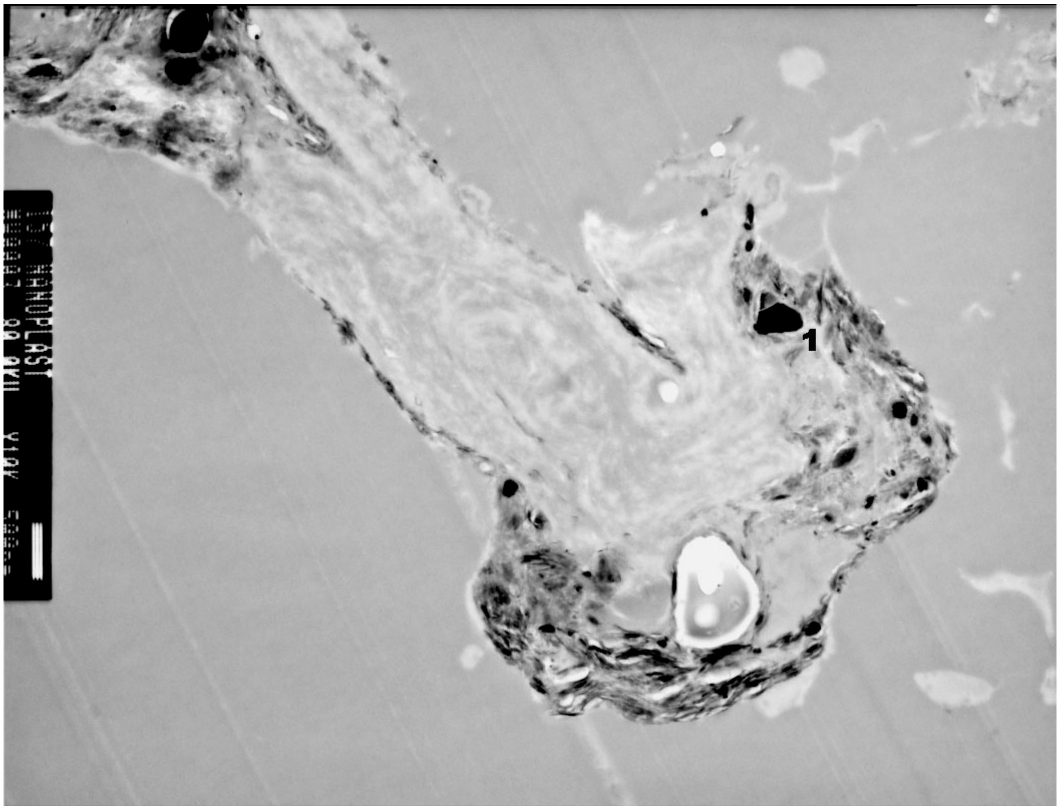


Figure A4h. TEM pictures of colloid samples. RF157N-1

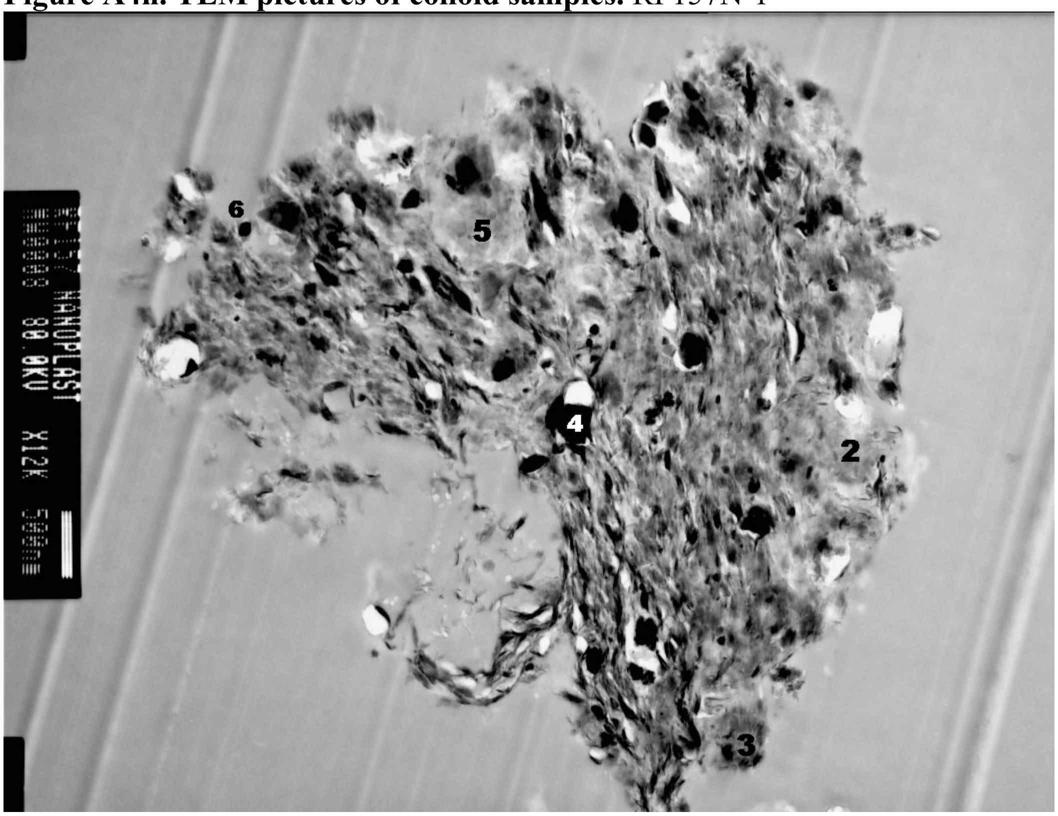


Figure A4i. TEM pictures of colloid samples. RF157N-2

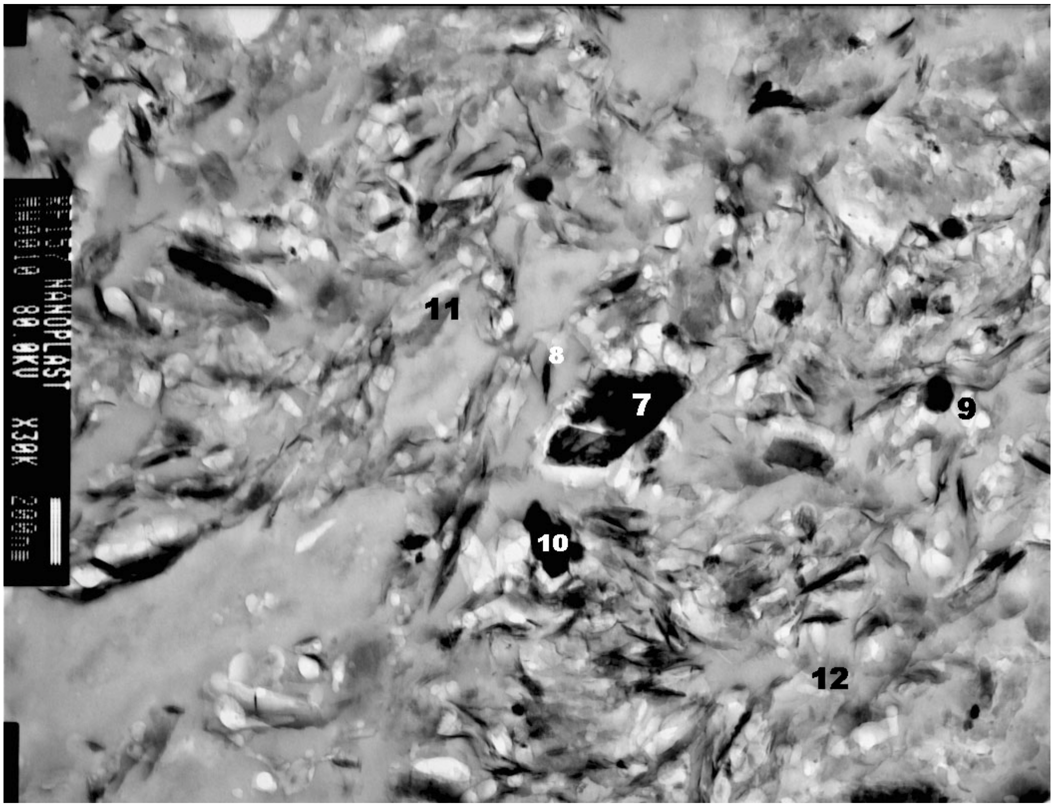


Figure A4j. TEM pictures of colloid samples. RF157N-3

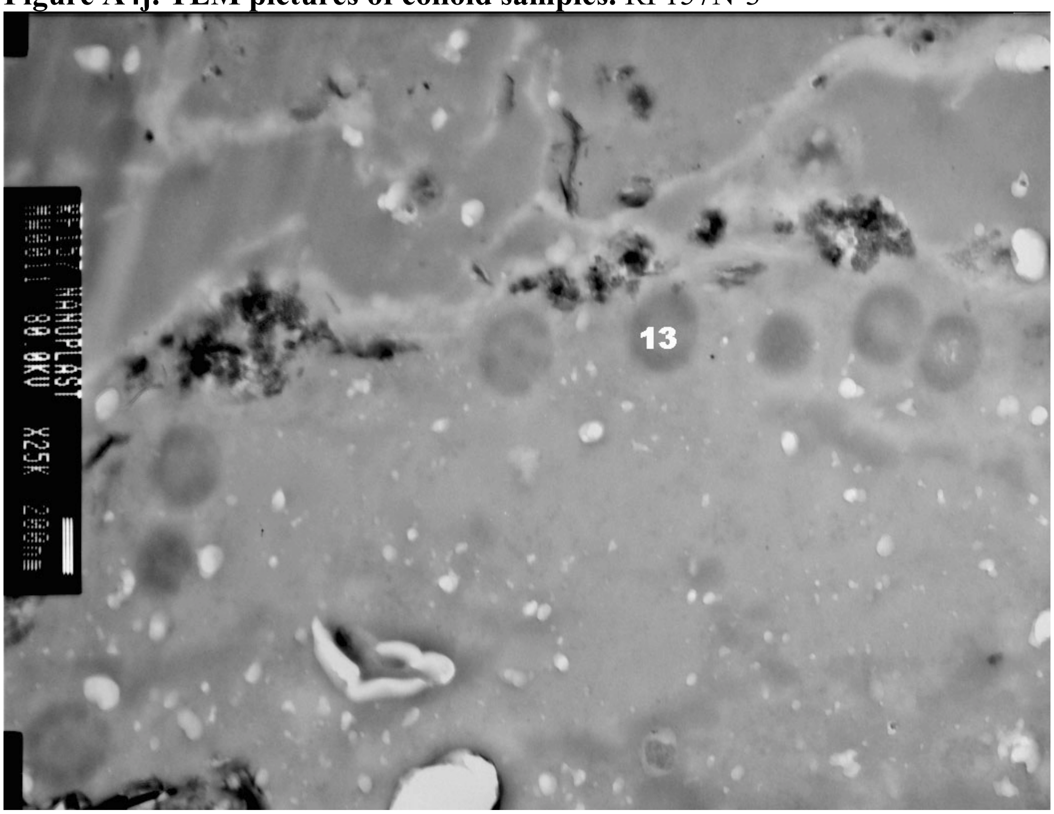


Figure A4k. TEM pictures of colloid samples. RF157N-4

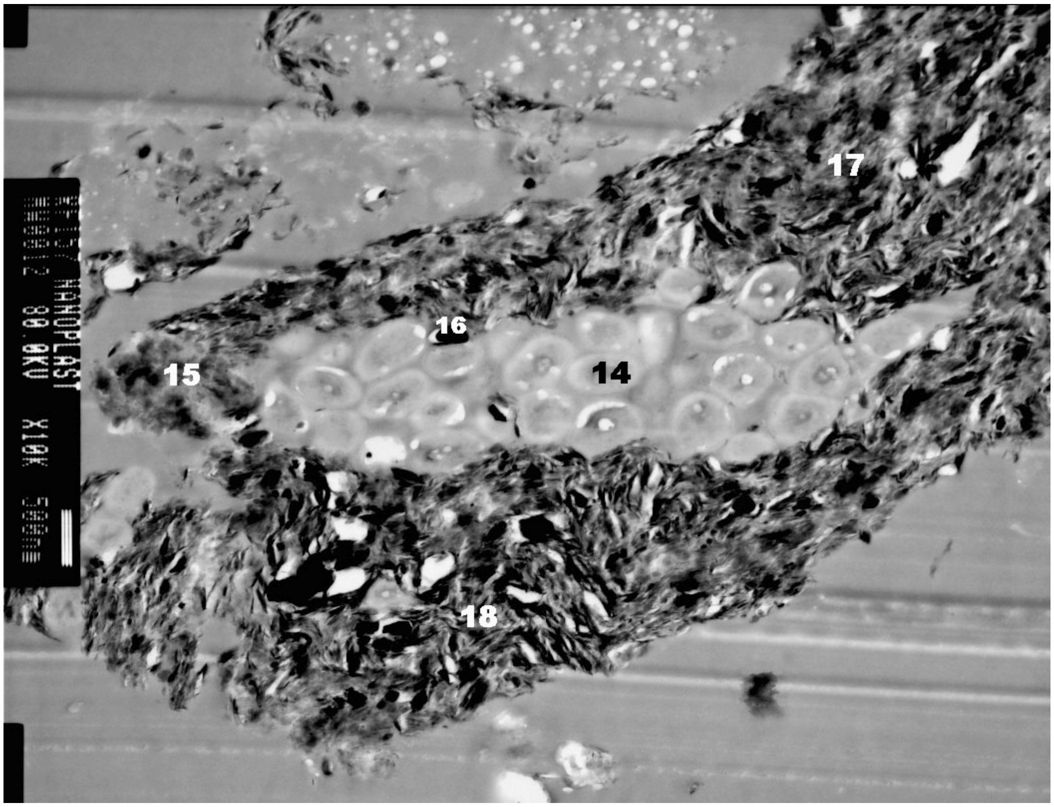


Figure A4l. TEM pictures of colloid samples. RF157N-5

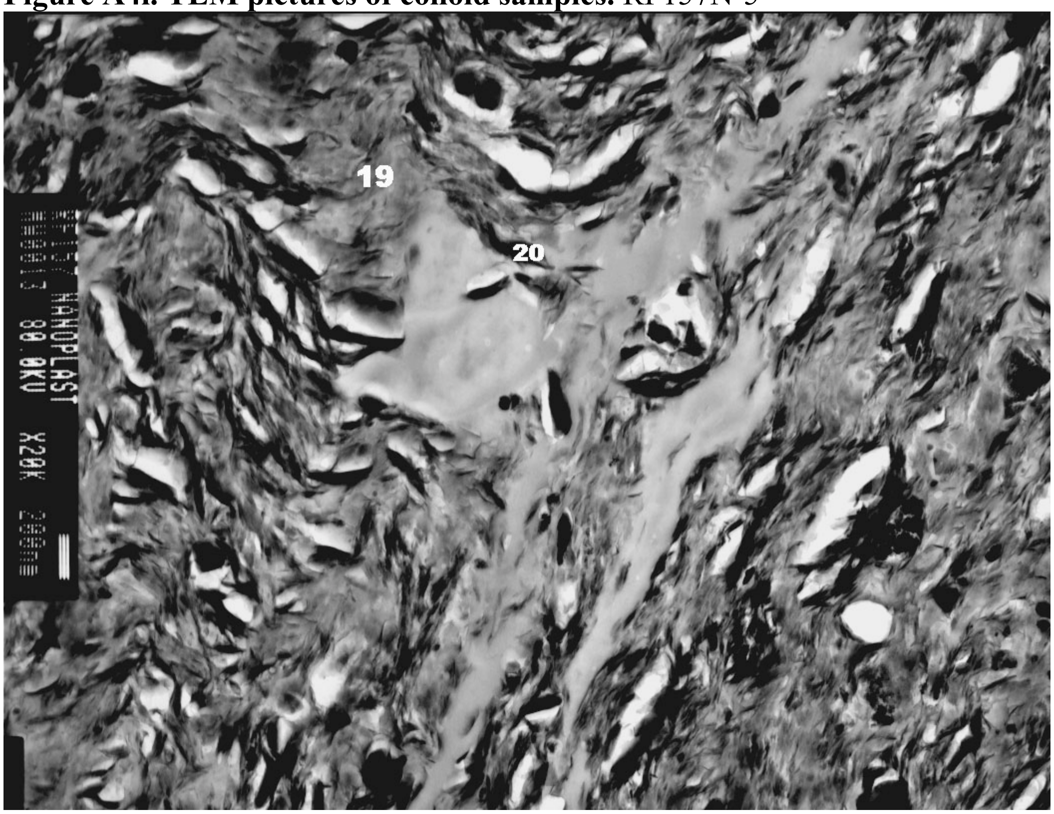
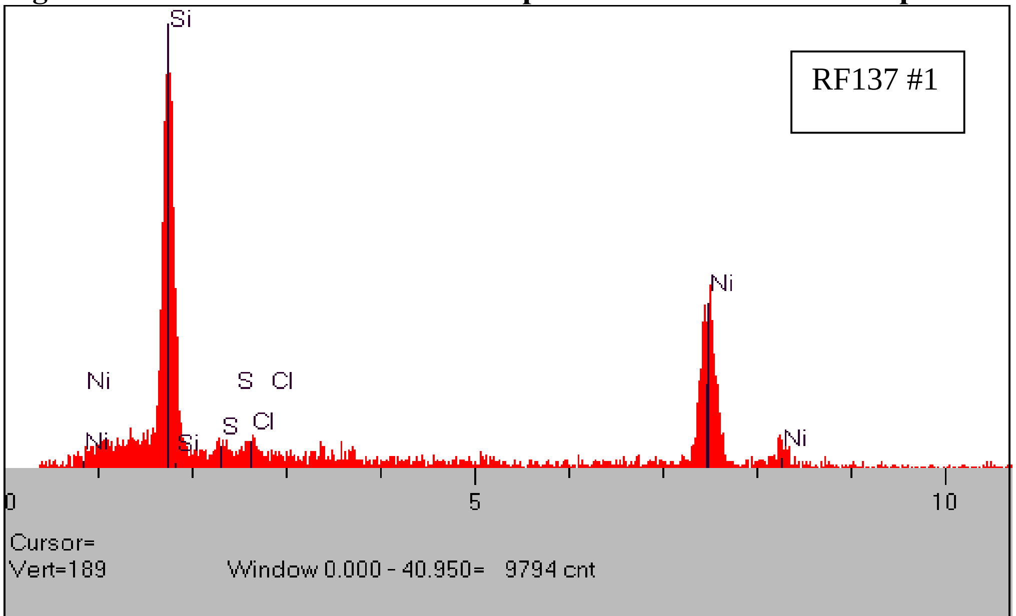
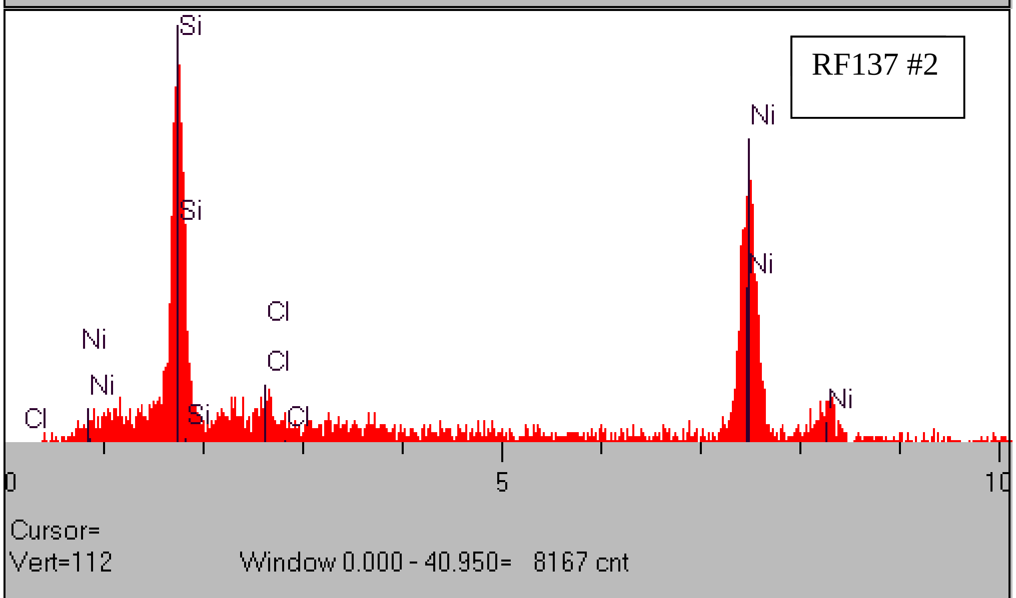
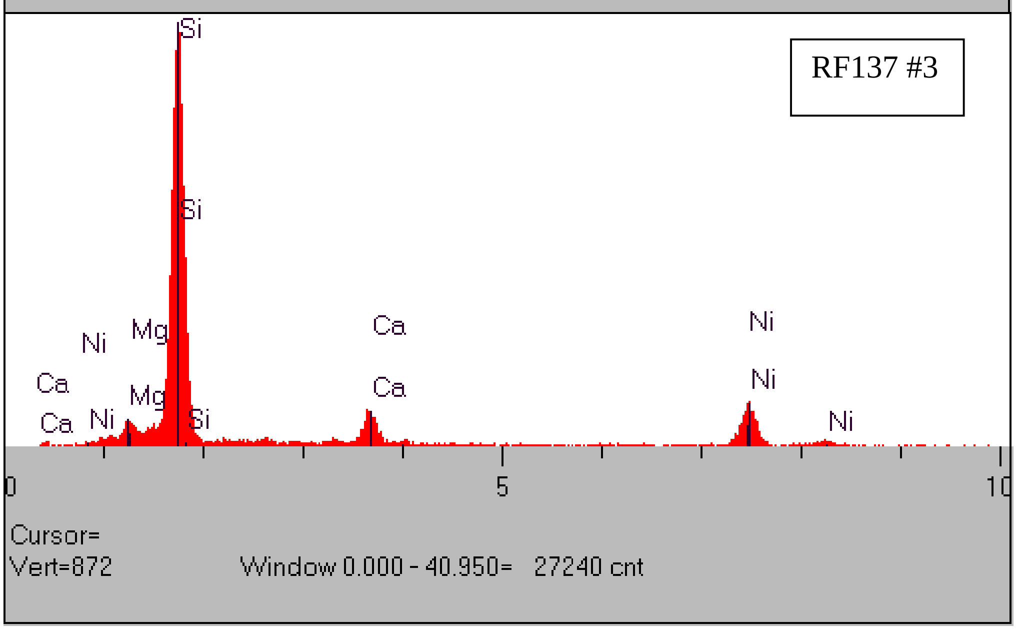


Figure A4m. TEM pictures of colloid samples. RF157N-6

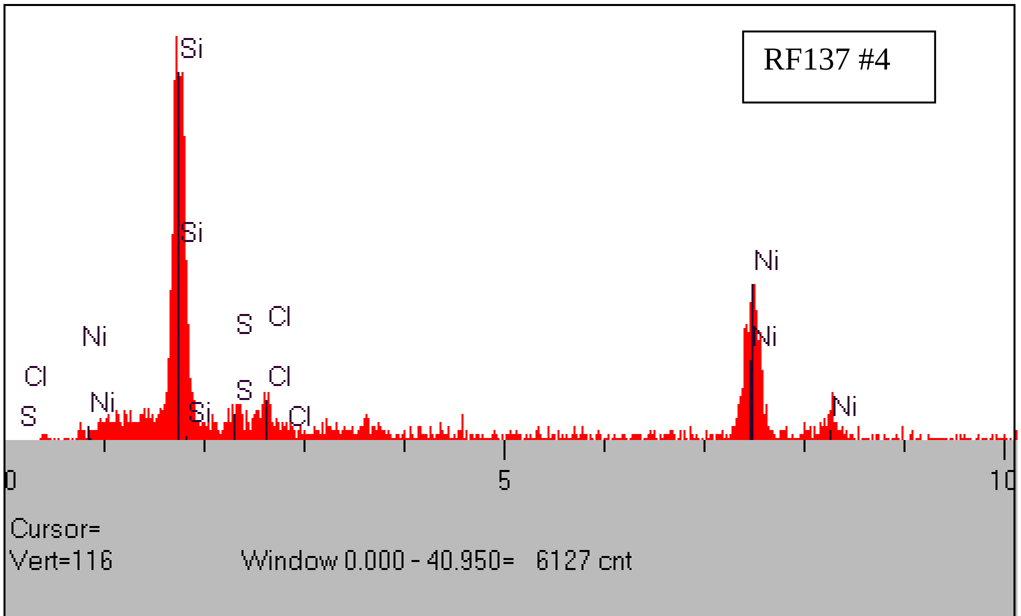
Figure A4n.1-20. EDAX of individual particles in the colloids samples: RF137Nanoplast

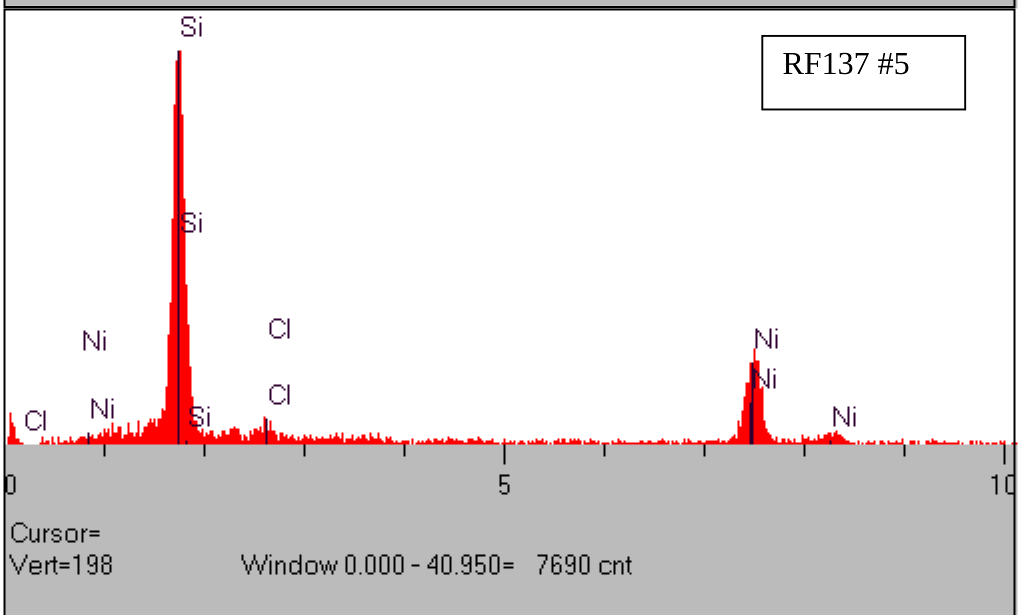


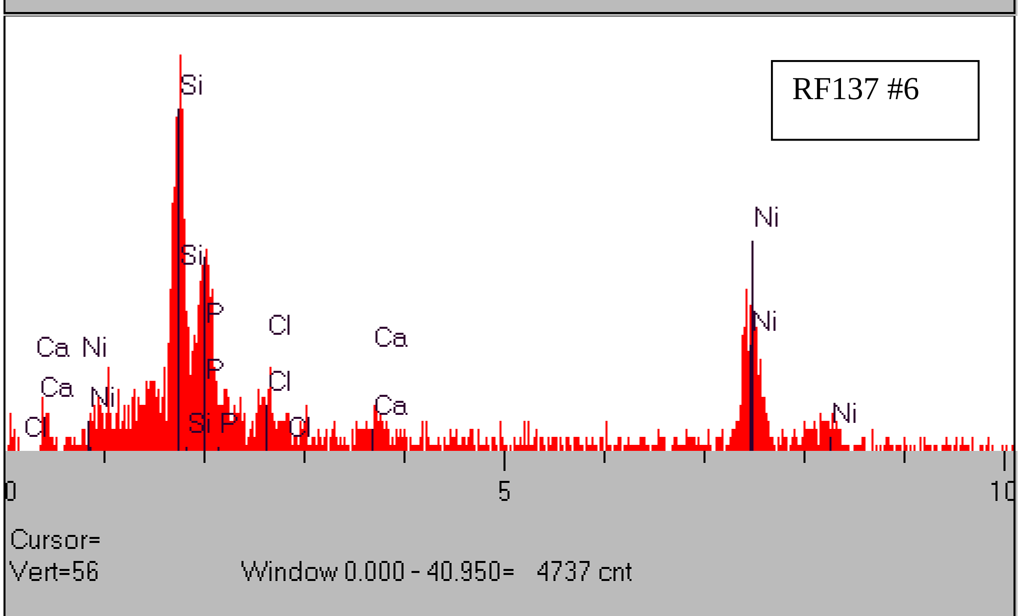




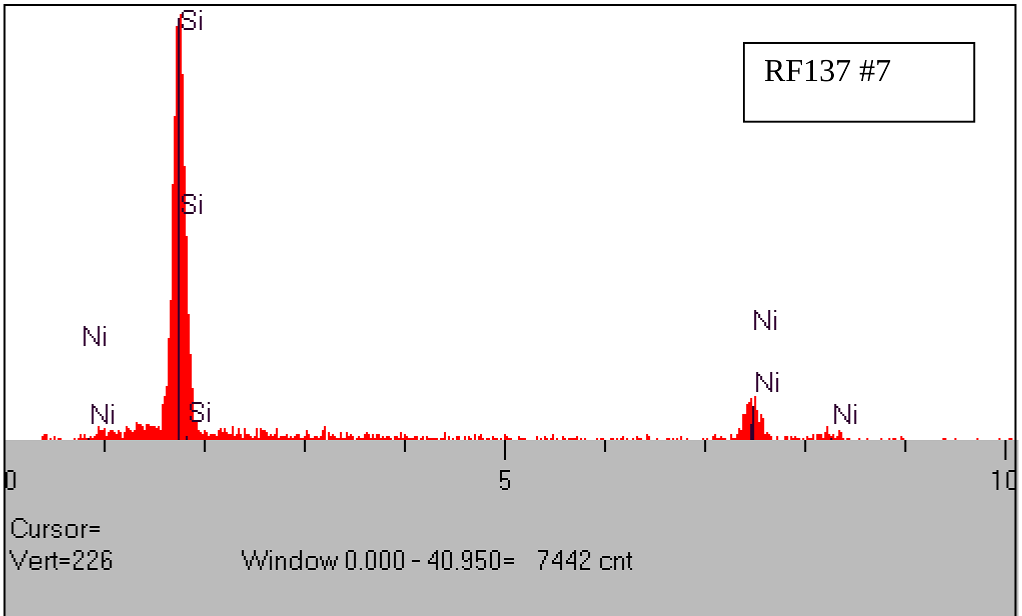
RF137Nanoplast

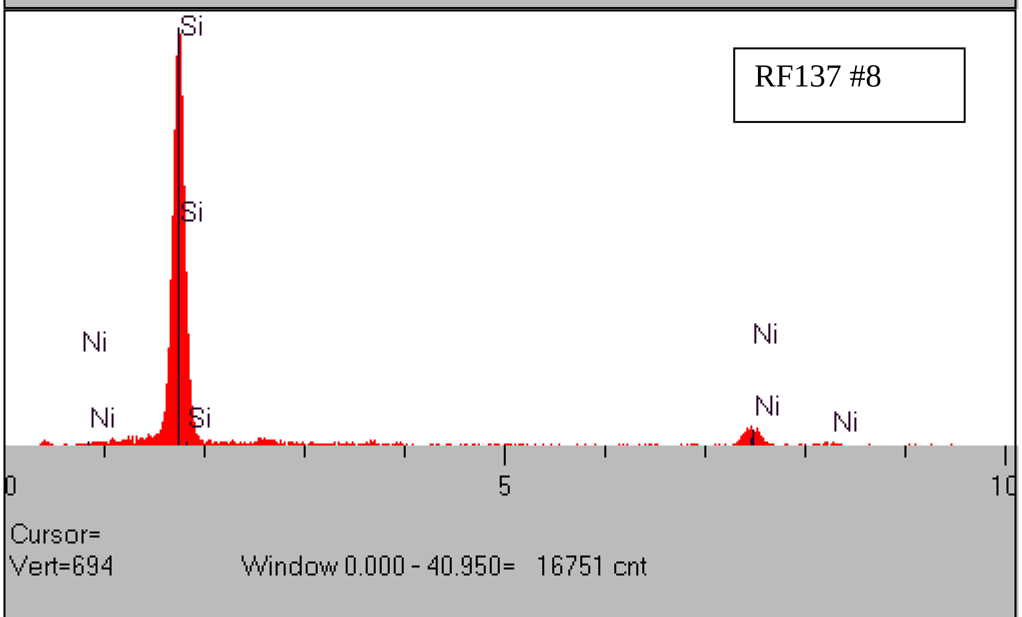


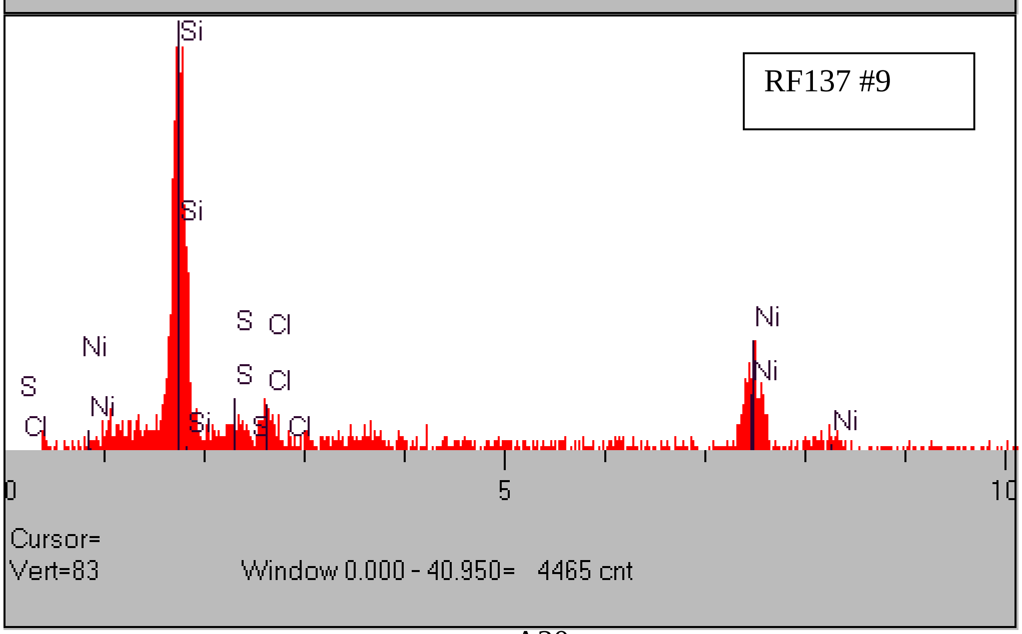


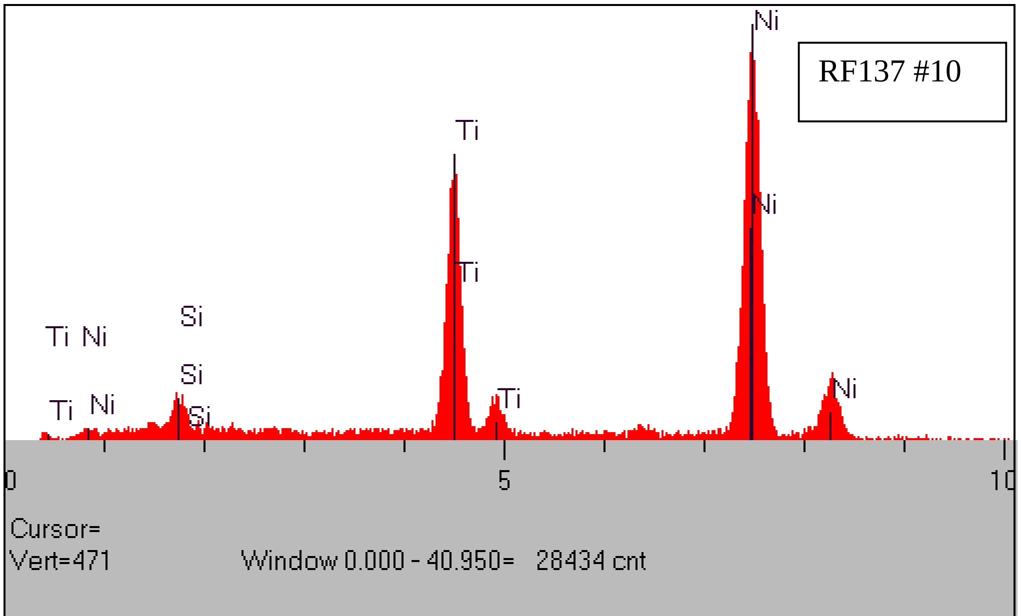


RF137Nanoplast





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Si
Ti Ni
Ti Ni
0
5
10
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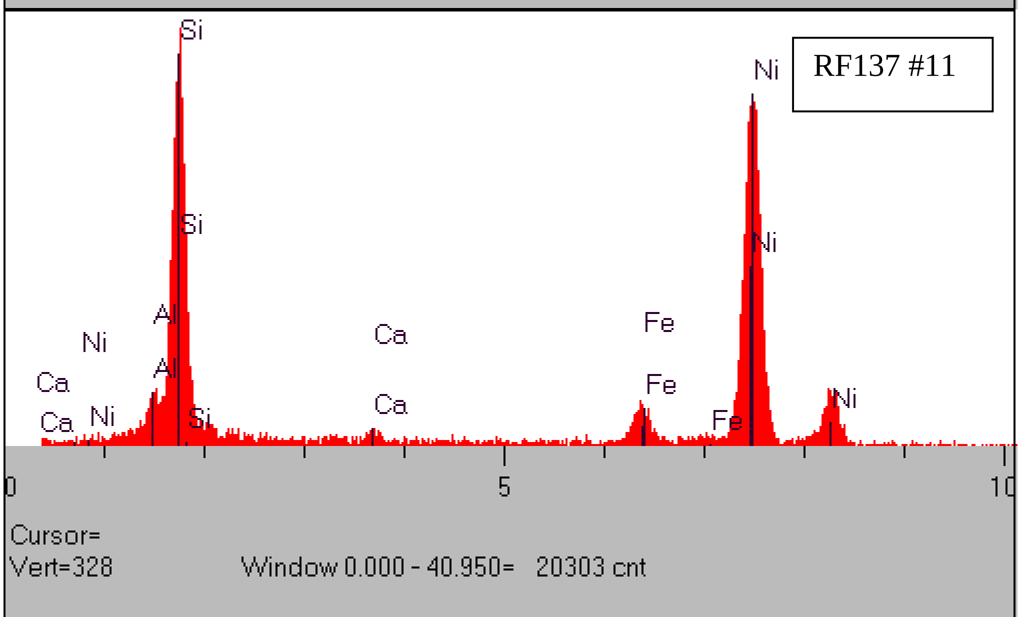
Vert=471
Window 0.000 - 40.950= 28434 cnt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Al
Ca
Fe
Ca Ni
0
5
10
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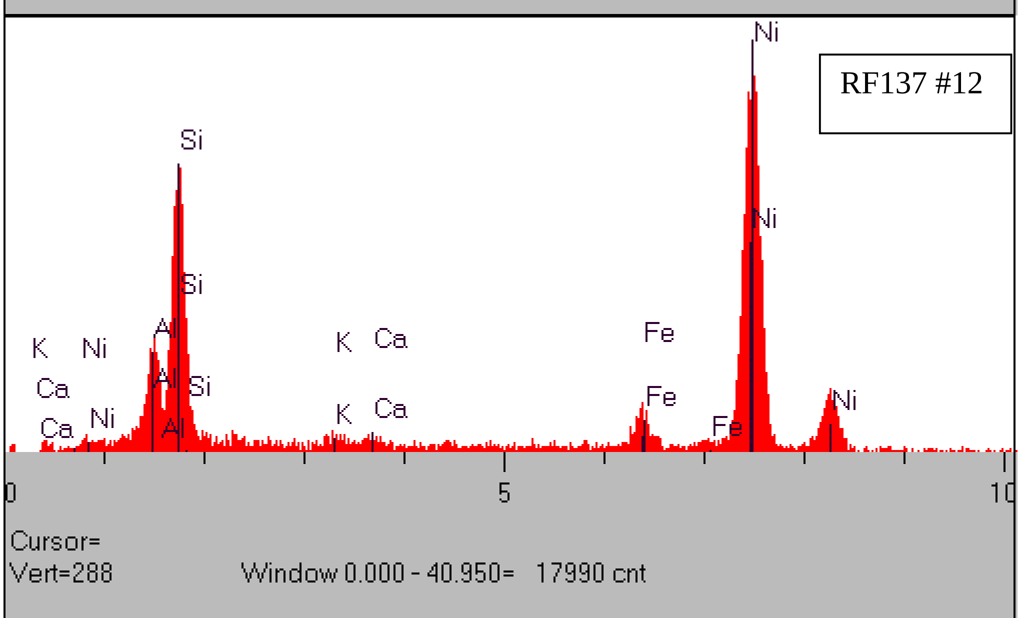
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Al
K
Ca
Fe
K Ni
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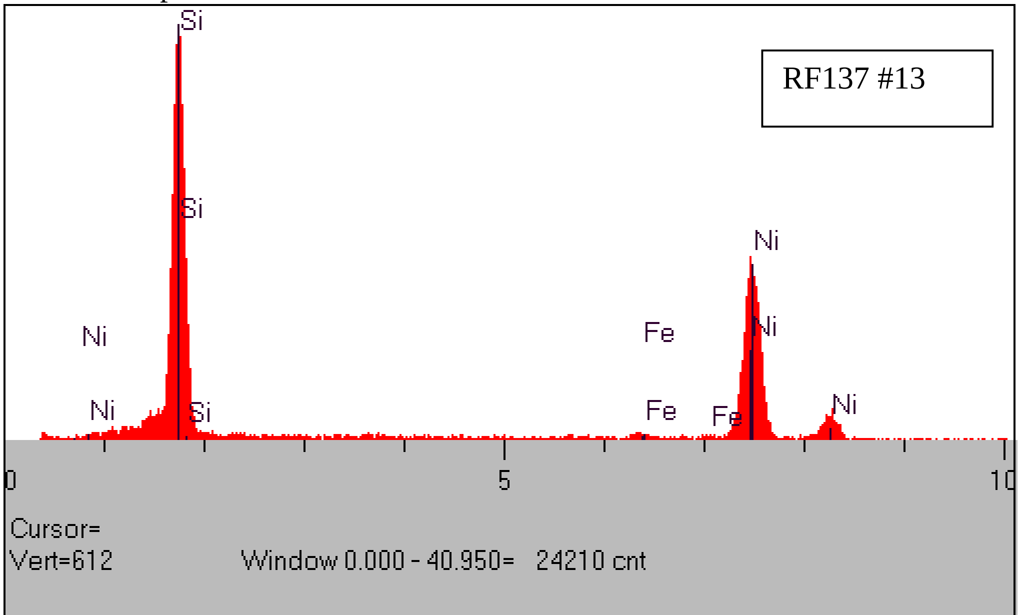
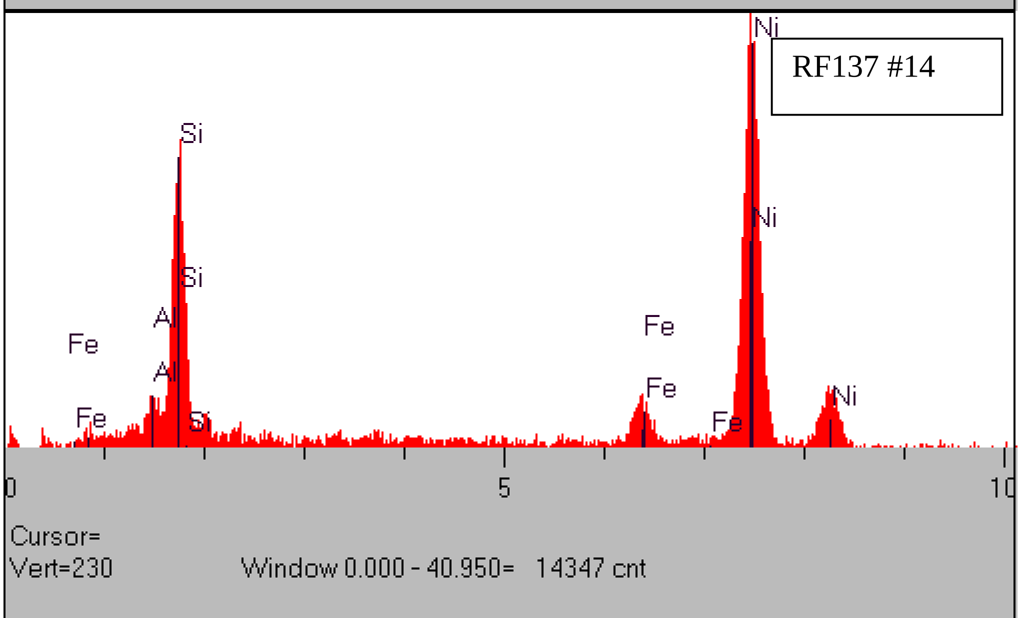
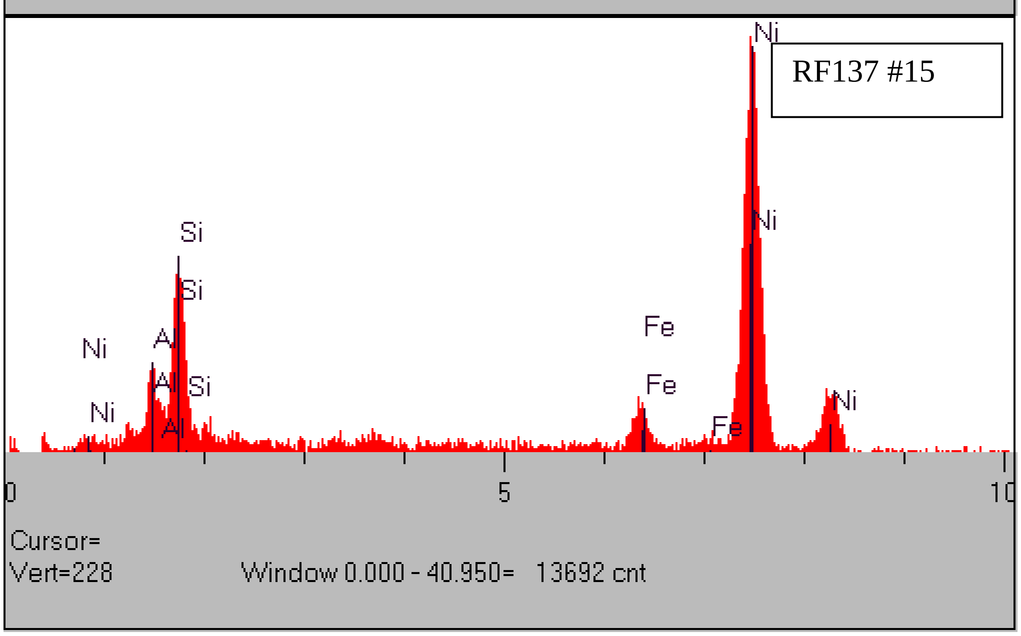
RF137Nanoplast







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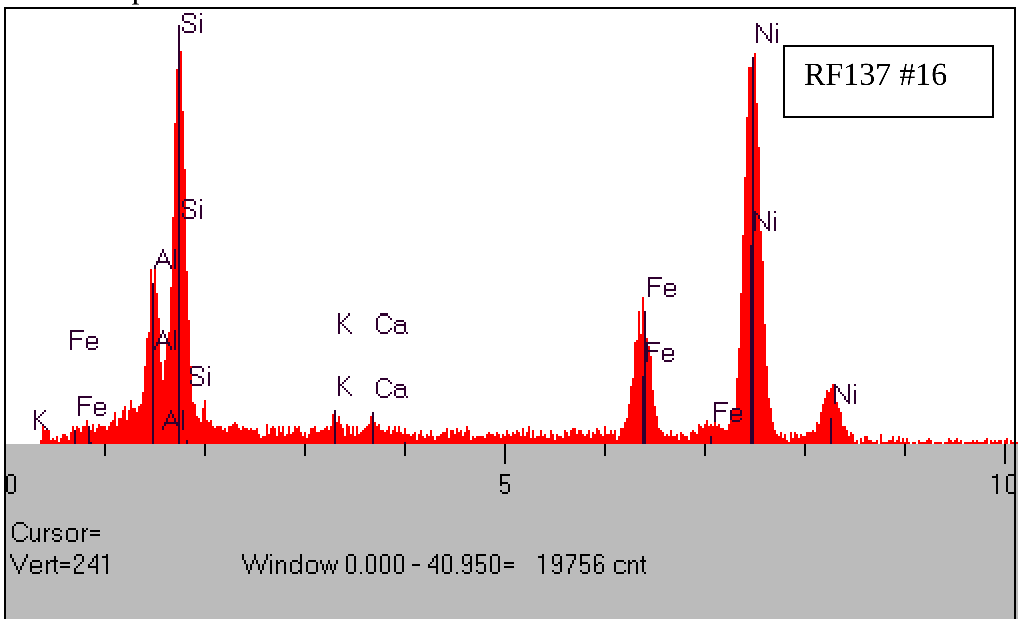

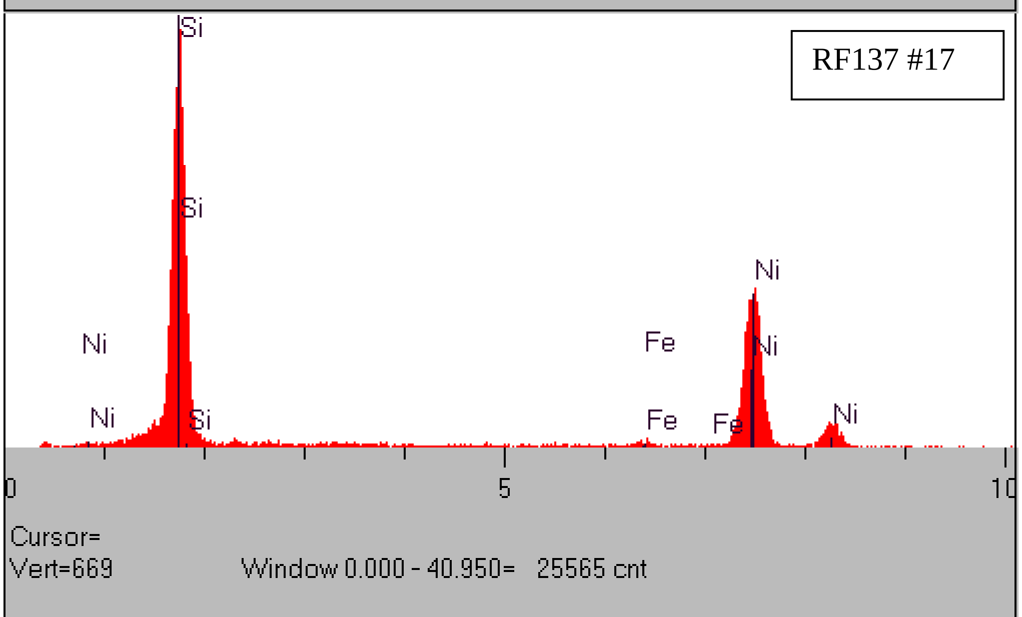

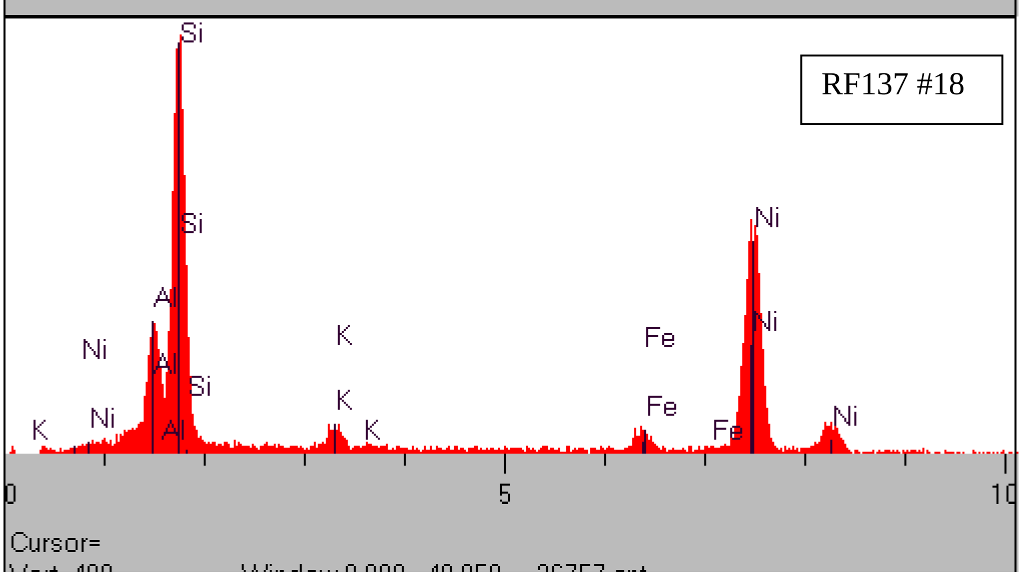

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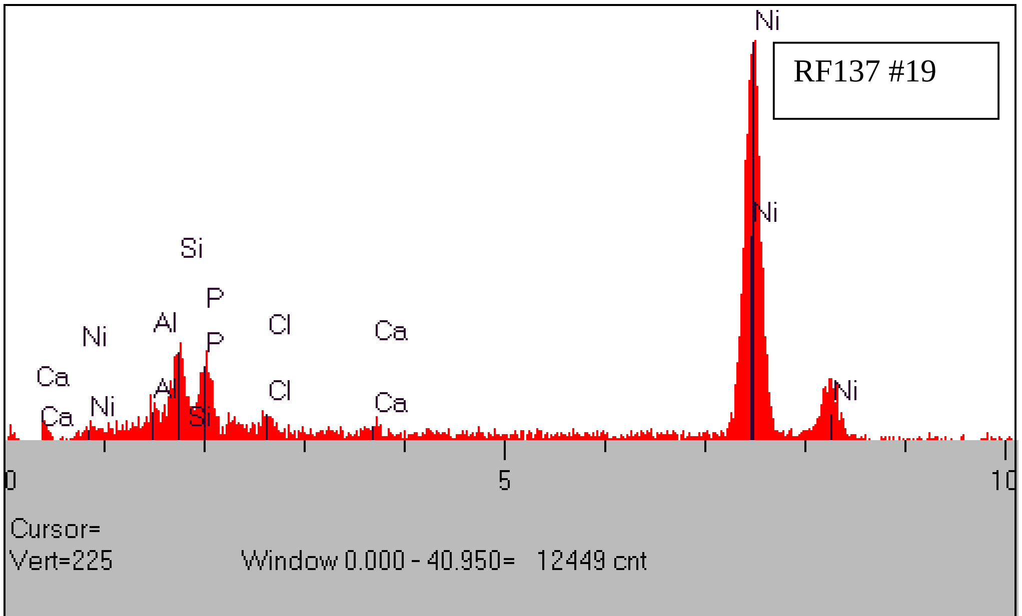


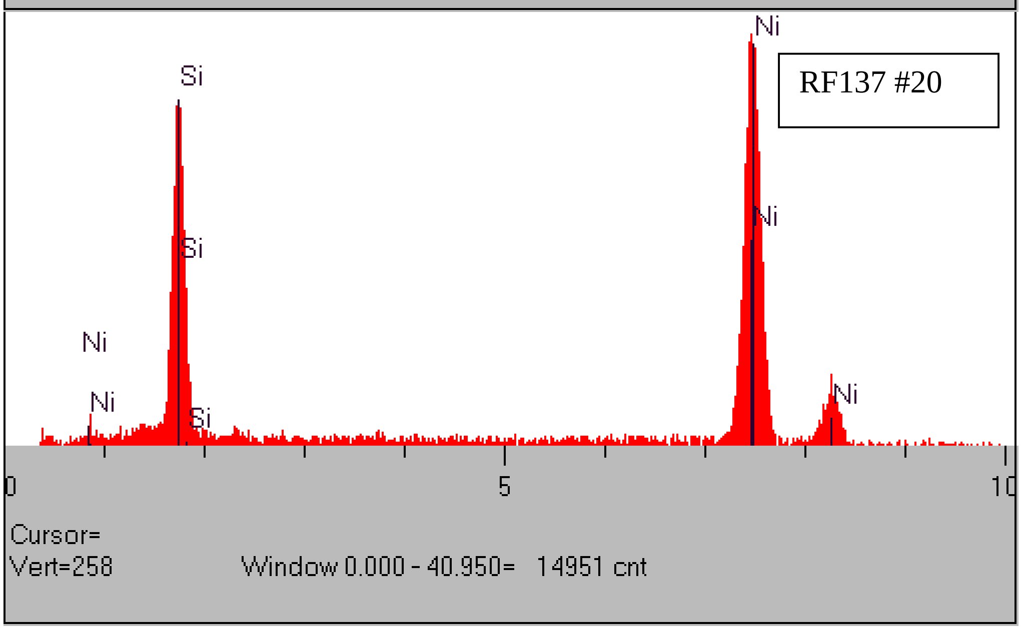
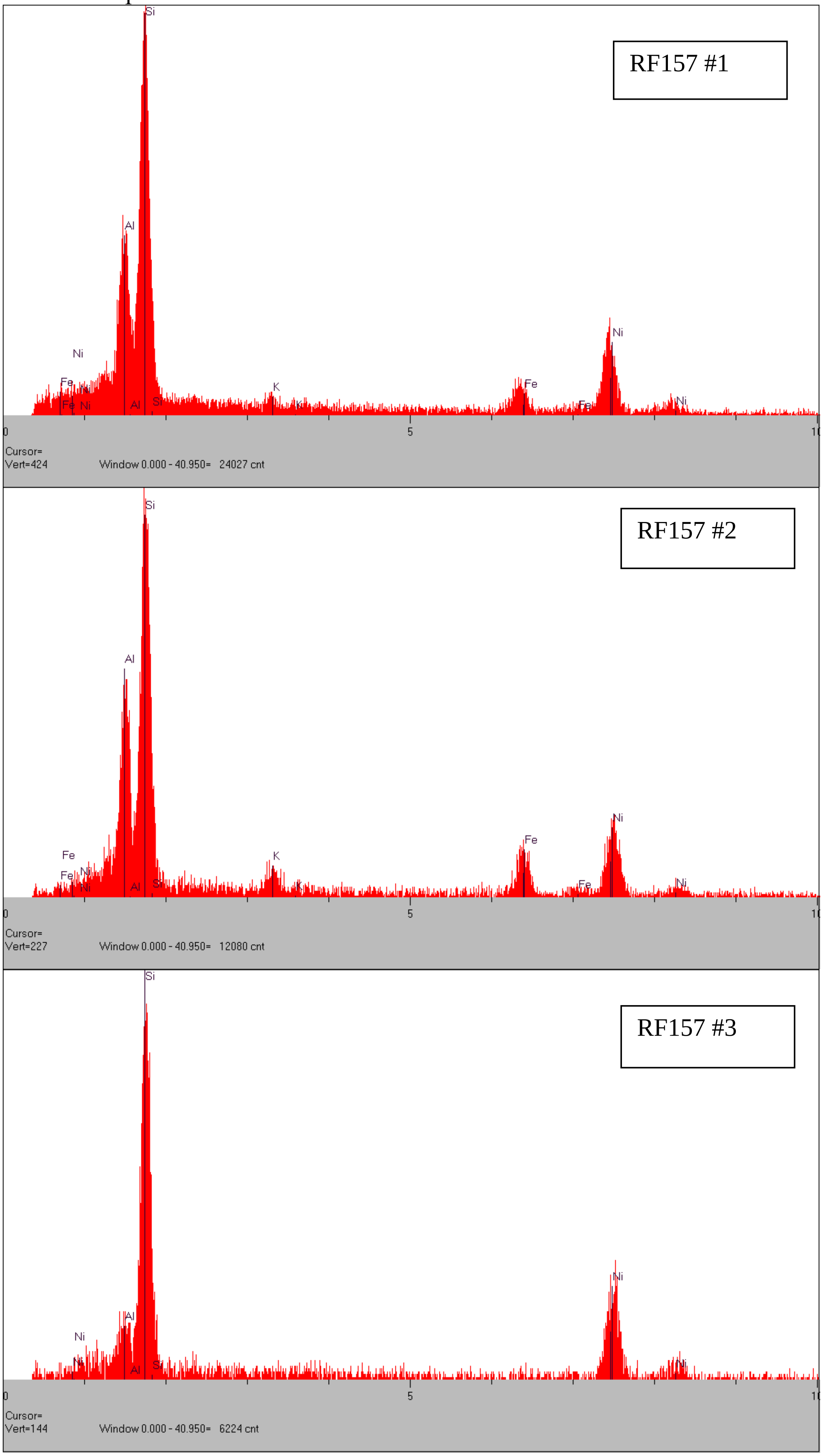


Figure A4o.1-20 EDAX of individual particles in the colloids samples:
RF157Nanoplast



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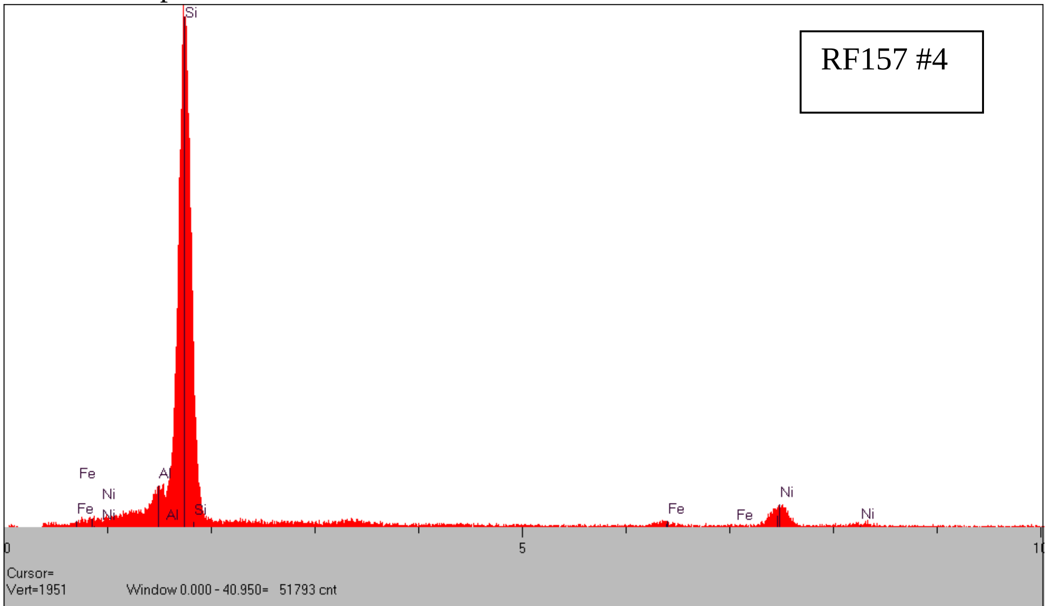
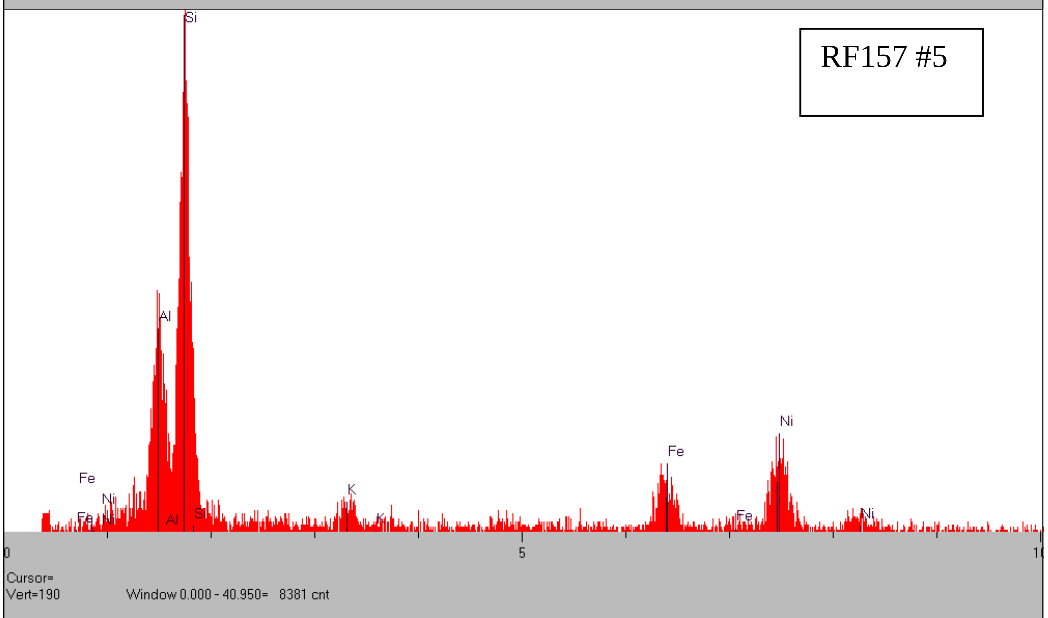
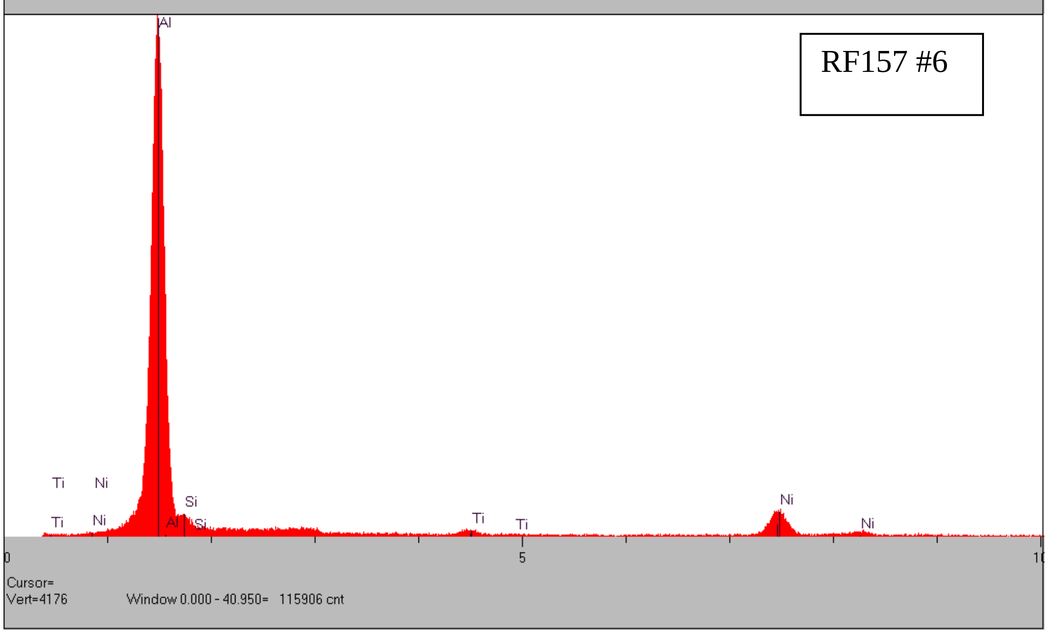
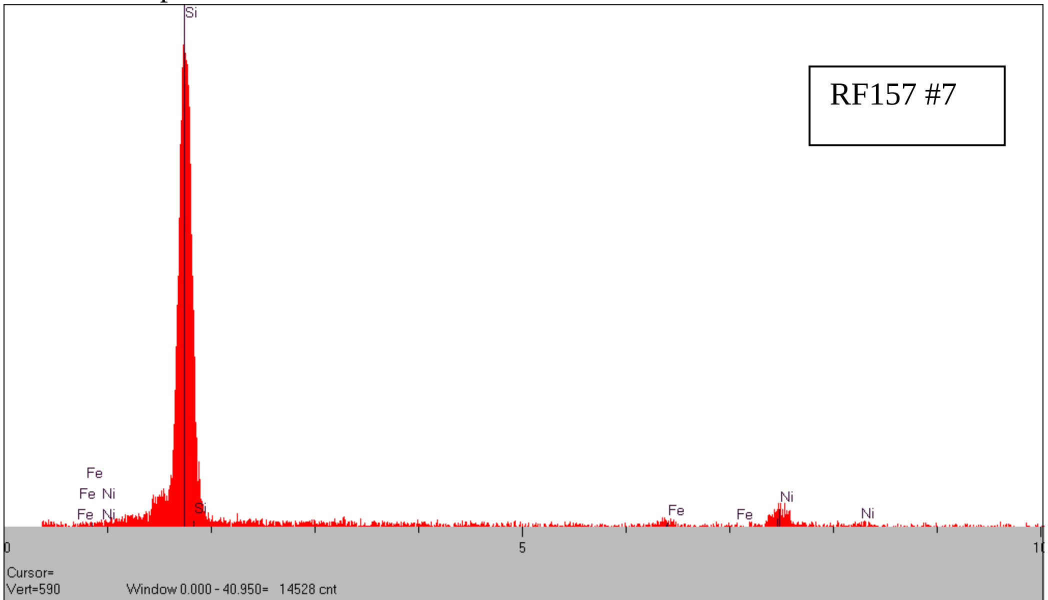
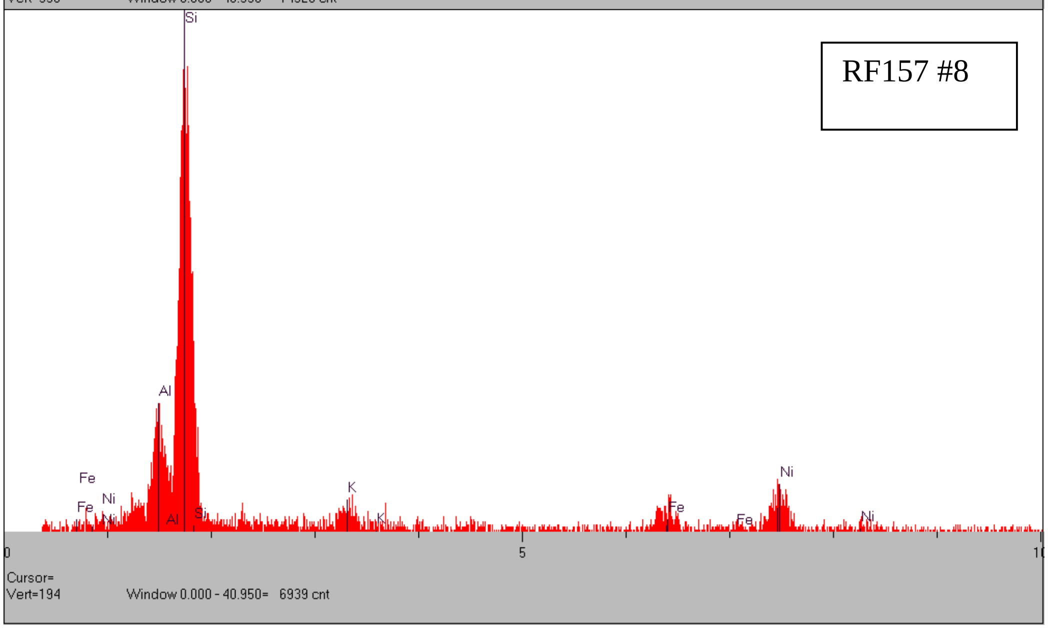


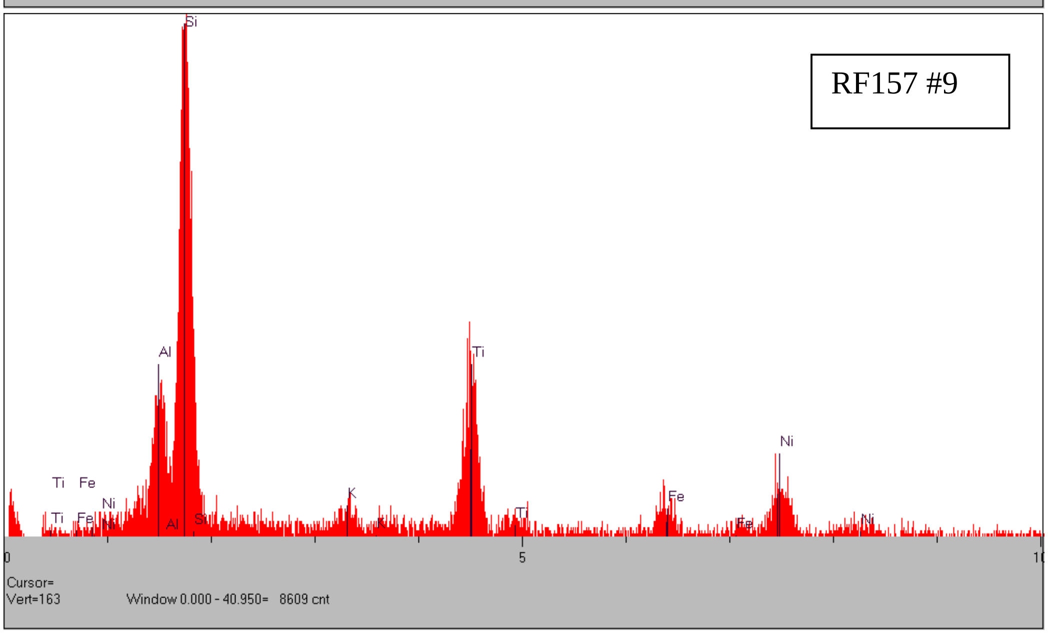




RF157Nanoplast







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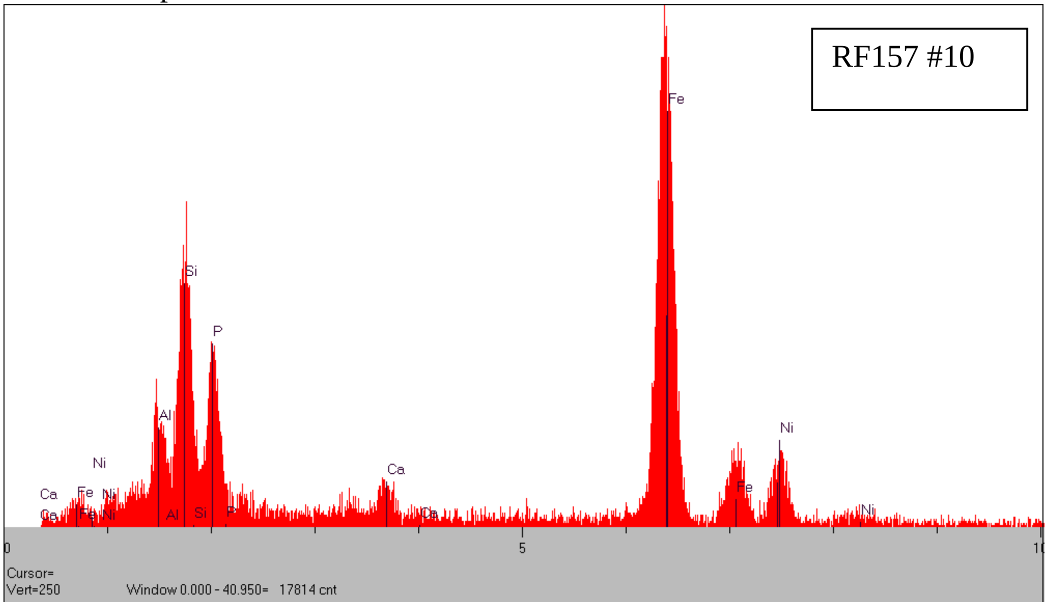

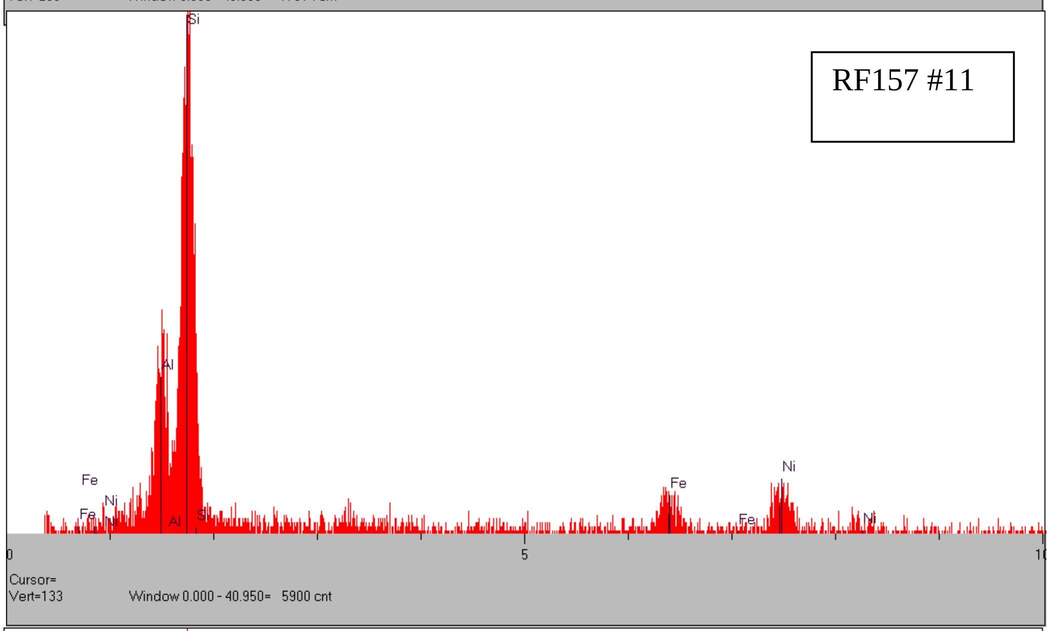

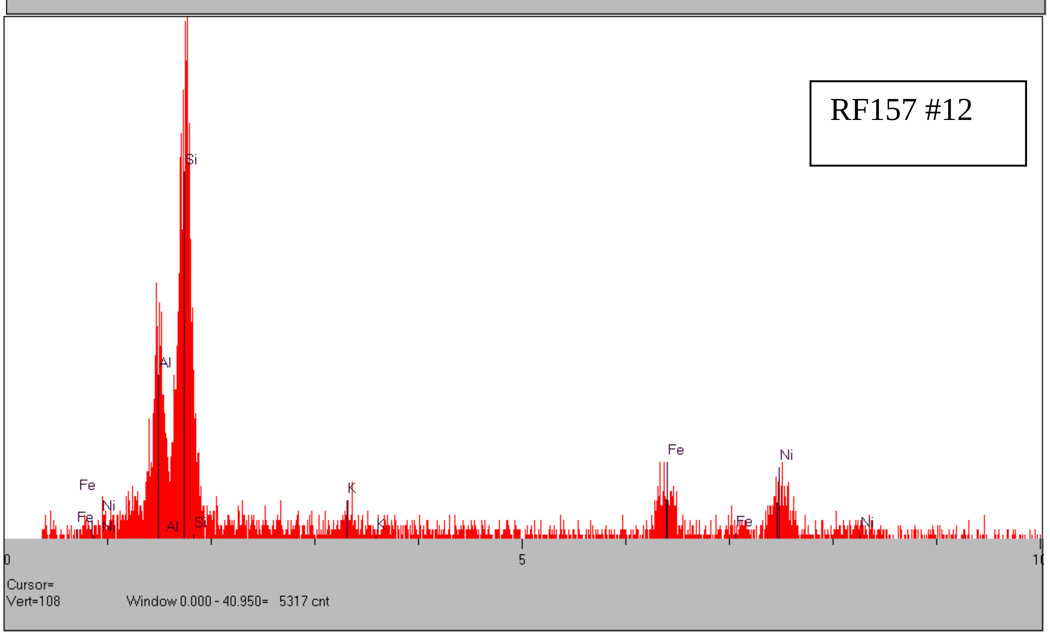

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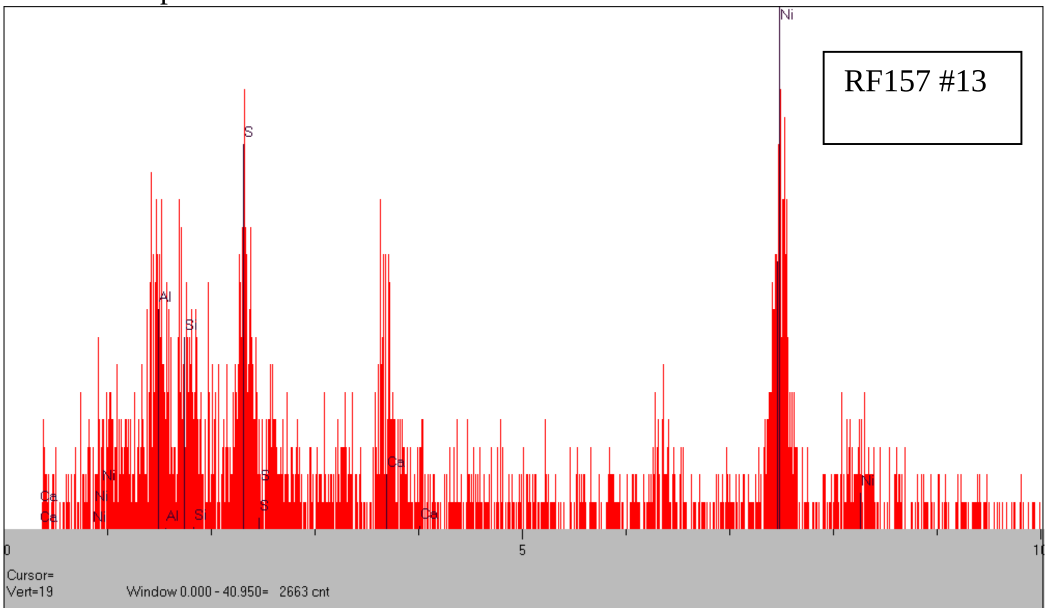

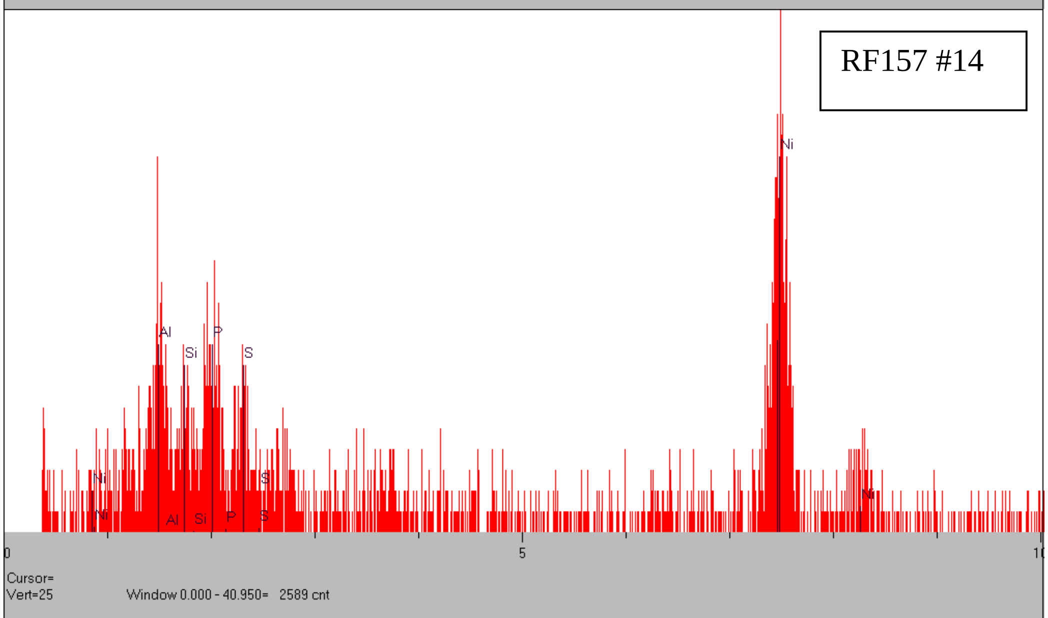

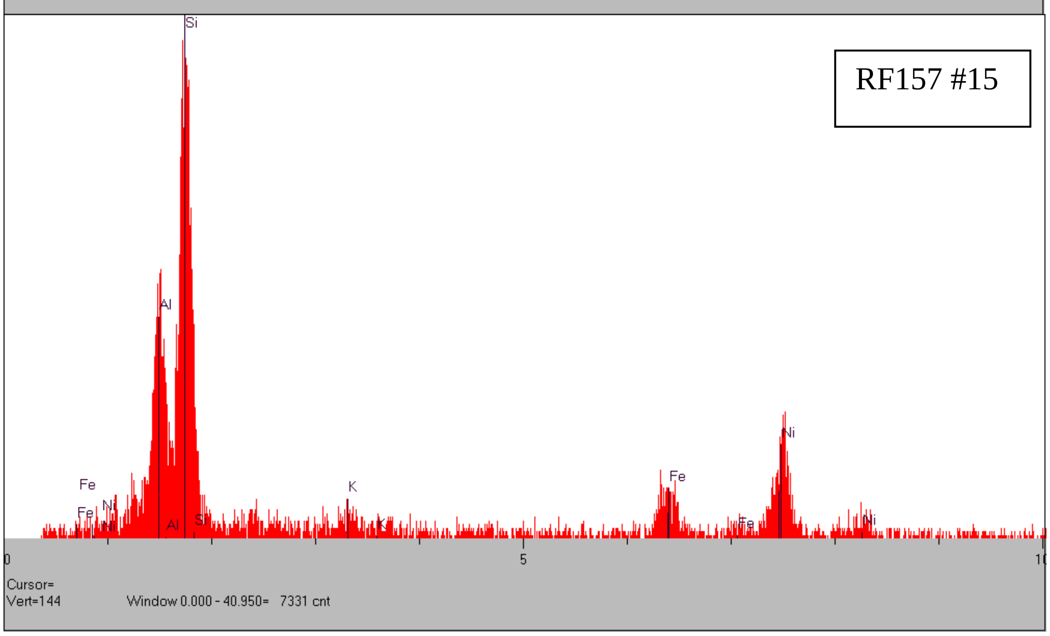

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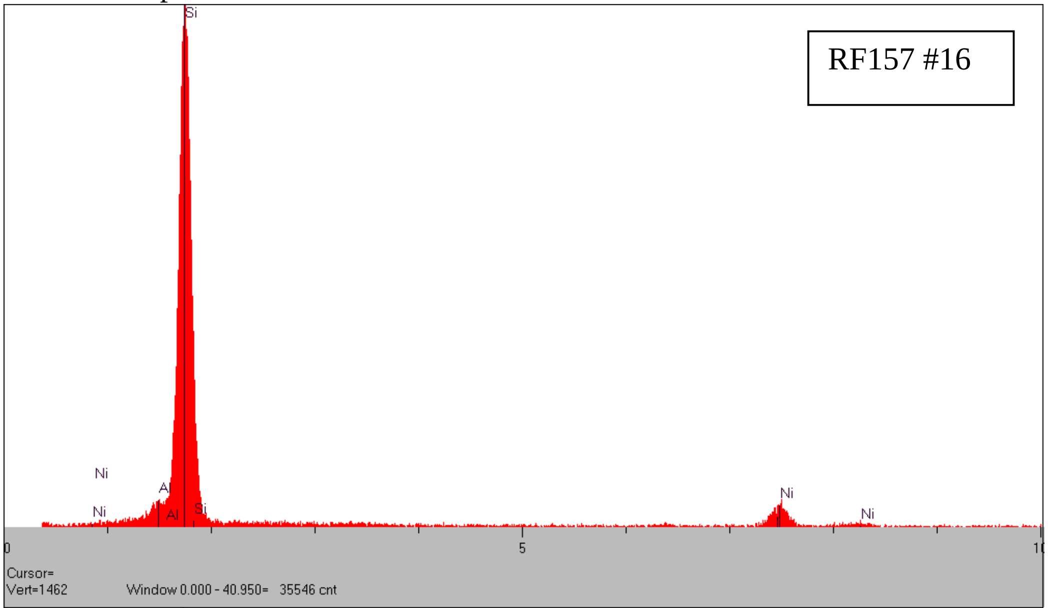

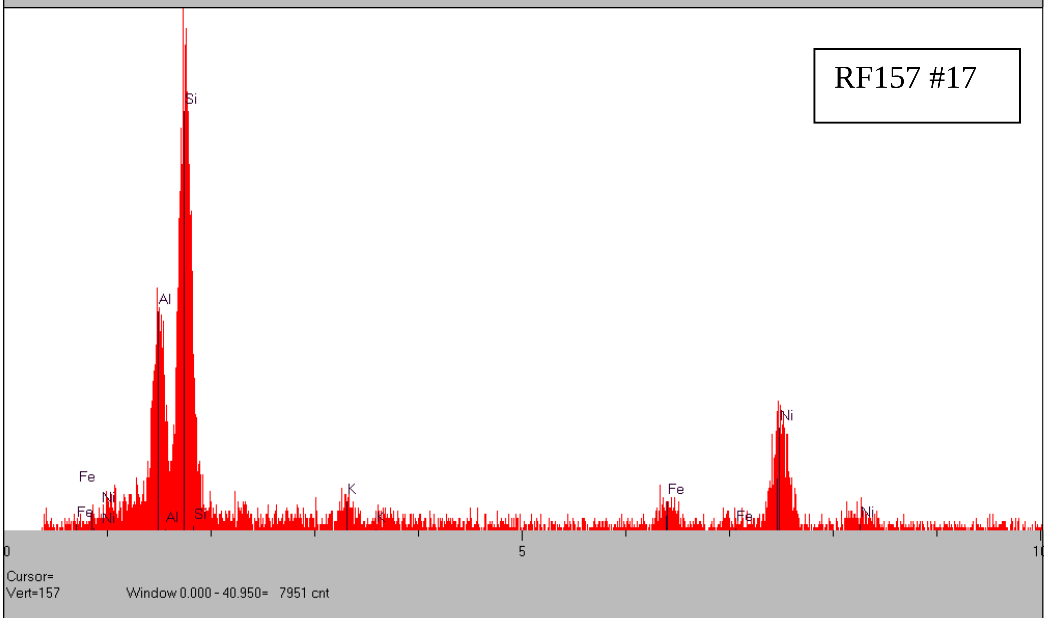

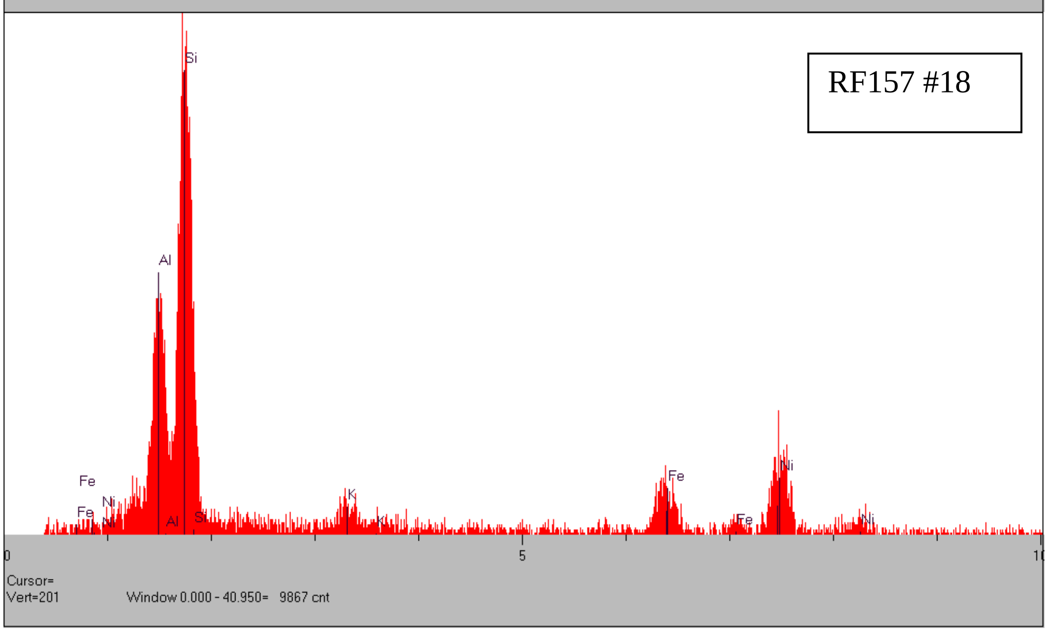

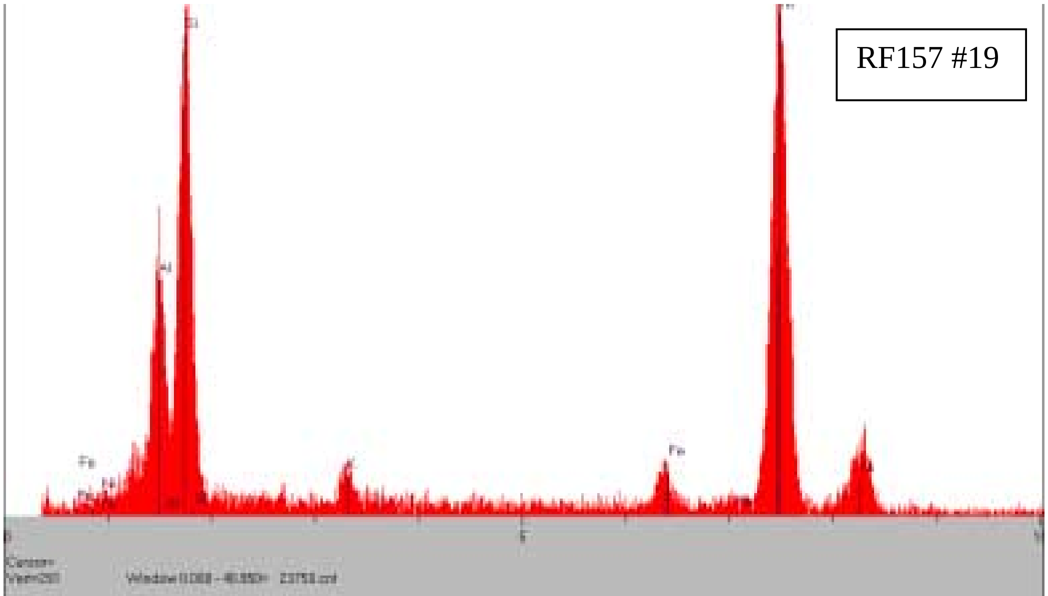

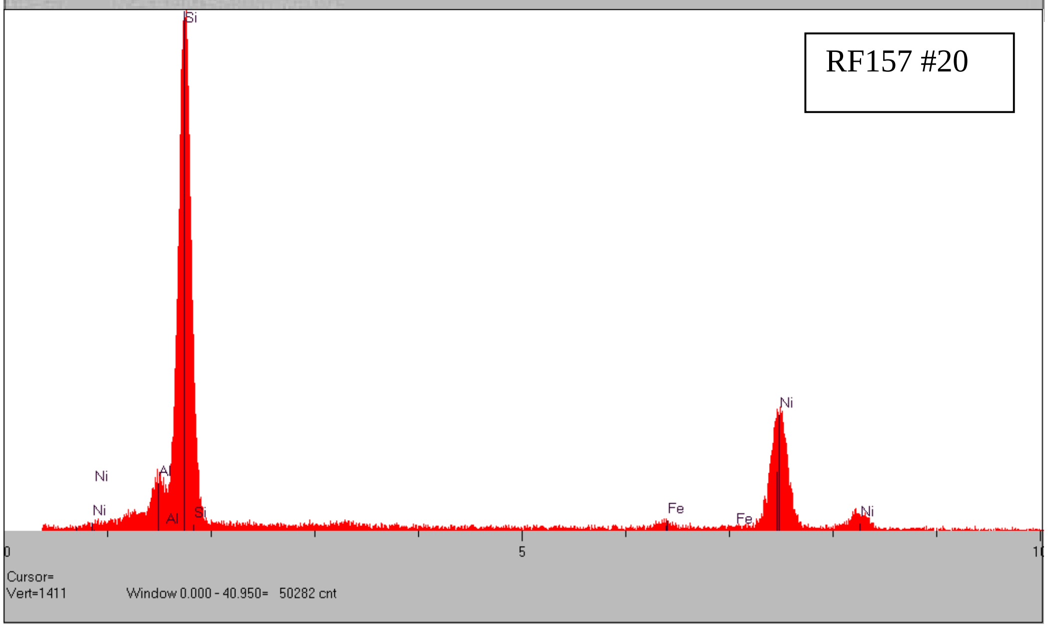

RF157Nanoplast

Appendix 5. Quality assurance summary

Precision

All samples were run in duplicate for the size fraction activity determination of Pu and Am and trace metals were run in duplicate and duplicate error ratio (DER = the difference between the duplicate measurement divided by the square root of the sum of the squares of the total propagated uncertainties) was calculated (Tables A5a – A5c). The Relative Percent Uncertainty (RPD) for other measured parameters such as SPM, DOC and trace metal concentrations in the less than 0.5μm fraction, and %C, %N and trace metal concentrations in freeze-dried were calculated based on the standard deviation of duplicate or triplicate measurements (Table A5d).

Table A5a: Duplicate error ratio (DER) for $^{239,240}Pu$. Maximum acceptable DER = 1.42.

ID	$^{239,240}Pu$ pCi/l	+/-	ID	$^{239,240}Pu$ pCi/l	+/-	Duplicate Error Ratio
RF130	0.0588	0.0018	RF140	0.0503	0.0166	0.5091
RF131	-	-	RF141	-	-	-
RF132	-	-	RF142	-	-	-
RF133	0.0225	0.0013	RF143	-	-	-
RF134	0.0077	0.0007	RF144	0.0015	0.0005	7.2074
RF135	0.0047	0.0006	RF145	0.0031	0.0007	1.7354
RF136	0.0172	0.0006	RF146	0.001	0.0005	20.7420
RF137	0.0421	0.0119	RF147	0.0059	0.0008	3.0352
RF138	0.0496	0.0095	RF148	0.0139	0.0012	3.7283
RF139	-	-	RF149	0.0025	0.0006	-
RF150	0.0822	0.0034	RF160	0.1082	0.0043	-4.7430
RF153	0.0142	0.0013	RF163	0.007	0.0012	4.0697
RF151	-	-	RF161	-	-	-
RF152	0.0834	0.0021	RF162	0.0424	0.0013	16.6004
RF154	0.0079	0.0006	RF164	0.0067	0.0007	1.3016
RF155	0.0041	0.0007	RF165	0.005	0.0012	-0.6478
RF156	0.0023	0.0005	RF166	0.0008	0.0005	2.1213
RF157	0.0164	0.0012	RF167	0.0175	0.0014	-0.5966
RF158	0.002	0.0006	RF168	0.0015	0.0007	0.5423
RF159	-	-	RF169	-	-	-
RF170	0.0944	0.0177	RF174	-0.0293	0.0187	4.8042
RF171	1.9695	0.0585	RF175	1.5155	1.7951	0.2528
RF172	-0.0414	0.0222	RF176	-0.0237	0.018	-0.6193
RF173	2.0927	0.0699	RF177	1.9628	0.0572	1.4382

Table A5b: Duplicate error ratio (DER) for ^{239,240}Pu from resuspension experiments. Maximum acceptable DER = 1.42.

ID	% of activity resuspended	+/-	ID	% of activity resuspended	+/-	Duplicate Error Ratio
P336	0.1812	0.0233	P337	-	-	-
P319	0.1200	0.0145	P321	0.1690	0.0184	-0.3687
P265	0.2157	0.0222	P267	0.2467	0.0280	-0.2677
P279	0.6856	0.0816	P281	0.8329	0.0951	-1.1164
P315	0.5309	0.0782	P317	1.0920	0.1300	-3.4743
P291	0.1330	0.0160	P293	0.1412	0.0173	-0.0609
P295	0.1686	0.0189	P297	0.2054	0.0222	-0.2974
P299	0.1965	0.0198	P301	0.1889	0.0205	0.0671
P303	0.1901	0.0207	P305	0.2171	0.0230	-0.2248
P178	0.0556	0.0085	P180	0.0529	0.0079	0.0153
P233	0.2071	0.0185	P235	0.2126	0.0195	-0.0567
P237	0.1509	0.0538	P239	0.2016	0.0179	-0.1391
P241	0.1949	0.0176	P243	0.1858	0.0167	0.0928
P269	0.4057	0.0376	P271	0.3554	0.0327	0.4977
P273	0.3454	0.0303	P275	0.3166	0.0272	0.3034
P245	0.8913	0.0826	P247	0.6992	0.0578	1.9311
P283	0.5823	0.0725	P285	0.6042	0.0791	-0.1551
P307	0.2215	0.0231	P309	0.2474	0.0249	-0.2266
P311	0.4581	0.0441	P313	0.4511	0.0429	0.0657
P323	0.4319	0.0410	P325	0.5103	0.0487	-0.7527
P331	0.6734	0.0735	P333	0.4012	0.0389	2.2962
P182	0.1518	0.0167	P184	0.1445	0.0154	0.0603
P338	0.1603	0.0190	P339	0.1624	0.0187	-0.0163
P340	0.1759	0.0202	P341	0.2110	0.0230	-0.2765
P342	0.3051	0.0316	P343	0.3513	0.0355	-0.4059
P344	0.5209	0.0484	P345	0.4119	0.0406	1.0617
P346	0.3890	0.0438	P347	0.3016	0.0330	0.7020
P348	0.3418	0.0334	P349	0.3593	0.0347	-0.1630
P249	0.1858	0.0158	P251	0.1810	0.0156	0.0514
P253	0.1766	0.0155	P255	0.1750	0.0150	0.0168
P257	0.1801	0.0154	P259	0.2029	0.0174	-0.2452
P261	0.1520	0.0133	P263	0.1739	0.0157	-0.2285
P287	0.4028	0.0555	P289	0.8299	0.0995	-2.8047

Table A5c. Duplicate error ratio (DER) for ^{241}Am. Maximum acceptable DER = 1.42.

ID	^{241}Am		ID	^{241}Am		Duplicate
	pCi/l	+/-		pCi/l	+/-	Error Ratio
RF130	0.0017	0.0008	RF140	0.003	0.001	-1.0151
RF131	-	-	RF141	-	-	-
RF132	-	-	RF142	-	-	-
RF133	-	-	RF143	-	-	-
RF134	0.0007	0.0004	RF144	-	-	-
RF135	0.0022	0.0004	RF145	-	-	-
RF136	0.0065	0.0005	RF146	-	-	-
RF137	0.003	0.0008	RF147	-	-	-
RF138	0.003	0.0005	RF148	-	-	-
RF139	0.067	0.0006	RF149	-	-	-
RF150	0.0672	0.0028	RF160	0.0768	0.0033	-2.2182
RF153	0.0023	0.0003	RF163	0.0092	0.0009	-7.2732
RF151	-	-	RF161	-	-	-
RF152	0.1377	0.0028	RF162	0.0406	0.002	28.2191
RF154	0.003	0.0005	RF164	0.0041	0.0004	-1.7179
RF155	0.0023	0.0005	RF165	0.003	0.0004	-1.0932
RF156	0.0031	0.0006	RF166	0.002	0.0004	1.5254
RF157	0.0106	0.0009	RF167	0.005	0.0004	5.6859
RF158	0.0018	0.0006	RF168	0.0006	0.0004	1.6641
RF159	0.0009	0.0004	RF169	0.0008	0.0004	0.1768
RF170	0.0018	0.0136	RF174	-0.0016	0.0133	0.1787
RF171	2.0192	0.0599	RF175	2.0152	0.0528	0.0501
RF172	0.0061	0.0155	RF176	-0.0063	0.0133	0.6071
RF173	1.9674	0.075	RF177	1.982	10.0524	-0.0015

Table A5d. Relative Percent Difference (RPD)

ID			ID			RPD
SPM	**mg/l**	**SD**		**mg/l**	**SD**	
4/27/00A	34.4	2.8	4/27/00B	27.5	1.9	22.29
DOC	**ppm**	**SD**		**ppm**	**SD**	
4/27/00A	3.94	0.41	4/27/00B	3.59	0.2	9.30
5/8/00A	6.66	2.94	5/8/00B	5.28	0.13	23.12
Carbon	**%**	**SD**		**%**	**SD**	
RF137	3.1	0.1	RF147	3.87	0.15	-22.09
RF157	11.94	0.05	RF167	10.68	0.35	-11.14
Nitrogen	**%**	**SD**		**%**	**SD**	
RF137	0.21	0.01	RF147	0.239	0.05	-12.92
RF157	0.6	0.01	RF167	0.56	0.03	6.9
Fe	**ppb**	**SD**	**ppb**	**SD**		
4/27/00A	40.31	1.1194087	4/27/00B	34.94	0.24458	14.27
5/8/00A	281.41	14.0705	5/8/00B	279.67	12.5852	0.62
RF137	0.15	0.002679	RF147	0.2	0.001	-28.57
RF157	10.3	0.185812	RF167	8.69	0.07291	16.96
Al	**ppb**	**SD**	**ppb**	**SD**		
4/27/00A	15.81	0.5229948	4/27/00B	14.81	0.22111	6.53
5/8/00A	291.67	13.577239	5/8/00B	290.43	25.099	0.43
RF137	0.07	0.0031493	RF147	0.16	0.00378	-78.26
RF157	13.32	0.240426	RF167	11.91	0.12148	11.18
Mn	**ppb**	**SD**	**ppb**	**SD**		
4/27/00A	35.14	0.1082312	4/27/00B	32.3	0.14664	8.42
5/8/00A	19.43	0.1775902	5/8/00B	21.87	0.19661	-11.82
RF137	86.05	0.254708	RF147	60.52	0.14706	34.84
RF157	131.41	0.696473	RF167	148.54	0.74567	-12.24

Accuracy

To ensure accuracy of activities determined by alpha spectrometry, the alpha spectrometer was calibrated from 3-7MeV using a NIST traceable 1” stainless steel planchete. Calibration checks were performed weekly during the time of instrument use. There were a few cases of samples being counted on detectors in which there was a >40kev shift of the standard peaks. In these cases, the samples were recounted on detectors without a shift and the recount data is the data reported. The efficiency for the detector is calculated with a 1” stainless steel standard. The efficiency is only used to approximate chemical yield as all samples have an internal tracer (^{242}Pu or ^{243}Am) added which provides the necessary efficiency information for data reduction. Chemical yields, referred to as tracer recoveries are listed in the raw data samples (Appendix 6).

Representativeness

Chain of custody forms for samples RIN 00R0972, 99A3372, 99A5936 and COC# TAMU42600, TAMU42700A, TAMU42700B, TAMU050800 were received and retained for this years work. All soil samples collected followed RFP method for soil collection. Water samples were collected according to site standard procedures. The water filtration followed the protocol as discussed in Baskaran, M., Murphy, D.J., Santschi, P.H., Orr, J.C., and Schink, D.R. 1993. A method for rapid in situ extraction and laboratory determination of Th, Pb, and Ra istopes from large volumes of seawater. Deep-Sea Research 40 (4), 849-865.

Comparability

Analytical methods used have followed established methods where necessary. All protocols have been referenced. The only deviation from protocol was adopted from Santschi et al. 1999 as noted in Appendix 2 p. A5.

Completeness

The number of samples analyzed matches the work plan.

Special Notes

There are two explanations for the low tracer recoveries for Pu and Am data in the size fractionated samples. First the initial samples that were run showed tracer recoveries and is attributed to poor equilibration. To combat this problem all subusequent samples at the $Fe(OH)_3$ stage were treated with concentrated hydrochloric and nitric acids and evaporated prior to beginning column chemistry. Samples run with this modification showed significant increases in tracer recovery. Second, some of the particulate samples (0.5- 20, >20 and total) caused severe clogging of the columns despite a “total” digestion procedure with consisted of concentrated nitric, hydrochloric, hydrofluoric and in some cases hydrogen peroxide. Also, the analyst neglected to add tracers to particulate sample numbers RF131, RF132, RF141 and RF142. The samples were run in hopes of determining activities via detector efficiency but these samples also experienced significant column clogging. These and all samples with <10% trac recovery were not used.

Appendix 6 Data Tables

*** These samples were split in half prior to analysis therefore final activities need to be doubled**

Sample	COC#	ID		vol	DET	eff	Background ^{242}Pu Cpm	+/-	^{239,240} Pu cpm	+/-	count time(s)	(min)
TOTAL	TAMU42700B	RF130	Pu	18	5	0.24	0.0028	0.0014	0	0	918010	15300
>20µm	TAMU42700B	RF131	Pu	90	5	0.24	0.0018	0.0005	0.0008	0.0003	513687	8561.5
0.5 -20µm	TAMU42700B	RF132	Pu	90	2	0.16	0.0035	0.0009	0.0012	0.0005	578118	9635.3
<0.5µm	TAMU42700B	RF133	Pu	18	6	0.21	0.0035	0.0016	0.0028	0.0014	918046	15301
100kDa - 0.5µm	TAMU42700B	RF134	Pu	36	7	0.19	0.0028	0.0014	0.0056	0.002	918047	15301
<100kDa	TAMU42700B	RF135	Pu	36	8	0.21	0	0	0.0063	0.0021	918045	15301
100kDa wash	TAMU42700B	RF136	Pu	36	9	0.22	0.0042	0.0017	0.0021	0.0012	918018	15300
3kDa - 0.5µm	TAMU42700B	RF137	Pu	36	10	0.21	0.0021	0.0012	0.0021	0.0012	918020	15300
<3kDa	TAMU42700B	RF138	Pu	36	6	0.21	0.0014	0.0004	0.002	0.0005	513720	8562
3kDa wash	TAMU42700B	RF139	Pu	36	2	0.16	0.0031	0.0009	0.0011	0.0005	559912	9331.9
TOTAL	TAMU42700B	RF140	Pu	18	3	0.2	0.0012	0.0005	0.0017	0.0006	559911	9331.9
>20µm	TAMU42700B	RF141	Pu	90	3	0.2	0.0023	0.0007	0.0019	0.0007	578119	9635.3
0.5 -20µm	TAMU42700B	RF142	Pu	90	4	0.14	0.0004	0.0004	0.0011	0.0006	578120	9635.3
<0.5µm	TAMU42700B	RF143	Pu	18	4	0.14	0.0002	0.0002	0.0012	0.0005	559913	9331.9
100kDa - 0.5µm	TAMU42700B	RF144	Pu	36	5	0.24	0.0028	0.0014	0	0	144832	2413.9
<100kDa	TAMU42700B	RF145	Pu	36	6	0.21	0.0035	0.0016	0.0028	0.0014	144755	2412.6
100kDa wash	TAMU42700B	RF146	Pu	36	7	0.19	0.0056	0.002	0.0028	0.0014	144840	2414
3kDa - 0.5µm	TAMU42700B	RF147	Pu	36	8	0.21	0.0028	0.0014	0	0	144842	2414
<3kDa	TAMU42700B	RF148	Pu	36	9	0.22	0.0021	0.0012	0.0021	0.0012	144859	2414.3
3kDa wash	TAMU42700B	RF149	Pu	36	10	0.21	0.0035	0.0016	0.0021	0.0012	144908	2415.1

ID	^{242}Pu Counts	+ / -	Bkg Corrected ^{242}Pu Counts	+ / -	$^{239,240}Pu$ Counts	+ / -	Bkg Corrected $^{239,240}Pu$ Counts	+ / -	^{242}Pu Cpm	+ / -	$^{239,240}Pu$ Cpm	+ / -	^{242}Pu Dpm Added	+ / -
RF130	4650	68.15	4607.2	71.437	2300	47.92	2300	47.92	0.3011	0.0047	0.1503	0.0031	4.72	0.035
RF131	34	5.83	18.589	7.2294	47	6.86	40.15084	7.408	0.0022	0.0008	0.0047	0.0009	4.72	0.035
RF132	43	6.56	9.2765	10.902	12	3.46	0.43764	6.222	0.0010	0.0011	0.0000	0.0006	4.72	0.035
RF133	5130	71.64	5076.4	75.537	1030	32.05	987.1579	38.55	0.3318	0.0049	0.0645	0.0025	4.72	0.035
RF134	4520	67.21	4477.2	70.541	689	26.25	603.3156	40.09	0.2926	0.0046	0.0394	0.0026	4.72	0.035
RF135	7160	84.64	7160	84.64	702	26.5	605.6053	41.65	0.4680	0.0055	0.0396	0.0027	4.72	0.035
RF136	11900	109.31	11836	112.41	3520	59.35	3487.869	62.18	0.7736	0.0073	0.2280	0.0041	4.72	0.035
RF137	168	12.96	135.87	22.631	129	11.36	96.8693	21.75	0.0089	0.0015	0.0063	0.0014	4.72	0.035
RF138	129	11.36	117.01	11.976	138	11.75	120.876	12.61	0.0137	0.0014	0.0141	0.0015	4.72	0.035
RF139	361	17.78	332.07	19.507	207	14.39	196.7349	15.10	0.0356	0.0021	0.0211	0.0016	4.72	0.035
RF140	84	9.17	72.802	10.448	47	6.86	31.13586	9.111	0.0078	0.0011	0.0033	0.0010	4.72	0.035
RF141	128	11.31	105.84	13.305	56	7.8	37.6929	10.14	0.0110	0.0014	0.0039	0.0011	4.72	0.035
RF142	10	3.16	6.1459	4.984	11	3.32	0.401133	6.962	0.0006	0.0005	0.0000	0.0007	4.72	0.035
RF143	153	12.37	151.13	12.51	122	11.05	110.8017	12.13	0.0162	0.0013	0.0119	0.0013	4.72	0.035
RF144	2510	50.1	2503.2	50.214	77	8.77	77	8.770	1.0370	0.0208	0.0319	0.0036	4.72	0.035
RF145	860	29.33	851.56	29.572	56	7.48	49.24477	8.207	0.3530	0.0123	0.0204	0.0034	4.72	0.035
RF146	1800	42.4	1786.5	42.669	47	6.86	40.2408	7.647	0.7401	0.0177	0.0167	0.0032	4.72	0.035
RF147	868	29.46	861.24	29.653	90	9.49	90	9.490	0.3568	0.0123	0.0373	0.0039	4.72	0.035
RF148	890	29.83	884.93	29.973	217	14.73	211.9299	15.02	0.3665	0.0124	0.0878	0.0062	4.72	0.035
RF149	1890	43.45	1881.5	43.614	95	9.75	89.92822	10.18	0.7791	0.0181	0.0372	0.0042	4.72	0.035

ID	$^{239,240}Pu$ dpm	+/-	Blank Corrected $^{239,240}Pu$ dpm	+/-	$^{239,240}Pu$ pCi	+/-	$^{239,240}Pu$ PCi/L	+/-	Tracer recovery	+/-
RF130	2.356	0.061	2.260	0.072	1.027	0.033	0.0571	0.0018	26.58	0.46
RF131	10.195	4.388	10.098	4.388	4.590	1.995	0.0510	0.0222	0.19	0.07
RF132	0.223	3.176	0.126	3.177	0.057	1.444	0.0006	0.0160	0.13	0.15
RF133	0.918	0.038	0.822	0.054	0.373	0.024	0.0207	0.0014	33.47	0.56
RF134	0.636	0.043	0.540	0.058	0.245	0.026	0.0068	0.0007	32.63	0.57
RF135	0.399	0.028	0.303	0.047	0.138	0.021	0.0038	0.0006	47.21	0.66
RF136	1.391	0.028	1.295	0.047	0.588	0.021	0.0163	0.0006	74.50	0.90
RF137	3.365	0.941	3.269	0.942	1.486	0.428	0.0413	0.0119	0.90	0.15
RF138	4.876	0.713	4.780	0.714	2.173	0.324	0.0603	0.0090	1.38	0.14
RF139	2.796	0.270	2.700	0.273	1.227	0.124	0.0341	0.0034	4.71	0.28
RF140	2.019	0.658	1.922	0.659	0.874	0.300	0.0485	0.0166	0.83	0.12
RF141	1.681	0.499	1.585	0.500	0.720	0.227	0.0080	0.0025	1.16	0.15
RF142	0.308	5.353	0.212	5.353	0.096	2.433	0.0011	0.0270	0.10	0.08
RF143	3.460	0.475	3.364	0.476	1.529	0.217	0.0850	0.0120	2.45	0.20
RF144	0.145	0.017	0.049	0.041	0.022	0.019	0.0006	0.0005	91.55	1.96
RF145	0.273	0.046	0.177	0.060	0.080	0.027	0.0022	0.0008	35.61	1.26
RF146	0.106	0.020	0.010	0.043	0.005	0.020	0.0001	0.0005	82.52	2.06
RF147	0.493	0.055	0.397	0.067	0.180	0.030	0.0050*	0.0008	35.99	1.27
RF148	1.130	0.089	1.034	0.097	0.470	0.044	0.0131	0.0012	35.30	1.22
RF149	0.226	0.026	0.129	0.046	0.059	0.021	0.0016	0.0006	78.60	1.91

Sample	COC#	ID		vol	DET	eff	Background ^{242}Pu Cpm	+/-	$^{239,240}Pu$ cpm	+/-	count time(s)	(min)
TOTAL	TAMU050800	RF150	Pu	12	7	0.19	0.0025	0.0006	0.004	0.0007	371417	6190.3
>20µm	TAMU050800	RF151	Pu	90	5	0.24	0.0004	0.0004	0.0004	0.0004	97029	1617.2
0.5 -20µm	TAMU050800	RF152	Pu	90	2	0.16	0.0038	0.001	0.0012	0.0005	405188	6753.1
0.5 -20µm	TAMU050800	RF152B	Pu	90	3	0.2	0.0019	0.0007	0.0019	0.0007	405206	6753.4
<0.5µm	TAMU050800	RF153	Pu	18	5	0.24	0.0035	0.0016	0	0	309632	5160.5
100kDa - 0.5µm	TAMU050800	RF154	Pu	36	6	0.21	0.0035	0.0016	0.0029	0.0014	309631	5160.5
<100kDa	TAMU050800	RF155	Pu	36	7	0.19	0.0028	0.0014	0.0056	0.002	309632	5160.5
100kDa wash	TAMU050800	RF156	Pu	36	8	0.21	0	0	0	0	309631	5160.5
3kDa - 0.5µm	TAMU050800	RF157	Pu	36	9	0.22	0.0042	0.0017	0.0028	0.0014	309639	5160.7
<3kDa	TAMU050800	RF158	Pu	36	10	0.21	0.0042	0.0017	0.0021	0.0012	309640	5160.7
3kDa wash	TAMU050800	RF159	Pu	36	11	19	0.0021	0.0007	0.0019	0.0007	309642	5160.7
TOTAL	TAMU050800	RF160	Pu	12	4	0.14	0.0018	0.0008	0.0018	0.0008	254291	4238.2
>20µm	TAMU050800	RF161	Pu	90	6	0.21	0.0018	0.0008	0.0022	0.0009	97031	1617.2
0.5 -20µm	TAMU050800	RF162	Pu	90	8	0.21	0.0022	0.0006	0.0025	0.0006	85920	1432
0.5 -20µm	TAMU050800	RF162B	Pu	90	2	0.16	0.0035	0.0009	0.0012	0.0005	154797	2580
<0.5µm	TAMU050800	RF163	Pu	18	5	0.24	0.0035	0.0016	0	0	167709	2795.2
100kDa - 0.5µm	TAMU050800	RF164	Pu	36	6	0.21	0.0035	0.0016	0.0028	0.0014	169662	2827.7
<100kDa	TAMU050800	RF165	Pu	36	7	0.19	0.0028	0.0014	0.0056	0.002	167719	2795.3
100kDa wash	TAMU050800	RF166	Pu	36	8	0.21	0.0014	0.001	0.0063	0.0021	167736	2795.6
3kDa - 0.5µm	TAMU050800	RF167	Pu	36	9	0.22	0.0035	0.0016	0.0021	0.0012	167754	2795.9
<3kDa	TAMU050800	RF168	Pu	36	10	0.21	0.0035	0.0016	0.0028	0.0014	167761	2796
3kDa wash	TAMU050800	RF169	Pu	36	11	0.19	0.0014	0.0006	0.0019	0.0008	445797	7430

ID	^{242}Pu Counts	+/-	Bkg Corrected ^{242}Pu Counts	+/-	$^{239,240}Pu$ Counts	+/-	Bkg Corrected $^{239,240}Pu$ Counts	+/-	^{242}Pu Cpm	+/-	$^{239,240}Pu$ Cpm	+/-	^{242}Pu Dpm Added	+/-
RF150	1290	35.9	1274.5	36.085	568	23.83	543.2389	24.27	0.2059	0.0058	0.0878	0.0039	4.72	0.035
RF151	15	3.87	14.353	3.9237	80	8.94	79.35314	8.963	0.0089	0.0024	0.0491	0.0055	4.72	0.035
RF152	1860	43.16	1834.3	43.634	5840	76.41	5831.896	76.50	0.2716	0.0065	0.8636	0.0113	4.72	0.035
RF152B	7360	85.78	7347.2	85.9	2460	49.57	2447.168	49.78	1.0879	0.0127	0.3624	0.0074	4.72	0.035
RF153	2270	47.62	2251.9	48.3	281	16.76	281	16.76	0.4364	0.0094	0.0545	0.0032	4.72	0.035
RF154	4890	69.92	4871.9	70.385	689	26.25	674.2925	27.26	0.9441	0.0136	0.1307	0.0053	4.72	0.035
RF155	1830	42.76	1815.6	43.366	166	12.88	137.101	16.44	0.3518	0.0084	0.0266	0.0032	4.72	0.035
RF156	3570	59.71	3570	59.71	156	12.49	156	12.49	0.6918	0.0116	0.0302	0.0024	4.72	0.035
RF157	4180	64.68	4158.3	65.282	613	24.76	598.5502	25.79	0.8058	0.0127	0.1160	0.0050	4.72	0.035
RF158	2260	47.53	2238.3	48.346	100	10	89.1626	11.80	0.4337	0.0094	0.0173	0.0023	4.72	0.035
RF159	4720	68.72	4709.2	68.815	68	8.25	58.19467	8.95	0.9125	0.0133	0.0113	0.0017	4.72	0.035
RF160	1910	43.7	1902.4	43.833	1170	34.22	1162.371	34.39	0.4489	0.0103	0.2743	0.0081	4.72	0.035
RF161	197	14.04	194.09	14.1	637	25.24	633.4422	25.28	0.1200	0.0087	0.3917	0.0156	4.72	0.035
RF162	1210	34.77	1206.8	34.779	1890	43.47	1886.42	43.48	0.8428	0.0243	1.3173	0.0304	4.72	0.035
RF162B	2110	45.98	2101	46.039	297	58.92	293.9041	58.94	0.8143	0.0178	0.1139	0.0228	4.72	0.035
RF163	1710	41.35	1700.2	41.581	110	10.49	110	10.49	0.6083	0.0149	0.0394	0.0038	4.72	0.035
RF164	2340	48.41	2330.1	48.612	283	16.82	275.0824	17.28	0.8240	0.0172	0.0973	0.0061	4.72	0.035
RF165	524	22.89	516.17	23.222	62	7.87	46.34623	9.622	0.1847	0.0083	0.0166	0.0034	4.72	0.035
RF166	2280	47.76	2276.1	47.84	62	7.87	44.38772	9.818	0.8142	0.0171	0.0159	0.0035	4.72	0.035
RF167	2330	48.27	2320.2	48.468	361	19	355.1286	19.30	0.8299	0.0173	0.1270	0.0069	4.72	0.035
RF168	966	31.08	956.21	31.387	38	6.16	30.17115	7.299	0.3420	0.0112	0.0108	0.0026	4.72	0.035
RF169	18	4.24	7.5981	6.0006	13	3.61	-1.11691	6.800	0.0010	0.0008	-0.0002	0.0009	4.72	0.035

ID	^{239,240} Pu dpm	+ / -	Blank Corrected ^{239,240} Pu dpm	+ / -	^{239,240} Pu pCi	+ / -	^{239,240} Pu PCi/L	+ / -	Tracer recovery	+ / -
RF150	2.012	0.106	1.916	0.113	0.871	0.051	0.0726	0.0043	22.96	0.67
RF151	26.095	7.719	25.999	7.719	11.818	3.508	0.1313	0.0390	0.78	0.21
RF152	15.006	0.408	14.910	0.409	6.777	0.186	0.0753	0.0021	35.97	0.90
RF152B	1.572	0.037	1.476	0.053	0.671	0.024	0.0075	0.0003	115.25	1.59
RF153	0.589	0.037	0.493	0.053	0.224	0.024	0.0124	0.0013	38.52	0.87
RF154	0.653	0.028	0.557	0.047	0.253	0.021	0.0070	0.0006	95.25	1.55
RF155	0.356	0.044	0.260	0.058	0.118	0.026	0.0033	0.0007	39.23	0.98
RF156	0.206	0.017	0.110	0.041	0.050	0.019	0.0014	0.0005	69.79	1.28
RF157	0.679	0.031	0.583	0.049	0.265	0.022	0.0074*****	0.0006	77.60	1.35
RF158	0.188	0.025	0.092	0.045	0.042	0.021	0.0012	0.0006	43.76	1.00
RF159	0.058	0.009	-0.038	0.039	-0.017	0.018	-0.0005	0.0005	1.02	0.02
RF160	2.884	0.108	2.788	0.115	1.267	0.052	0.1056	0.0043	67.93	1.64
RF161	15.405	1.277	15.308	1.277	6.958	0.581	0.0773	0.0065	12.11	0.88
RF162	7.378	0.272	7.282	0.275	3.310	0.125	0.0368	0.0014	85.03	2.53
RF162B	0.660	0.133	0.564	0.138	0.256	0.063	0.0028	0.0007	107.83	2.49
RF163	0.305	0.030	0.209	0.048	0.095	0.022	0.0053	0.0012	53.70	1.37
RF164	0.557	0.037	0.461	0.053	0.210	0.024	0.0058	0.0007	83.13	1.84
RF165	0.424	0.090	0.328	0.098	0.149	0.044	0.0041	0.0012	20.59	0.94
RF166	0.092	0.020	-0.004	0.043	-0.002	0.020	-0.0001	0.0005	82.14	1.83
RF167	0.722	0.042	0.626	0.057	0.285	0.026	0.0079*****	0.0007	79.92	1.77
RF168	0.149	0.036	0.053	0.052	0.024	0.024	0.0007	0.0007	34.50	1.16
RF169	-0.694	-4.260	-0.790	4.260	-0.359	1.936	-0.0100	0.0538	0.11	0.09

Sample	Spike Added Dpm	ID		vol	DET	eff	Background ^{242}Pu Cpm	+ / -	^{239,240} Pu cpm	+ / -	count time(s)	(min)
Blank		RF170	Pu	1	5	0.24	0.0035	0.0016	0	0	227767	3796.1
Blank Spike	5.34	RF171	Pu	1	6	0.21	0.0035	0.0016	0.0028	0.0014	227766	3796.1
Blank		RF172	Pu	1	7	0.19	0.0028	0.0014	0.0049	0.0019	227766	3796.1
Blank Spike	5.3	RF173	Pu	1	8	0.21	0.0014	0.001	0.0063	0.0021	227766	3796.1
Blank		RF174	Pu	1	9	0.22	0.0035	0.0016	0.0021	0.0012	227757	3796
Blank Spike	5.23	RF175	Pu	1	10	0.21	0.0028	0.0014	0.0028	0.5	227756	3795.9
Blank		RF176	Pu	1	11	0.19	0.0016	0.0006	0.0023	0.0007	227756	3795.9
Blank Spike	5.3	RF177	Pu	1	12	0.2	0.0045	0.001	0.0026	0.0008	227756	3795.9

ID	^{242}Pu Counts	+ / -	Bkg Corrected ^{242}Pu Counts	+ / -	^{239,240} Pu Counts	+ / -	Bkg Corrected ^{239,240} Pu Counts	+ / -	^{242}Pu Cpm	+ / -	^{239,240} Pu Cpm	+ / -	^{242}Pu Dpm Added	+ / -
RF170	17400	131.96	17387	132.09	1120	33.51	1120	33.510	4.5801	0.0348	0.2950	0.0088	4.72	0.035
RF171	2730	52.26	2716.7	52.597	2560	50.56	2549.371	50.839	0.7157	0.0139	0.6716	0.0134	4.72	0.035
RF172	1280	35.81	1269.4	36.202	20	4.47	1.39911	8.332	0.3344	0.0095	0.0004	0.0022	4.72	0.035
RF173	2050	45.24	2044.7	45.393	2060	45.42	2036.085	46.114	0.5386	0.0120	0.5364	0.0121	4.72	0.035
RF174	1950	44.12	1936.7	44.518	21	4.58	13.02851	6.493	0.5102	0.0117	0.0034	0.0017	4.72	0.035
RF175	2280	47.7	2269.4	47.995	1660	40.68	1649.371	1898.4	0.5978	0.0126	0.4345	0.5001	4.72	0.035
RF176	2600	50.99	2593.9	51.042	33	5.74	24.26935	6.369	0.6833	0.0134	0.0064	0.0017	4.72	0.035
RF177	2840	53.25	2822.9	53.394	2650	51.43	2640.131	51.516	0.7437	0.0141	0.6955	0.0136	4.72	0.035

ID	^{239,240} Pu dpm	+ / -	Blank Corrected ^{239,240} Pu dpm	+ / -	^{239,240} Pu pCi	+ / -	^{239,240} Pu PCi/L	+ / -	Tracer recovery	+ / -	Spike recovery	+ / -
RF170	0.304	0.009	0.208	0.039	0.094	0.018	0.0944	0.0177	404.32	4.29		
RF171	4.429	0.123	4.333	0.129	1.970	0.059	1.9695	0.0585	72.20	1.50	81.14	2.58
RF172	0.005	0.031	-0.091	0.049	-0.041	0.022	-0.0414	0.0222	37.29	1.10		
RF173	4.700	0.149	4.604	0.154	2.093	0.070	2.0927	0.0699	54.34	1.27	86.87	517.29
RF174	0.032	0.016	-0.065	0.041	-0.029	0.019	-0.0293	0.0187	49.13	1.19		
RF175	3.430	3.949	3.334	3.949	1.516	1.795	1.5155	1.7951	60.32	1.35	63.75	31.81
RF176	0.044	0.012	-0.052	0.040	-0.024	0.018	-0.0237	0.0180	76.20	1.60		
RF177	4.414	0.120	4.318	0.126	1.963	0.057	1.9628	0.0572	78.78	1.60	81.47	21.45

Sample	COC#	ID		vol	DET	eff	Background ^{243}Am Cpm	+/-	^{241}Am cpm	+/-	count time(s)	(min)
TOTAL	TAMU42700B	RF130	Am	18	9	0.220	0.0039	0.0007	0.001	0.0004	86124	1435.4
>20µm	TAMU42700B	RF131	Am	90	7	0.190	0.0042	0.0017	0.0014	0.0010	72330	1205.5
0.5 -20µm	TAMU42700B	RF132	Am	90	8	0.210	0.0042	0.0017	0	0.0000	72330	1205.5
<0.5µm	TAMU42700B	RF133	Am	18	2	0.160	0.0017	0.0006	0.0002	0.0002	224236	3737.3
100kDa - 0.5µm	TAMU42700B	RF134	Am	36	3	0.200	0.0014	0.0006	0.0007	0.0004	224236	3737.3
<100kDa	TAMU42700B	RF135	Am	36	4	0.140	0.0014	0.0006	0	0.0000	224237	3737.3
100kDa wash	TAMU42700B	RF136	Am	36	5	0.240	0.0007	0.0007	0.0014	0.0010	224235	3737.3
3kDa - 0.5µm	TAMU42700B	RF137	Am	36	6	0.210	0.0021	0.0012	0.0028	0.0014	224236	3737.3
<3kDa	TAMU42700B	RF138	Am	36	7	0.190	0.0056	0.0020	0.0014	0.0010	224237	3737.3
3kDa wash	TAMU42700B	RF139	Am	36	8	0.210	0.0042	0.0017	0	0.0000	224236	3737.3
TOTAL	TAMU42700B	RF140	Am	18	9	0.220	0.0021	0.0012	0	0.0000	224231	3737.2
>20µm	TAMU42700B	RF141	Am	90	10	0.210	0.0028	0.0014	0.0007	0.0007	267293	4454.9
0.5 -20µm	TAMU42700B	RF142	Am	90	11	0.190	0.0021	0.0007	0.0005	0.0004	267293	4454.9
<0.5µm	TAMU42700B	RF143	Am	18	10	0.240	0.0028	0.0014	0	0.0000	224231	3737.2
100kDa - 0.5µm	TAMU42700B	RF144	Am	36	11	0.190	0.0019	0.0007	0.0002	0.0002	224233	3737.2
<100kDa	TAMU42700B	RF145	Am	36	12	0.200	0.0005	0.0004	0.0005	0.0004	224249	3737.5
100kDa wash	TAMU42700B	RF146	Am	36	5	0.240	0.0004	0.0004	0.0004	0.0004	53421	890.4
3kDa - 0.5µm	TAMU42700B	RF147	Am	36	6	0.210	0.0011	0.0006	0.0037	0.0012	53421	890.4
<3kDa	TAMU42700B	RF148	Am	36	7	0.190	0.0004	0.0004	0.0007	0.0005	53421	890.4
3kDa wash	TAMU42700B	RF149	Am	36	8	0.210	0.003	0.0011	0	0.0000	53420	890.3

ID	^{243}Am Counts	+ / -	Bkg Corrected ^{243}Am Counts	+ / -	^{241}Am Counts	+ / -	Bkg Corrected ^{241}Am Counts	+ / -	^{243}Am Cpm	+ / -	^{241}Am Cpm	+ / -	^{243}Am Dpm Added	+ / -
RF130	1630	40.32	1624.40	40.33	24	4.9	22.56	4.93	1.13	0.03	0.02	0.00	6.45	0.050
RF131	7	2.65	1.94	3.36	36	6	34.31	6.12	0.00	0.00	0.03	0.01	6.45	0.050
RF132	5	2.24	-0.06	3.05	1	1	1.00	1.00	0.00	0.00	0.00	0.00	6.45	0.050
RF133	3980	63.06	3973.65	63.11	19	4.36	18.25	4.42	1.06	0.02	0.00	0.00	6.45	0.050
RF134	4390	66.29	4384.77	66.32	71	8.43	68.38	8.56	1.17	0.02	0.02	0.00	6.45	0.050
RF135	4080	63.87	4074.77	63.91	141	11.87	141.00	11.87	1.09	0.02	0.04	0.00	6.45	0.050
RF136	5150	71.77	5147.38	71.82	456	21.35	450.77	21.67	1.38	0.02	0.12	0.01	6.45	0.050
RF137	4340	65.89	4332.15	66.05	122	11.05	111.54	12.23	1.16	0.02	0.03	0.00	6.45	0.050
RF138	2870	53.56	2849.07	54.07	129	11.36	123.77	11.95	0.76	0.01	0.03	0.00	6.45	0.050
RF139	2930	54.11	2914.30	54.49	261	16.16	261.00	16.16	0.78	0.01	0.07	0.00	6.45	0.050
RF140	5460	73.86	5452.15	74.00	125	11.18	125.00	11.18	1.46	0.02	0.03	0.00	6.45	0.050
RF141	27	5.2	14.53	8.12	93	9.64	89.88	10.13	0.00	0.00	0.02	0.00	6.45	0.050
RF142	14	3.74	4.64	4.87	0	0	-2.23	1.58	0.00	0.00	0.00	0.00	6.45	0.050
RF143	5290	72.73	5279.54	72.92	29	5.39	29.00	5.39	1.41	0.02	0.01	0.00	6.45	0.050
RF144	73	8.54	65.90	8.90	19	4.36	18.25	4.42	0.02	0.00	0.00	0.00	6.45	0.050
RF145	401	20.02	399.13	20.06	9	3	7.13	3.28	0.11	0.01	0.00	0.00	6.45	0.050
RF146	108	10.39	107.64	10.40	8	2.83	7.64	2.85	0.12	0.01	0.01	0.00	6.45	0.050
RF147	7	2.65	6.02	2.71	2	1.41	-1.29	1.75	0.01	0.00	0.00	0.00	6.45	0.050
RF148	11	3.16	10.64	3.18	0	0	-0.62	0.44	0.01	0.00	0.00	0.00	6.45	0.050
RF149	1160	34.06	1157.33	34.07	50	7.07	50.00	7.07	1.30	0.04	0.06	0.01	6.45	0.050

ID	^{241}Am dpm	+ / -	Blank Corrected ^{241}Am	+ / -	^{241}Am pCi	+ / -	^{241}Am PCi/L	+ / -	Tracer recovery	+ / -
RF130	0.090	0.020	0.0432	0.0338	0.0196	0.0154	0.0011	0.0009	79.75	2.08
RF131	114.262	199.296	114.2157	199.2965	51.9162	90.5893	0.5768	1.0065	0.13	0.23
RF132	-102.219	-4938.335	-102.2651	4938.3350	-46.4842	2244.6977	-0.5165	24.9411	0.00	-0.19
RF133	0.030	0.007	-0.0168	0.0284	-0.0076	0.0129	-0.0004	0.0007	103.03	1.82
RF134	0.101	0.013	0.0541	0.0303	0.0246	0.0138	0.0007	0.0004	90.95	1.55
RF135	0.223	0.019	0.1767	0.0335	0.0803	0.0152	0.0022	0.0004	120.74	2.11
RF136	0.565	0.028	0.5184	0.0394	0.2356	0.0179	0.0065	0.0005	88.97	1.42
RF137	0.166	0.018	0.1196	0.0331	0.0544	0.0150	0.0015*	0.0004	85.58	1.47
RF138	0.280	0.028	0.2338	0.0389	0.1063	0.0177	0.0030	0.0005	62.21	1.28
RF139	0.578	0.037	0.5312	0.0464	0.2415	0.0211	0.0067	0.0006	57.57	1.17
RF140	0.148	0.013	0.1014	0.0306	0.0461	0.0139	0.0026	0.0008	102.81	1.61
RF141	39.909	22.758	39.8629	22.7585	18.1195	10.3448	0.2013	0.1149	0.24	0.13
RF142	-3.093	-3.911	-3.1396	3.9115	-1.4271	1.7780	-0.0159	0.0198	0.09	0.09
RF143	0.035	0.007	-0.0110	0.0283	-0.0050	0.0128	-0.0003	0.0007	91.26	1.45
RF144	1.786	0.496	1.7401	0.4964	0.7909	0.2257	0.0220	0.0063	1.44	0.19
RF145	0.115	0.053	0.0688	0.0600	0.0313	0.0273	0.0009	0.0008	8.28	0.42
RF146	0.458	0.177	0.4116	0.1787	0.1871	0.0812	0.0052	0.0023	7.81	0.76
RF147	-1.387	-1.979	-1.4330	1.9792	-0.6514	0.8996	-0.0181*	0.0250	0.50	0.22
RF148	-0.378	-0.290	-0.4241	0.2912	-0.1928	0.1324	-0.0054	0.0037	0.98	0.29
RF149	0.279	0.040	0.2322	0.0487	0.1056	0.0222	0.0029	0.0006	95.97	2.92

Sample	COC#	ID		vol	DET	eff	Background ^{243}**Am** Cpm	+/-	^{241}**Am** cpm	+/-	count time(s)	(min)
TOTAL	TAMU050800	RF150	Am	12	10	0.240	0.0028	0.0006	0.0006	0.0003	86160	1436.0
0.5 -20µm	TAMU050800	RF152	Am	90	2	0.160	0.0019	0.0007	0.0002	0.0002	244188	4069.8
0.5 -20µm	TAMU050800	RF152B	Am	90	3	0.200	0.0017	0.0006	0.0007	0.0004	244188	4069.8
<0.5µm	TAMU050800	RF153	Am	90	11	0.190	0.0021	0.0005	0.0004	0.0002	86174	1436.2
100kDa - 0.5µm	TAMU050800	RF154	Am	36	2	0.160	0.0011	0.0005	0.0002	0.0002	445457	7424.3
<100kDa	TAMU050800	RF155	Am	36	3	0.200	0.0011	0.0005	0.0007	0.0004	445457	7424.3
100kDa wash	TAMU050800	RF156	Am	36	4	0.140	0.0094	0.0047	0	0.0000	445458	7424.3
3kDa - 0.5µm	TAMU050800	RF157	Am	36	3	0.200	0.0009	0.0005	0.0007	0.0004	154797	2580.0
<3kDa	TAMU050800	RF158	Am	36	4	0.140	0.0012	0.0005	0	0.0000	154798	2580.0
3kDa wash	TAMU050800	RF159	Am	36	5	0.240	0.0007	0.0007	0.0014	0.0010	581251	9687.5
TOTAL	TAMU050800	RF160	Am	12	4	0.140	0.0014	0.0006	0	0.0000	244190	4069.8
>20µm	TAMU050800	RF161	Am	90	7	0.190	0.0007	0.0005	0.0004	0.0004	71002	1183.4
0.5 -20µm	TAMU050800	RF162	Am	90	2	0.160	0.0033	0.0011	0.0007	0.0005	41299	688.3
0.5 -20µm	TAMU050800	RF162B	Am	90	4	0.140	0.0004	0.0004	0.0004	0.0004	41339	689.0
<0.5µm	TAMU050800	RF163	Am	18	6	0.210	0.0028	0.0014	0.0028	0.0014	581251	9687.5
100kDa - 0.5µm	TAMU050800	RF164	Am	36	7	0.190	0.0056	0.0020	0.0014	0.0010	581243	9687.4
<100kDa	TAMU050800	RF165	Am	36	8	0.210	0.0063	0.0021	0	0.0000	581242	9687.4
100kDa wash	TAMU050800	RF166	Am	36	9	0.220	0.0021	0.0012	0.0007	0.0007	581225	9687.1
3kDa - 0.5µm	TAMU050800	RF167	Am	36	10	0.210	0.0035	0.0016	0.0007	0.0007	581225	9687.1
<3kDa	TAMU050800	RF168	Am	36	11	0.190	0.0023	0.0007	0.0002	0.0002	581226	9687.1
3kDa wash	TAMU050800	RF169	Am	36	12	0.2	0.0031	0.0009	0.0005	0.0004	581226	9687.1

ID	^{243}Am Counts	+ / -	Bkg Corrected ^{243}Am Counts	+ / -	^{241}Am Counts	+ / -	Bkg Corrected ^{241}Am Counts	+ / -	^{243}Am Cpm	+ / -	^{241}Am Cpm	+ / -	^{243}Am Dpm Added	+ / -
RF150	1220	34.99	1215.98	35.00	309	17.28	308.14	17.29	0.85	0.02	0.21	0.01	6.45	0.050
RF152	2440	49.38	2432.27	49.46	9270	96.28	9269.19	96.28	0.60	0.01	2.28	0.02	6.45	0.050
RF152B	6370	79.82	6363.08	79.86	1760	41.98	1757.15	42.01	1.56	0.02	0.43	0.01	6.45	0.050
RF153	372	19.29	368.98	19.31	23	4.8	22.43	4.81	0.26	0.01	0.02	0.00	6.45	0.050
RF154	2160	46.45	2151.83	46.59	95	9.75	93.52	9.86	0.29	0.01	0.01	0.00	6.45	0.050
RF155	2950	54.35	2941.83	54.47	108	10.39	102.80	10.81	0.40	0.01	0.01	0.00	6.45	0.050
RF156	1170	34.19	1100.21	48.85	50	7.07	50.00	7.07	0.15	0.01	0.01	0.00	6.45	0.050
RF157	748	27.35	745.68	27.37	56	7.48	54.19	7.55	0.29	0.01	0.02	0.00	6.45	0.050
RF158	921	30.35	917.90	30.38	27	5.2	27.00	5.20	0.36	0.01	0.01	0.00	6.45	0.050
RF159	4260	65.3	4253.22	65.65	89	9.43	75.44	13.45	0.44	0.01	0.01	0.00	6.45	0.050
RF160	2580	50.77	2574.30	50.82	828	28.77	828.00	28.77	0.63	0.01	0.20	0.01	6.45	0.050
RF161	743	27.26	742.17	27.27	2110	45.95	2109.53	45.95	0.63	0.02	1.78	0.04	6.45	0.050
RF162	577	24.02	574.73	24.03	670	25.88	669.52	25.88	0.83	0.03	0.97	0.04	6.45	0.050
RF162B	795	28.2	794.72	28.20	76	8.72	75.72	8.72	1.15	0.04	0.11	0.01	6.45	0.050
RF163	9770	98.86	9742.87	99.79	650	25.5	622.87	28.88	1.01	0.01	0.06	0.00	6.45	0.050
RF164	9310	96.49	9255.75	98.38	550	23.45	536.44	25.34	0.96	0.01	0.06	0.00	6.45	0.050
RF165	10900	104.31	10838.97	106.27	472	21.73	472.00	21.73	1.12	0.01	0.05	0.00	6.45	0.050
RF166	10900	104.57	10879.66	105.23	349	18.68	342.22	19.87	1.12	0.01	0.04	0.00	6.45	0.050
RF167	13000	113.83	12966.10	114.84	894	29.9	887.22	30.66	1.34	0.01	0.09	0.00	6.45	0.050
RF168	8090	89.97	8067.72	90.25	119	10.91	117.06	11.08	0.83	0.01	0.01	0.00	6.45	0.050
RF169	11700	108.12	11669.97	108.44	206	14.35	201.16	14.75	1.20	0.01	0.02	0.00	6.45	0.050

ID	^{241}Am dpm	+ / -	Blank Corrected ^{241}Am	+ / -	^{241}Am pCi	+ / -	^{241}Am PCi/L	+ / -	Tracer recovery	+ / -
RF150	1.634	0.103	1.5880	0.1067	0.7218	0.0485	0.0602	0.0040	54.70	1.63
RF152	24.580	0.561	24.5340	0.5619	11.1518	0.2554	0.1239	0.0028	57.91	1.26
RF152B	1.781	0.048	1.7347	0.0554	0.7885	0.0252	0.0088	0.0003	121.20	1.79
RF153	0.392	0.087	0.3456	0.0908	0.1571	0.0413	0.0017	0.0005	20.96	1.11
RF154	0.280	0.030	0.2339	0.0408	0.1063	0.0186	0.0030	0.0005	28.08	0.65
RF155	0.225	0.024	0.1790	0.0365	0.0813	0.0166	0.0023	0.0005	30.72	0.62
RF156	0.293	0.043	0.2467	0.0514	0.1121	0.0234	0.0031	0.0006	16.41	0.74
RF157	0.469	0.068	0.4223	0.0729	0.1920	0.0332	0.0053*	0.0009	22.41	0.84
RF158	0.190	0.037	0.1433	0.0462	0.0651	0.0210	0.0018	0.0006	39.40	1.34
RF159	0.114	0.020	0.0680	0.0343	0.0309	0.0156	0.0009	0.0004	28.36	0.49
RF160	2.075	0.083	2.0281	0.0873	0.9219	0.0397	0.0768	0.0033	70.05	1.49
RF161	18.333	0.783	18.2868	0.7835	8.3122	0.3561	0.0924	0.0040	51.18	1.92
RF162	7.514	0.428	7.4674	0.4288	3.3943	0.1949	0.0377	0.0022	80.91	3.44
RF162B	0.615	0.074	0.5681	0.0790	0.2582	0.0359	0.0029	0.0004	127.74	4.64
RF163	0.412	0.020	0.3659	0.0337	0.1663	0.0153	0.0092	0.0009	74.25	0.96
RF164	0.374	0.018	0.3274	0.0329	0.1488	0.0150	0.0041	0.0004	77.96	1.03
RF165	0.281	0.013	0.2344	0.0305	0.1066	0.0139	0.0030	0.0004	82.60	1.03
RF166	0.203	0.012	0.1564	0.0300	0.0711	0.0136	0.0020	0.0004	79.15	0.98
RF167	0.441	0.016	0.3949	0.0317	0.1795	0.0144	0.0050*	0.0004	98.82	1.17
RF168	0.094	0.009	0.0471	0.0289	0.0214	0.0131	0.0006	0.0004	67.96	0.93
RF169	0.111	0.008	0.0647	0.0287	0.0294	0.0130	0.0008	0.0004	93.39	1.13

Sample	Spike Added Dpm	ID		vol	DET	eff	Background ^{243}Am Cpm	+ / -	^{241}Am cpm	+ / -	count time(s)	(min)
Blank		RF170	Am	1	5	0.24	0.0007	0.0007	0.0014	0.0010	235664	3927.7
Blank Spike	4.94	RF171	Am	1	6	0.21	0.0021	0.0012	0.0028	0.0014	235665	3927.8
Blank		RF172	Am	1	7	0.19	0.0042	0.0017	0.0014	0.0010	235666	3927.8
Blank Spike	5.11	RF173	Am	1	8	0.21	0.0042	0.0017	0	0.0000	235666	3927.8
Blank		RF174	Am	1	9	0.22	0.0028	0.0014	0	0.0000	235663	3927.7
Blank Spike	5.07	RF175	Am	1	10	0.21	0.0028	0.0014	0.0007	0.0007	235662	3927.7
Blank		RF176	Am	1	11	0.19	0.0021	0.0007	0.0005	0.0004	235664	3927.7
Blank Spike	4.91	RF177	Am	1	12	0.2	0.0021	0.0007	0.0005	0.0004	235663	3927.7

ID	^{243}Am Counts	+ / -	Bkg Corrected ^{243}Am Counts	+ / -	^{241}Am Counts	+ / -	Bkg Corrected ^{241}Am Counts	+ / -	^{243}Am Cpm	+ / -	^{241}Am Cpm	+ / -	^{243}Am Dpm Added	+ / -
RF170	3900	62.45	3897.25	62.51	36	6	30.50	7.15	0.99	0.02	0.01	0.00	6.45	0.050
RF171	3010	54.87	3001.75	55.08	2100	45.79	2089.00	46.12	0.76	0.01	0.53	0.01	6.45	0.050
RF172	2010	44.79	1993.50	45.29	24	4.9	18.50	6.26	0.51	0.01	0.00	0.00	6.45	0.050
RF173	1830	42.81	1813.50	43.34	1230	35.07	1230.00	35.07	0.46	0.01	0.31	0.01	6.45	0.050
RF174	2870	53.58	2859.00	53.86	19	4.36	19.00	4.36	0.73	0.01	0.00	0.00	6.45	0.050
RF175	3880	62.26	3869.00	62.50	2690	51.85	2687.25	51.92	0.99	0.02	0.68	0.01	6.45	0.050
RF176	2590	50.87	2581.75	50.94	15	3.87	13.04	4.11	0.66	0.01	0.00	0.00	6.45	0.050
RF177	3840	61.95	3831.75	62.01	2620	51.22	2618.04	51.24	0.98	0.02	0.67	0.01	6.45	0.050

ID	^{241}Am dpm	+ / -	Blank Corrected ^{241}Am	+ / -	^{241}Am pCi	+ / -	^{241}Am PCi/L	+ / -	Tracer recovery	+ / -	Spike recovery	+ / -
RF170	0.050	0.012	0.0040	0.0299	0.0018	0.0136	0.0018	0.0136	64.10	1.14		
RF171	4.489	0.129	4.4423	0.1318	2.0192	0.0599	2.0192	0.0599	56.42	1.12	88.80	2.78
RF172	0.060	0.020	0.0134	0.0342	0.0061	0.0155	0.0061	0.0155	41.42	0.99		
RF173	4.375	0.163	4.3282	0.1651	1.9674	0.0750	1.9674	0.0750	34.09	0.86	81.94	3.23
RF174	0.043	0.010	-0.0036	0.0292	-0.0016	0.0133	-0.0016	0.0133	51.30	1.05		
RF175	4.480	0.113	4.4335	0.1161	2.0152	0.0528	2.0152	0.0528	72.72	1.30	85.64	2.40
RF176	0.033	0.010	-0.0139	0.0293	-0.0063	0.0133	-0.0063	0.0133	53.64	1.14		
RF177	4.407	0.112	4.3605	0.1152	1.9820	0.0524	1.9820	0.0524	75.63	1.36	87.40	2.47

Sample		ID		Wt.	DET	eff	Background ^{242}Pu Cpm	+/-	$^{239,240}Pu$ cpm	+/-	count time(s)	(min)
RF99A3372	<1mm	P130	Pu	0.51	10	0.21	0	0	0	0	57977	966.28
RF99A3372	<1mm	P142	Pu	0.52	7	0.19	0	0	0	0	86807	1446.8
RF99A3372	<1mm	P328	Pu	0.1	5	0.24	0.0011	0.0006	0.0004	0.0004	3347	55.783
RF99A3372	<1mm	P329	Pu	0.1	6	0.21	0.0022	0.0009	0.0018	0.0008	3347	55.783
RF99A3372	<1mm	P334	Pu	0.22	5	0.24	0.0011	0.0006	0.0004	0.0004	6759	112.65
RF99A3372	<1mm	P335	Pu	0.17	6	0.21	0.0022	0.0009	0.0022	0.0009	6755	112.58

ID	^{242}Pu Counts	+/-	Bkg Corrected ^{242}Pu Counts	+/-	$^{239,240}Pu$ Counts	+/-	Bkg Corrected $^{239,240}Pu$ Counts	+/-	^{242}Pu Cpm	+/-	$^{239,240}Pu$ Cpm	+/-	^{242}Pu Dpm Added	+/-
P130	3250	57.01	3250	57.01	177000	421.2	177000	421.17	3.3634	0.0590	183.1761	0.4359	19.92	0.1474
P142	1970	44.33	1970	44.33	133000	188.5	133000	188.53	1.3616	0.0306	91.9281	0.1303	9.96	0.0737
P328	384	19.6	383.94	19.6	2600	50.96	2599.978	50.960	6.8827	0.3514	46.6085	0.9135	23.50	0.1739
P329	451	21.24	450.88	21.24	2360	48.56	2359.9	48.560	8.0827	0.3808	42.3047	0.8705	23.50	0.1739
P334	797	28.23	796.88	28.23	13100	114.4	13099.95	114.41	7.0739	0.2506	116.2890	1.0156	23.50	0.1739
P335	686	26.19	685.75	26.19	7350	85.71	7349.752	85.71	6.0911	0.2326	65.2828	0.7613	23.50	0.1739

ID	$^{239,240}Pu$ dpm	+/-	$^{239,240}Pu$ dpm/g	+/-	$^{239,240}Pu$ pCi/g	+/-	Tracer recovery	+/-
P130	1084.874	19.205	2125.537	37.627	966.153	17.103	80.40	1.53
P142	672.426	15.161	1293.128	29.156	587.785	13.253	71.95	1.70
P328	159.139	8.702	1620.556	88.617	736.617	40.281	122.03	6.29
P329	122.999	6.323	1290.655	66.348	586.661	30.158	163.78	7.81
P334	386.320	14.096	1740.963	63.522	791.347	28.874	125.42	4.54
P335	251.868	10.058	1518.193	60.625	690.088	27.557	123.43	4.80

Sample	ID	DOC ppm	+/-	Sed Wt. (g)	DET	eff	Background ^{242}Pu Cpm	+/-	^{239,240}Pu cpm	+/-	count time(s)	(min)
Control 0.5g/20ml 4d	P369	7.2	0.1	0.514	5	0.24	0.0018	0.0005	0.0008	0.0003	84840	1414
Control 0.5g/20ml 4d	P370	6.92	0.09	0.499	6	0.21	0.007	0.001	0.0011	0.0003	84915	1415.3
Control 0.5g/20ml 4d	P336	9.02	0.02	0.477	5	0.24	0.0004	0.0004	0.0004	0.0004	46112	768.53
Control 0.5g/20ml 4d	P337	10.2	0.02	0.492	6	0.21	0.0026	0.001	0.0022	0.0009	46114	768.57
Control 0.5g/100ml 4d	P319	3.51	0.03	0.503	7	0.19	0.003	0.0011	0.0077	0.0017	53981	899.68
Control 0.5g/100ml 4d	P321	3.3	0.05	0.507	8	0.21	0.0063	0.0015	0.0085	0.0018	53982	899.7
Control 190ml/0.5g	P265	7.75	0.06	0.505	2	0.16	0.0022	0.0009	0.0029	0.001	73282	1221.4
Control 190ml/0.5g	P267	7.29	0.04	0.505	4	0.14	0.0011	0.0006	0.0004	0.0004	73296	1221.6
Control 400ml/0.1g	P279	6.74	0.08	0.102	5	0.24	0.0004	0.0004	0.0004	0.0004	41853	697.55
Control 400ml/0.1g	P281	5.09	0.06	0.101	6	0.21	0.0018	0.0008	0.0011	0.0006	41856	697.6
Control 0.05g/2L 4d	P315	2.26	0.04	0.051	5	0.24	0.0011	0.0006	0.0004	0.0004	53979	899.65
Control 0.05g/2L 4d	P317	3.08	0.01	0.052	6	0.21	0.0022	0.0009	0.0018	0.0008	53980	899.67
Control 190ml/0.5g 1d	P291	2.74	0.01	0.495	11	0.19	0.0007	0.0005	0.0011	0.0006	41962	699.37
Control 190ml/0.5g 1d	P293	2.64	0.06	0.493	12	0.2	0.0018	0.0008	0.0022	0.0009	41977	699.62
Control 190ml/0.5g 3d	P295	2.52	0.02	0.500	9	0.22	0.0018	0.0008	0.0011	0.0006	55931	932.18
Control 190ml/0.5g 3d	P297	2.47	0.02	0.500	12	0.2	0.0018	0.0008	0.0026	0.001	53421	890.35
Control 190ml/0.5g 5d	P299	2.41	0.03	0.505	5	0.24	0.0004	0.0004	0.0004	0.0004	55895	931.58
Control 190ml/0.5g 5d	P301	2.34	0.01	0.500	6	0.21	0.0018	0.0008	0.0018	0.0008	55897	931.62
Control 190ml/0.5g 7d	P303	2.69	0.03	0.504	7	0.19	0.0022	0.0009	0.0073	0.0016	55897	931.62
Control 190ml/0.5g 7d	P305	2.48	0.05	0.499	8	0.21	0.0041	0.0012	0.0077	0.0017	55923	932.05

ID	^{242}Pu Counts	+ / -	Bkg Corrected ^{242}Pu Counts	+ / -	$^{239,240}Pu$ Counts	+ / -	Bkg Corrected $^{239,240}Pu$ Counts	+ / -	^{242}Pu Cpm	+ / -	$^{239,240}Pu$ Cpm	+ / -	^{242}Pu Dpm Added	+ / -
P369	1630	40.41	1627.5	40.416	64	8	62.8688	8.013	1.1510	0.0286	0.0445	0.0057	4.70	0.035
P370	1100	33.17	1090.1	33.2	37	6.08	35.44323	6.091	0.7702	0.0235	0.0250	0.0043	4.70	0.035
P336	412	20.3	411.69	20.302	120	10.95	119.6926	10.95	0.5357	0.0264	0.1557	0.0143	2.35	0.017
P337	431	20.76	429	20.774	51	7.14	49.30915	7.173	0.5582	0.0270	0.0642	0.0093	2.35	0.017
P319	442	21.02	439.3	21.042	167	12.92	160.0724	13.01	0.4883	0.0234	0.1779	0.0145	2.36	0.017
P321	509	22.56	503.33	22.602	268	16.37	260.3526	16.45	0.5594	0.0251	0.2894	0.0183	2.36	0.017
P265	825	28.72	822.31	28.741	288	16.97	284.458	17.02	0.6733	0.0235	0.2329	0.0139	4.72	0.035
P267	503	22.43	501.66	22.443	199	14.11	198.5114	14.12	0.4107	0.0184	0.1625	0.0116	4.72	0.035
P279	383	19.57	382.72	19.572	175	13.23	174.721	13.23	0.5487	0.0281	0.2505	0.0190	2.36	0.017
P281	392	19.8	390.74	19.808	216	14.7	215.2326	14.71	0.5601	0.0284	0.3085	0.0211	2.36	0.017
P315	415	20.37	414.01	20.378	75	8.66	74.64014	8.67	0.4602	0.0227	0.0830	0.0096	2.36	0.017
P317	442	21.02	440.02	21.036	168	12.96	166.3806	12.98	0.4891	0.0234	0.1849	0.0144	2.36	0.017
P291	396	19.9	395.51	19.903	166	12.88	165.2307	12.89	0.5655	0.0285	0.2363	0.0184	2.36	0.017
P293	357	18.89	355.74	18.898	159	12.61	157.4608	12.63	0.5085	0.0270	0.2251	0.0180	2.36	0.017
P295	426	20.64	424.32	20.654	228	15.1	226.9746	15.11	0.4552	0.0222	0.2435	0.0162	2.36	0.017
P297	438	20.93	436.4	20.942	287	16.94	284.6851	16.96	0.4901	0.0235	0.3197	0.0191	2.36	0.017
P299	601	24.52	600.63	24.523	379	19.47	378.6274	19.47	0.6447	0.0263	0.4064	0.0209	2.36	0.017
P301	454	21.31	452.32	21.323	273	16.52	271.3231	16.54	0.4855	0.0229	0.2912	0.0178	2.36	0.017
P303	451	21.24	448.95	21.256	280	16.73	273.1992	16.80	0.4819	0.0228	0.2933	0.0180	2.36	0.017
P305	466	21.59	462.18	21.621	325	18.03	317.8232	18.08	0.4959	0.0232	0.3410	0.0194	2.36	0.017

ID	^{239,240}Pu dpm	+/-	Blank Corrected ^{239,240}Pu dpm	+/-	^{239,240}Pu pCi	+/-	^{239,240}Pu PCi/L	+/-	%of Activity Resuspended	+/-	Tracer recovery	+/-
P369	0.182	0.024	0.363	0.047	18.156	2.358	8.253	1.072	0.0434	0.0065	102.04	2.64
P370	0.153	0.027	0.306	0.053	15.282	2.667	6.946	1.212	0.0376	0.0072	78.04	2.45
P336	0.683	0.071	1.366	0.142	68.322	7.103	31.056	3.229	0.1759	0.0227	94.98	4.74
P337	0.270	0.041	0.540	0.083	27.011	4.141	12.278	1.882	0.0673	0.0115	113.11	5.54
P319	0.860	0.081	0.955	0.090	9.555	0.901	4.343	0.410	0.1165	0.0141	108.89	5.28
P321	1.221	0.095	1.356	0.105	13.564	1.051	6.165	0.478	0.1641	0.0178	112.88	5.14
P265	1.633	0.113	1.723	0.119	9.071	0.628	4.123	0.286	0.2095	0.0216	89.15	3.19
P267	1.868	0.157	1.972	0.166	10.376	0.872	4.717	0.396	0.2396	0.0272	62.15	2.82
P279	1.077	0.098	1.105	0.101	2.763	0.252	1.256	0.115	0.6658	0.0792	96.87	5.01
P281	1.300	0.111	1.333	0.113	3.333	0.284	1.515	0.129	0.8089	0.0924	113.02	5.79
P315	0.425	0.054	0.428	0.054	0.214	0.027	0.09718	0.01226	0.5155	0.0760	81.25	4.04
P317	0.892	0.082	0.897	0.082	0.448	0.041	0.20383	0.01865	1.0604	0.1262	98.69	4.77
P291	0.986	0.092	1.041	0.097	5.477	0.508	2.490	0.231	0.1291	0.0155	126.12	6.41
P293	1.045	0.100	1.103	0.106	5.803	0.558	2.638	0.254	0.1371	0.0168	107.73	5.78
P295	1.262	0.104	1.333	0.110	7.013	0.578	3.188	0.263	0.1637	0.0184	87.67	4.32
P297	1.540	0.118	1.625	0.124	8.553	0.654	3.888	0.297	0.1995	0.0215	103.84	5.04
P299	1.488	0.098	1.570	0.103	8.265	0.543	3.757	0.247	0.1908	0.0192	113.83	4.72
P301	1.416	0.109	1.494	0.115	7.865	0.606	3.575	0.275	0.1835	0.0199	97.97	4.67
P303	1.436	0.111	1.516	0.118	7.978	0.619	3.627	0.281	0.1846	0.0201	107.47	5.15
P305	1.623	0.120	1.713	0.126	9.016	0.664	4.098	0.302	0.2108	0.0223	100.06	4.74

Sample	ID	Conc (mg/L)	Sed Wt. (g)	DET	eff	Background ^{242}Pu Cpm	+/-	$^{239,240}Pu$ cpm	+/-	count time(s)	(min)
Humic Acid 3d 190ml/0.5g	P233	15	0.502	5	0.24	0.0028	0.0014	0	0	101236	1687.3
Humic Acid 3d 190ml/0.5g	P235	15	0.505	6	0.21	0.0035	0.0016	0.0028	0.0014	101236	1687.3
Humic Acid 3d 190ml/0.5g	P237	30	0.497	7	0.19	0.0028	0.0014	0.0056	0.002	101237	1687.3
Humic Acid 3d 190ml/0.5g	P239	30	0.501	8	0.21	0.0014	0.0007	0	0	101237	1687.3
Humic Acid 3d 190ml/0.5g	P241	60	0.500	9	0.22	0.0028	0.0014	0.0021	0.0012	101236	1687.3
Humic Acid 3d 190ml/0.5g	P243	60	0.507	10	0.21	0.0028	0.0014	0.0021	0.0012	101236	1687.3
Humic Acid 3d 190ml/0.5g	P269	79	0.497	6	0.21	0.0018	0.0008	0.0018	0.0008	77361	1289.4
Humic Acid 3d 190ml/0.5g	P271	79	0.497	8	0.21	0.0033	0.0025	0.0029	0.0026	77364	1289.4
Humic Acid 3d 190ml/0.5g	P273	99	0.505	10	0.21	0.0015	0.0008	0.0048	0.0013	77364	1289.4
Humic Acid 3d 190ml/0.5g	P275	99	0.509	12	0.2	0.0026	0.001	0.0026	0.001	77365	1289.4
Humic Acid 3d 190ml/0.5g	P245	120	0.507	11	0.19	0.0014	0.0014	0.0006	0.0019	101236	35797
Humic Acid 3d 190ml/0.5g	P247	120	0.500	12	0.2	0.004	0.001	0.0026	0.0008	101236	1687.3
Humic Acid 400ml/0.1g	P283	60	0.100	7	0.19	0.0022	0.0009	0.0074	0.0017	41875	697.92
Humic Acid 400ml/0.1g	P285	60	0.102	8	0.21	0.0044	0.0013	0.0077	0.0017	41896	698.27
Humic Acid 190ml/0.5g 1d	P307	59	0.497	9	0.22	0.0007	0.0007	0.0007	0.0007	53458	890.97
Humic Acid 190ml/0.5g 1d	P309	59	0.493	10	0.21	0.0011	0.0006	0.0041	0.0012	53471	891.18
Humic Acid 190ml/0.5g 3d	P311	59	0.501	11	0.19	0.0007	0.0005	0.0018	0.0008	53472	891.2
Humic Acid 190ml/0.5g 3d	P313	59	0.505	12	0.2	0.0022	0.0009	0.0026	0.001	53472	891.2
Humic Acid 190ml/0.5g 5d	P323	59	0.497	9	0.22	0.0018	0.0008	0.0011	0.0006	53981	899.68
Humic Acid 190ml/0.5g 5d	P325	59	0.502	10	0.21	0.0015	0.0008	0.0048	0.0013	53981	899.68
Humic Acid 190ml/0.5g 7d	P331	59	0.501	7	0.19	0.0029	0.001	0.0077	0.0017	28933	482.22
Humic Acid 190ml/0.5g 7d	P333	59	0.500	4	0.14	0.0015	0.0008	0.0011	0.0006	53973	899.55

ID	^{242}Pu Counts	+/-	Bkg Corrected ^{242}Pu Counts	+/-	$^{239,240}Pu$ Counts	+/-	Bkg Corrected $^{239,240}Pu$ Counts	+/-	^{242}Pu Cpm	+/-	$^{239,240}Pu$ Cpm	+/-	^{242}Pu Dpm Added	+/-
P233	1830	42.76	1825.3	42.825	603	24.56	603	24.560	1.0818	0.0254	0.3574	0.0146	4.72	0.0349
P235	1520	38.95	1514.1	39.039	521	22.83	516.2757	22.952	0.8974	0.0231	0.3060	0.0136	4.72	0.0349
P237	914	310.23	909.28	310.24	226	15.03	216.5512	15.397	0.5389	0.1839	0.1283	0.0091	4.72	0.0349
P239	2020	44.96	2017.6	44.976	647	25.44	647	25.440	1.1958	0.0267	0.3835	0.0151	4.72	0.0349
P241	1820	42.67	1815.3	42.735	565	23.77	561.4567	23.858	1.0759	0.0253	0.3328	0.0141	4.72	0.0349
P243	1910	43.75	1905.3	43.814	574	23.96	570.4567	24.047	1.1292	0.0260	0.3381	0.0143	4.72	0.0349
P269	929	30.48	926.68	30.498	596	24.41	593.6792	24.432	0.7187	0.0237	0.4604	0.0189	4.72	0.0349
P271	1080	32.92	1075.7	33.082	607	24.64	603.2607	24.860	0.8343	0.0257	0.4679	0.0193	4.72	0.0349
P273	1510	38.88	1508.1	38.892	841	29	834.8109	29.051	1.1696	0.0302	0.6474	0.0225	4.72	0.0349
P275	1880	43.32	1876.6	43.339	964	31.05	960.6475	31.076	1.4554	0.0336	0.7450	0.0241	4.72	0.0349
P245	1700	41.28	1649.9	64.928	2390	48.23	2369.543	83.379	0.0461	0.0018	0.0662	0.0023	4.72	0.0349
P247	1840	42.84	1833.3	42.871	2040	45.19	2035.613	45.209	1.0865	0.0254	1.2065	0.0268	4.72	0.0349
P283	389	19.72	387.46	19.73	152	12.33	146.8354	12.384	0.5552	0.0283	0.2104	0.0177	2.36	0.0175
P285	323	17.97	319.93	17.992	134	11.58	128.6233	11.639	0.4582	0.0258	0.1842	0.0167	2.36	0.0175
P307	479	21.89	478.38	21.899	335	18.3	334.3763	18.311	0.5369	0.0246	0.3753	0.0206	2.36	0.0175
P309	533	23.09	532.02	23.097	416	20.4	412.3461	20.430	0.5970	0.0259	0.4627	0.0229	2.36	0.0175
P311	489	22.11	488.38	22.114	714	26.72	712.3958	26.730	0.5480	0.0248	0.7994	0.0300	2.36	0.0175
P313	523	22.87	521.04	22.884	757	27.51	754.6829	27.524	0.5846	0.0257	0.8468	0.0309	2.36	0.0175
P323	539	23.22	537.38	23.231	734	27.09	733.0103	27.096	0.5973	0.0258	0.8147	0.0301	2.36	0.0175
P325	490	22.14	488.65	22.15	799	28.27	794.6815	28.295	0.5431	0.0246	0.8833	0.0315	2.36	0.0175
P331	243	15.59	241.6	15.598	521	22.83	517.2869	22.844	0.5010	0.0323	1.0727	0.0474	2.36	0.0175
P333	499	22.34	497.65	22.35	635	25.2	634.0105	25.206	0.5532	0.0248	0.7048	0.0280	2.36	0.0175

ID	^{239,240} Pu dpm	+ / -	Blank Corrected ^{239,240} Pu dpm	+ / -	^{239,240} Pu pCi	+ / -	^{239,240} Pu PCi/L	+ / -	%of Activity Resuspended	+ / -	Tracer recovery	+ / -
P233	1.559	0.073	1.646	0.077	8.663	0.407	3.938	0.185	0.2011	0.0180	95.50	2.35
P235	1.609	0.083	1.699	0.087	8.941	0.460	4.064	0.209	0.2065	0.0190	90.53	2.43
P237	1.124	0.392	1.187	0.414	6.245	2.177	2.839	0.989	0.1465	0.0523	60.09	20.51
P239	1.514	0.068	1.598	0.072	8.409	0.380	3.822	0.173	0.1958	0.0173	120.64	2.83
P241	1.460	0.071	1.541	0.075	8.110	0.394	3.687	0.179	0.1893	0.0171	103.61	2.56
P243	1.413	0.068	1.492	0.072	7.851	0.377	3.569	0.171	0.1804	0.0162	113.92	2.75
P269	3.024	0.159	3.192	0.168	16.799	0.885	7.636	0.402	0.3939	0.0365	72.51	2.45
P271	2.647	0.136	2.794	0.144	14.705	0.756	6.684	0.344	0.3451	0.0317	84.17	2.66
P273	2.613	0.113	2.758	0.119	14.516	0.629	6.598	0.286	0.3354	0.0294	118.00	3.17
P275	2.416	0.096	2.550	0.101	13.423	0.534	6.101	0.243	0.3074	0.0264	154.18	3.74
P245	6.779	0.358	7.155	0.378	37.660	1.988	17.118	0.904	0.8656	0.0802	5.14	0.21
P247	5.241	0.169	5.532	0.178	29.117	0.939	13.235	0.427	0.6790	0.0561	115.10	2.82
P283	0.894	0.088	0.917	0.090	2.293	0.226	1.042	0.103	0.5654	0.0704	123.81	6.37
P285	0.949	0.101	0.973	0.104	2.433	0.259	1.106	0.118	0.5868	0.0768	92.45	5.24
P307	1.650	0.118	1.741	0.124	9.164	0.654	4.166	0.297	0.2151	0.0224	103.41	4.80
P309	1.829	0.120	1.931	0.127	10.162	0.669	4.619	0.304	0.2403	0.0242	120.46	5.30
P311	3.443	0.202	3.634	0.214	19.125	1.125	8.693	0.511	0.4448	0.0428	122.21	5.61
P313	3.418	0.195	3.608	0.206	18.990	1.084	8.632	0.493	0.4381	0.0417	123.87	5.52
P323	3.219	0.183	3.398	0.193	17.884	1.017	8.129	0.462	0.4194	0.0399	115.04	5.05
P325	3.838	0.221	4.051	0.234	21.322	1.229	9.692	0.559	0.4955	0.0473	109.59	5.03
P331	5.053	0.395	5.334	0.417	28.072	2.196	12.760	0.998	0.6540	0.0714	111.74	7.26
P333	3.007	0.180	3.174	0.190	16.704	1.002	7.593	0.455	0.3896	0.0378	167.44	7.62

Sample	ID	Conc (mg/L)	Sed Wt. (g)	DET	eff	Background ^{242}Pu Cpm	+/-	$^{239,240}Pu$ cpm	+/-	count time(s)	(min)
Ext Humic Acid 190ml 0.5g	P338	15	0.478	7	0.19	0.0033	0.0011	0.0085	0.0018	46114	768.57
Ext Humic Acid 190ml 0.5g	P339	15	0.484	8	0.21	0.0055	0.0014	0.0077	0.0017	46113	768.55
Ext Humic Acid 190ml 0.5g	P340	30	0.500	9	0.22	0.0007	0.0005	0.0007	0.0005	46117	768.62
Ext Humic Acid 190ml 0.5g	P341	30	0.516	10	0.21	0.0015	0.0008	0.0044	0.0013	46117	768.62
Ext Humic Acid 190ml 0.5g	P342	60	0.497	11	0.19	0.0007	0.0005	0.0018	0.0008	46117	768.62
Ext Humic Acid 190ml 0.5g	P343	60	0.497	12	0.2	0.0022	0.0009	0.0026	0.001	46118	768.63
Ext Humic Acid 190ml 0.5g	P344	79	0.497	5	0.54	0.0011	0.0006	0.0004	0.0004	56685	944.75
Ext Humic Acid 190ml 0.5g	P345	79	0.497	6	0.21	0.0022	0.0009	0.0022	0.0009	56685	944.75
Ext Humic Acid 190ml 0.5g	P346	99	0.494	7	0.19	0.0022	0.0009	0.0085	0.0018	56686	944.77
Ext Humic Acid 190ml 0.5g	P347	99	0.496	8	0.21	0.0048	0.0013	0.1	0.0193	56686	944.77
Ext Humic Acid 190ml 0.5g	P348	118	0.518	9	0.22	0.0007	0.0005	0.0007	0.0005	56685	944.75
Ext Humic Acid 190ml 0.5g	P349	118	0.515	10	0.21	0.0011	0.0006	0.0041	0.0012	56685	944.75
Alginic Acid3d 190ml/0.5g	P249	15	0.505	5	0.24	0.0035	0.0016	0	0	166976	2782.9
Alginic Acid3d 190ml/0.5g	P251	15	0.503	6	0.21	0.0035	0.0016	0.0028	0.0014	166978	2783
Alginic Acid3d 190ml/0.5g	P253	30	0.505	7	0.19	0.0028	0.0014	0.0056	0.002	166979	2783
Alginic Acid3d 190ml/0.5g	P255	30	0.497	8	0.21	0.0099	0.0026	0.0049	0.0019	166980	2783
Alginic Acid3d 190ml/0.5g	P257	60	0.505	9	0.22	0.0021	0.0012	0.0021	0.0012	166977	2783
Alginic Acid3d 190ml/0.5g	P259	60	0.494	10	0.21	0.0021	0.0012	0.0021	0.0012	166978	2783
Alginic Acid3d 190ml/0.5g	P261	120	0.501	11	0.19	0.0014	0.0006	0.0019	0.0007	166978	2783
Alginic Acid3d 190ml/0.5g	P263	120	0.504	12	0.2	0.0045	0.001	0.0026	0.0008	166978	2783
Alginic Acid 400ml/0.1g	P287	60	0.099	9	0.22	0.0007	0.0005	0.0026	0.001	41914	698.57
Alginic Acid 400ml/0.1g	P289	60	0.100	10	0.19	0.0011	0.0006	0.0026	0.001	41942	699.03

ID	^{242}Pu Counts	+ / -	Bkg Corrected ^{242}Pu Counts	+ / -	$^{239,240}Pu$ Counts	+ / -	Bkg Corrected $^{239,240}Pu$ Counts	+ / -	^{242}Pu Cpm	+ / -	$^{239,240}Pu$ Cpm	+ / -	^{242}Pu Dpm Added	+ / -
P338	384	19.6	381.46	19.618	193	13.89	186.4672	13.957	0.4963	0.0255	0.2426	0.0182	2.35	0.0174
P339	415	20.37	410.77	20.399	212	14.56	206.0822	14.617	0.5345	0.0265	0.2681	0.0190	2.35	0.0174
P340	376	19.39	375.46	19.394	211	14.53	210.462	14.535	0.4885	0.0252	0.2738	0.0189	2.35	0.0174
P341	404	20.1	402.85	20.108	283	16.82	279.6181	16.848	0.5241	0.0262	0.3638	0.0219	2.35	0.0174
P342	415	20.37	414.46	20.374	402	20.05	400.6165	20.060	0.5392	0.0265	0.5212	0.0261	2.35	0.0174
P343	436	20.88	434.31	20.891	486	22.05	484.0016	22.063	0.5650	0.0272	0.6297	0.0287	2.35	0.0174
P344	568	23.83	566.96	23.838	936	30.59	935.6221	30.592	0.6001	0.0252	0.9903	0.0324	2.35	0.0174
P345	455	21.33	452.92	21.347	594	24.37	591.9216	24.385	0.4794	0.0226	0.6265	0.0258	2.35	0.0174
P346	271	16.46	268.92	16.482	338	18.38	329.9695	18.456	0.2846	0.0174	0.3493	0.0195	2.35	0.0174
P347	494	22.23	489.47	22.266	562	23.71	467.5233	29.882	0.5181	0.0236	0.4949	0.0316	2.35	0.0174
P348	501	22.38	500.34	22.385	566	23.79	565.3387	23.795	0.5296	0.0237	0.5984	0.0252	2.35	0.0174
P349	527	22.96	525.96	22.968	625	25	621.1265	25.027	0.5567	0.0243	0.6575	0.0265	2.35	0.0174
P249	2990	54.71	2980.3	54.883	888	29.8	888	29.800	1.0709	0.0197	0.3191	0.0107	4.72	0.0349
P251	2740	52.36	2730.3	52.541	797	28.23	789.2077	28.498	0.9811	0.0189	0.2836	0.0102	4.72	0.0349
P253	2570	50.73	2562.2	50.879	741	27.22	725.4153	27.772	0.9207	0.0183	0.2607	0.0100	4.72	0.0349
P255	3150	56.11	3122.4	56.591	877	29.61	863.3633	30.055	1.1220	0.0203	0.3102	0.0108	4.72	0.0349
P257	3050	55.24	3044.2	55.343	885	29.75	879.1558	29.941	1.0939	0.0199	0.3159	0.0108	4.72	0.0349
P259	2750	52.44	2744.2	52.548	880	29.66	874.1558	29.851	0.9861	0.0189	0.3141	0.0107	4.72	0.0349
P261	2740	52.38	2736.1	52.404	667	25.83	661.7124	25.898	0.9832	0.0188	0.2378	0.0093	4.72	0.0349
P263	1950	44.2	1937.5	44.293	547	23.29	539.7643	23.392	0.6962	0.0159	0.1940	0.0084	4.72	0.0349
P287	374	19.34	373.51	19.343	99	9.95	97.18373	9.974	0.5347	0.0277	0.1391	0.0143	2.36	0.0175
P289	336	18.33	335.23	18.335	183	13.53	181.1825	13.547	0.4796	0.0262	0.2592	0.0194	2.36	0.0175

ID	^{239,240}Pu dpm	+ / -	Blank Corrected ^{239,240}Pu dpm	+ / -	^{239,240}Pu pCi	+ / -	^{239,240}Pu PCi/L	+ / -	%of Activity Resuspended	+ / -	Tracer recovery	+ / -
P338	1.149	0.104	1.213	0.110	6.382	0.580	2.901	0.263	0.1557	0.0184	109.37	5.68
P339	1.179	0.102	1.244	0.108	6.550	0.567	2.977	0.258	0.1578	0.0182	108.30	5.44
P340	1.317	0.114	1.390	0.120	7.318	0.631	3.326	0.287	0.1708	0.0197	94.49	4.93
P341	1.631	0.128	1.722	0.135	9.062	0.709	4.119	0.322	0.2049	0.0224	106.20	5.36
P342	2.271	0.159	2.398	0.168	12.619	0.885	5.736	0.402	0.2963	0.0307	120.77	6.00
P343	2.619	0.174	2.764	0.183	14.549	0.964	6.613	0.438	0.3411	0.0344	120.22	5.85
P344	3.878	0.207	4.094	0.218	21.545	1.148	9.793	0.522	0.5058	0.0470	47.29	2.02
P345	3.071	0.192	3.242	0.203	17.062	1.068	7.756	0.485	0.4000	0.0394	97.14	4.63
P346	2.883	0.239	3.044	0.253	16.019	1.329	7.282	0.604	0.3778	0.0425	63.75	3.94
P347	2.245	0.176	2.369	0.186	12.470	0.978	5.668	0.445	0.2929	0.0320	104.98	4.84
P348	2.655	0.163	2.803	0.172	14.752	0.906	6.705	0.412	0.3319	0.0325	102.44	4.65
P349	2.775	0.165	2.929	0.174	15.418	0.916	7.008	0.416	0.3489	0.0337	112.81	5.00
P249	1.406	0.054	1.485	0.057	7.813	0.299	3.551	0.136	0.1804	0.0154	94.54	1.88
P251	1.364	0.056	1.440	0.059	7.580	0.310	3.445	0.141	0.1758	0.0152	98.98	2.04
P253	1.336	0.058	1.411	0.061	7.424	0.320	3.375	0.146	0.1715	0.0150	102.66	2.18
P255	1.305	0.051	1.378	0.054	7.250	0.285	3.296	0.129	0.1700	0.0146	113.19	2.22
P257	1.363	0.053	1.439	0.056	7.573	0.292	3.442	0.133	0.1749	0.0149	105.34	2.07
P259	1.504	0.059	1.587	0.062	8.353	0.327	3.797	0.149	0.1970	0.0169	99.48	2.04
P261	1.142	0.050	1.205	0.053	6.342	0.276	2.883	0.126	0.1476	0.0130	109.63	2.25
P263	1.315	0.064	1.388	0.068	7.305	0.358	3.321	0.163	0.1689	0.0153	73.75	1.77
P287	0.614	0.071	0.630	0.072	1.574	0.181	0.716	0.082	0.3912	0.0539	102.98	5.39
P289	1.276	0.118	1.308	0.121	3.271	0.303	1.487	0.138	0.8059	0.0966	106.95	5.90

Sample		Size	ID	Conc (mg/L)	DOC ppm	+/-	Sed Wt. (g)	DET	eff	Background ^{242}Pu Cpm	+/-	^{239,240}Pu cpm	+/-	count time(s)	(min
Control 0.5g/190	UF	<0.45	P353	0	3.06	0.01	0.510	5	0.24	0.0015	0.0005	0.0008	0.0003	64733	1078
Control 0.5g/190	UF	0.45-1kDa	P354	0	6.72	0.01	0.510	6	0.21	0.002	0.0005	0.002	0.0005	64733	1078
Control 0.5g/190	UF	<1kDa	P355	0	3.11	0.01	0.510	7	0.19	0.0014	0.0004	0.0033	0.0007	64734	1078
Control 0.5g/190	UF	1kDa filter	P356	0			0.510	5	0.24	0.0018	0.0005	0.0008	0.0003	65124	1085
Control 0.5g/190	UF	<0.45	P357	0			0.508	8	0.21	0.0017	0.0005	0.0022	0.0006	64733	1078
Control 0.5g/190	UF	0.45-1kDa	P358	0	7.08	0.04	0.508	9	0.22	0.0006	0.0003	0.0022	0.0006	64732	1078
Control 0.5g/190	UF	<1kDa	P359	0	3.64	0.03	0.508	10	0.21	0.002	0.0005	0.0021	0.0005	64733	1078
Control 0.5g/190	UF	1kDa filter	P360	0	3.1	0.02	0.508	6	0.21	0.0022	0.0006	0.002	0.0005	65124	1085
Humic Acid 0.5g/190ml	UF	<0.45	P361	59			0.514	11	0.19	0.0022	0.0006	0.002	0.0005	64732	1078
Humic Acid 0.5g/190ml	UF	0.45-1kDa	P362	59			0.514	12	0.2	0.0014	0.0004	0.0021	0.0005	64732	1078
Humic Acid 0.5g/190ml	UF	<1kDa	P363	59			0.514	9	0.22	0.0008	0.0199	0.0022	0.0006	65125	1085
Humic Acid 0.5g/190ml	UF	1kDa filter	P364	59			0.514	7	0.19	0.002	0.0005	0.0035	0.0007	65125	1085
Humic Acid 0.5g/190ml	UF	<0.45	P365	59			0.493	10	0.21	0.002	0.0005	0.0024	0.0006	65126	1085
Humic Acid 0.5g/190ml	UF	0.45-1kDa	P366	59			0.493	11	0.19	0.0021	0.0005	0.0022	0.0006	65126	1085
Humic Acid 0.5g/190ml	UF	<1kDa	P367	59			0.493	12	0.2	0.0014	0.0004	0.0022	0.0006	65128	1085
Humic Acid 0.5g/190ml	UF	1kDa filter	P368	59			0.493	8	0.21	0.0017	0.0005	0.0022	0.0006	65125	1085

ID	^{242}Pu Counts	+/-	Bkg Corrected ^{242}Pu Counts	+/-	^{239,240}Pu Counts	+/-	Bkg Corrected ^{239,240}Pu Counts	+/-	^{242}Pu Cpm	+/-	^{239,240}Pu Cpm	+/-	^{242}Pu Dpm Added	+/-
P353	648	25.46	646.38	25.465	154	12.41	153.1369	12.415	0.5991	0.0236	0.1419	0.0115	2.35	0.0174
P354	532	23.07	529.84	23.077	81	9	78.84223	9.018	0.4911	0.0214	0.0731	0.0084	2.35	0.0174
P355	402	20.05	400.49	20.056	32	5.66	28.43963	5.706	0.3712	0.0186	0.0264	0.0053	2.35	0.0174
P356	562	23.71	560.05	23.716	3	1.73	2.13168	1.766	0.5160	0.0219	0.0020	0.0016	2.35	0.0174
P357	583	24.15	581.17	24.156	274	16.55	271.6265	16.561	0.5387	0.0224	0.2518	0.0153	2.35	0.0174
P358	547	23.39	546.35	23.392	112	10.58	109.6265	10.597	0.5064	0.0217	0.1016	0.0098	2.35	0.0174
P359	618	24.86	615.84	24.867	41	6.4	38.73435	6.427	0.5708	0.0230	0.0359	0.0060	2.35	0.0174
P360	496	22.27	493.61	22.278	8	2.83	5.8292	2.889	0.4548	0.0205	0.0054	0.0027	2.35	0.0174
P361	534	23.11	531.63	23.118	322	17.94	319.8423	17.949	0.4928	0.0214	0.2965	0.0166	2.35	0.0174
P362	562	23.71	560.49	23.715	146	12.08	143.7344	12.094	0.5195	0.0220	0.1332	0.0112	2.35	0.0174
P363	621	24.92	620.13	33.004	9	3	6.612083	3.059	0.5713	0.0304	0.0061	0.0028	2.35	0.0174
P364	1110	33.36	1107.8	33.365	26	5.1	22.20104	5.156	1.0206	0.0307	0.0205	0.0048	4.70	0.0348
P365	603	24.56	600.83	24.567	328	18.11	325.395	18.121	0.5535	0.0226	0.2998	0.0167	2.35	0.0174
P366	552	23.49	549.72	23.497	114	10.68	111.612	10.697	0.5065	0.0216	0.1028	0.0099	2.35	0.0174
P367	514	22.67	512.48	22.675	16	4	13.61197	4.044	0.4721	0.0209	0.0125	0.0037	2.35	0.0174
P368	538	23.19	536.15	23.196	21	4.58	18.61208	4.619	0.4940	0.0214	0.0171	0.0043	4.70	0.0348

ID	^{239,240}Pu dpm	+/-	Blank Corrected ^{239,240}Pu dpm	+/-	^{239,240}Pu pCi	+/-	^{239,240}Pu PCi/L	+/-	%of Activity Resuspended	+/-	Tracer recovery	+/-
P353	0.557	0.050	0.619	0.056	6.186	0.558	2.812	0.253	0.0745	0.0088	106.23	4.26
P354	0.350	0.043	0.525	0.064	5.828	0.713	2.649	0.324	0.0632	0.0091	99.51	4.40
P355	0.167	0.035	0.200	0.041	2.225	0.460	1.011	0.209	0.0241	0.0053	83.14	4.21
P356	0.009	0.007	0.009	0.007	0.009	0.007	0.004	0.003	0.0011	0.0009	91.49	3.93
P357	1.098	0.081	1.220	0.090	12.204	0.901	5.547	0.409	0.1476	0.0156	109.15	4.61
P358	0.472	0.050	0.707	0.075	7.859	0.831	3.572	0.378	0.0855	0.0111	97.95	4.26
P359	0.148	0.025	0.177	0.030	1.971	0.337	0.896	0.153	0.0214	0.0040	115.67	4.75
P360	0.028	0.014	0.028	0.014	0.028	0.014	0.013	0.006	0.0034	0.0017	92.15	4.21
P361	1.414	0.100	1.571	0.112	15.709	1.115	7.141	0.507	0.1875	0.0195	110.36	4.87
P362	0.603	0.057	0.904	0.085	10.044	0.946	4.565	0.430	0.1079	0.0131	110.54	4.75
P363	0.025	0.012	0.030	0.014	0.334	0.156	0.152	0.071	0.0036	0.0017	110.51	5.94
P364	0.094	0.022	0.094	0.022	0.094	0.022	0.043	0.010	0.0112	0.0028	114.29	3.54
P365	1.273	0.088	1.414	0.098	14.141	0.977	6.428	0.444	0.1760	0.0181	112.17	4.66
P366	0.477	0.050	0.954	0.100	10.603	1.113	4.819	0.506	0.1188	0.0154	113.43	4.92
P367	0.062	0.019	0.073	0.022	0.809	0.243	0.368	0.110	0.0091	0.0028	100.45	4.51
P368	0.163	0.041	0.163	0.041	0.163	0.041	0.074	0.019	0.0203	0.0053	50.05	2.20