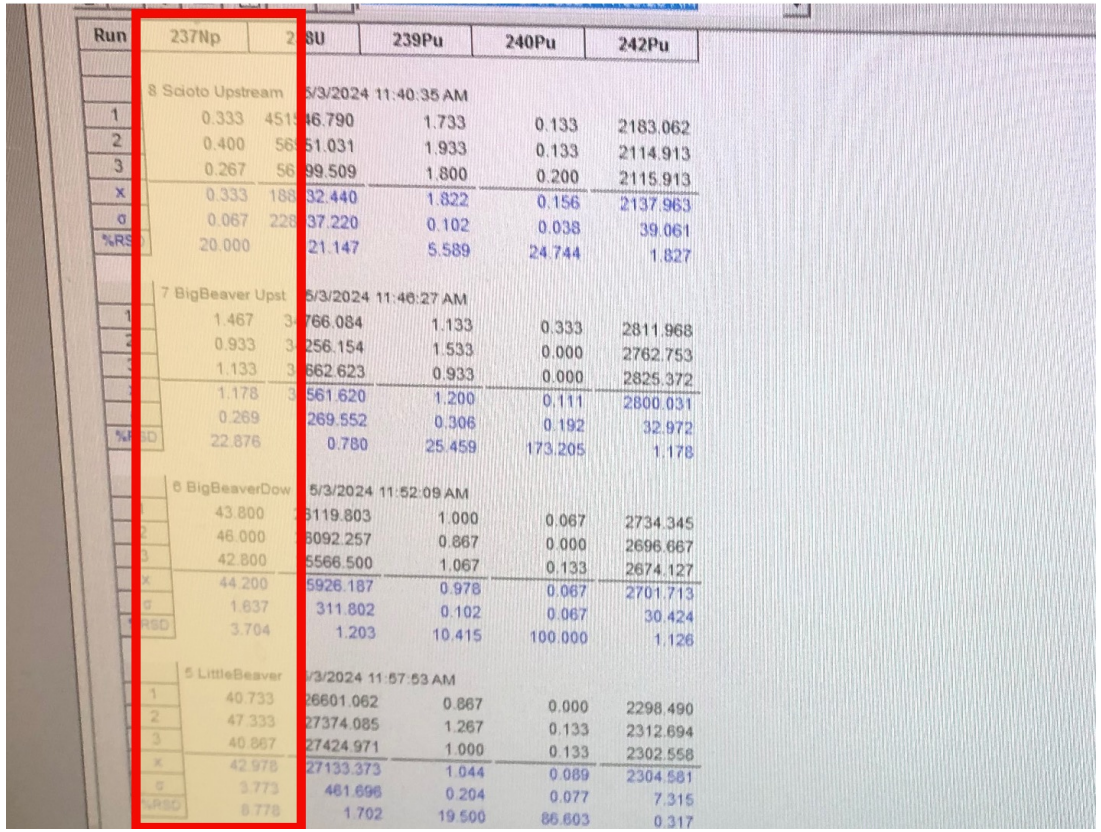
Plant uptake of neptunium-237 in the riparian zone at the Little Beaver Creek – Big Beaver Creek confluence area near the former Portsmouth Gaseous Diffusion Plant



Run	237Np	238U	239Pu	240Pu	242Pu
8 Scioto Upstream		5/3/2024 11:40:35 AM			
1	0.333	46.790	1.733	0.133	2183.062
2	0.400	51.031	1.933	0.133	2114.913
3	0.267	99.509	1.800	0.200	2115.913
x	0.333	82.440	1.822	0.156	2137.963
σ	0.067	37.220	0.102	0.038	39.061
%RSD	20.000	21.147	5.589	24.744	1.827
7 BigBeaver Upst		5/3/2024 11:46:27 AM			
1	1.467	766.084	1.133	0.333	2811.968
2	0.933	256.154	1.533	0.000	2762.753
3	1.133	662.623	0.933	0.000	2825.372
x	1.178	561.620	1.200	0.111	2800.031
σ	0.269	269.552	0.306	0.192	32.972
%RSD	22.876	0.780	25.459	173.205	1.178
6 BigBeaverDow		5/3/2024 11:52:09 AM			
1	43.800	3119.803	1.000	0.067	2734.345
2	46.000	3092.257	0.867	0.000	2696.667
3	42.800	5566.500	1.067	0.133	2674.127
x	44.200	5926.187	0.978	0.067	2701.713
σ	1.637	311.802	0.102	0.067	30.424
%RSD	3.704	1.203	10.415	100.000	1.126
5 LittleBeaver		5/3/2024 11:57:53 AM			
1	40.733	26601.062	0.867	0.000	2298.490
2	47.333	27374.085	1.267	0.133	2312.694
3	40.867	27424.971	1.000	0.133	2302.558
x	42.978	27133.373	1.044	0.089	2304.581
σ	3.773	461.696	0.204	0.077	7.315
%RSD	8.778	1.702	19.500	86.603	0.317

Michael E. Ketterer, PhD

Professor Emeritus, Chemistry and Biochemistry, Northern Arizona University

Flagstaff, AZ 86911-5698 USA Michael.Ketterer@nau.edu

NAU NORTHERN ARIZONA UNIVERSITY

Executive Summary. We investigated, and have confirmed, a significant extent of plant uptake of neptunium-237, a 2.1 million year half-life synthetic radioisotope attributed to the Portsmouth Gaseous Diffusion Plant (PGDP). We studied vegetation growing in the riparian zone at the confluence of Little Beaver Creek with Big Beaver Creek, located downstream in the drainage of the north end of the former PGDP.

Previous work by the author and community members has demonstrated the widespread presence of PGDP-attributed ^{237}Np in creek water and sediments at the confluence. Unclassified DOE documents dating back to the mid-1970's show that DOE and its contractors were well aware of the presence of ^{237}Np and made unsuccessful efforts to contain soluble neptunyl cation (NpO_2^+) and to prevent its entry into groundwater and into Little Beaver Creek upstream of the riparian study area.

Surface soils, sediments and creek waters were sampled by the author on March 2-3, 2024 in collaboration with Lee Blackburn and Vina Colley; a sample of live/dead standing riparian vegetation was collected on August 31, 2023 by Lee Blackburn and Vina Colley. The samples were chemically processed at Northern Arizona University, and subsequently analyzed by inductively coupled plasma mass spectrometry in an Arizona-licensed radioisotope lab facility to determine the concentrations of neptunium-237 in soil/sediment samples, and in plant ash samples. The results were used to calculate the neptunium-237 concentrations in picograms ^{237}Np per gram for solid samples and in picograms ^{237}Np per liter for the water samples.

The results demonstrate the following:

1. The riparian zone at the confluence of Little Beaver Creek and Big Beaver Creek, downstream of the former uranium enrichment facility, has become contaminated with enriched uranium,^{237}Np, and $^{239+240}$Pu. The contaminant stems from past/present PGDP discharges into soil and water on the northern portion of the DOE reservation, draining into Little Beaver Creek. Riparian zone concentrations of ^{237}Np in soils and sediments ranged from 8 to 21 picograms ^{237}Np per gram. ***Concentrations of neptunium from PGDP in soils/sediments within the confluence zone are approximately one hundred times higher than accountable from ubiquitous 1950's-1960's nuclear weapons test fallout.***

2. The fact that the neptunium originated from PGDP and not fallout, is established by measurements of ^{237}Np/^{239}Pu in sediments of the confluence zone isotope compositions in riparian zone sediments. The Np-Pu isotope signatures in the confluence sediments are grossly elevated, with ^{237}Np/^{239}Pu atom ratios exceeding 100, unlike global fallout (^{237}Np/^{239}Pu = 0.47; Kelley *et al*., 1999). The confluence zone contains input from material described in Lawrence Livermore National Laboratory's "Moody report" (Moody, 1995), a study that directly sampled materials within the PGDP process buildings.

3. Neptunium-237, attributed to the PGDP, is present in Little Beaver and Big Beaver Creek waters at concentrations of 0.8-1.0 picograms of ^{237}Np per liter, in the probable form of neptunyl cation. Plutonium could not be detected in 0.5 liter creek water samples.

4. Neptunium-237 was found at concentrations of approximately 2 pg/g in dry plant matter; there is significant neptunium uptake into grasses, based on limited measurements of one composite streambank grass sample. A soil-to-plant transfer factor, or concentration ratio, of 0.16 $\pm$ 0.06 was calculated from the results. This transfer factor is intermediate between literature values ranging from 0.001 to 10 for neptunium observed in different published studies of transfer factors measured in greenhouse and field studies. The Department of Energy is well aware of ^{237}Np plant uptake; DOE Basic Energy Sciences has funded studies (e.g., Montgomery *et al.,* 2023), yet DOE has not, in five decades, spoken directly to the citizens of southern Ohio about radionuclide uptake in plants near the PGDP.

5. The soil-to-plant transfer factor shows that ***in areas near the former PGDP that have been affected by water or air releases of ^{237}Np, this isotope is plant-available. Persons consuming foods grown within 10-15 miles of the DOE Portsmouth reservation are cautioned that they may be ingesting ^{237}Np.*** The study also observed that uranium is plant-available in the riparian zone, as the plant ash ^{235}U/^{238}U and ^{236}U/^{238}U atom ratio signatures reflected uptake of the PGDP-attributed enriched uranium in water and sediments.

6. Further work is warranted to understand the soil-to-plant transfer behavior of neptunium, along with uptake of enriched uranium, plutonium-239/240, and technetium-99 in native and cultivated plants growing near the former Portsmouth Gaseous Diffusion Plant.

Context of the study. Piketon, Ohio is the location of a decommissioned, partially demolished uranium enrichment facility that formerly produced low- and high-enriched uranium in three vast-scale uranium hexafluoride gaseous diffusion process buildings. At the US Government-owned Portsmouth gaseous diffusion plant (PGDP), which operated from 1954 until 2001, specific grades of enriched uranium were produced for power reactors, naval submarine reactors, and nuclear weapons. The recently demolished X-326 "high enrichment" process building was capable of refining uranium hexafluoride up to enrichment levels of 97% uranium-235.

Annual site environmental reports (ASER) published by the US Department of Energy (e.g., DOE, 2023) reveal various legacy and ongoing, contemporary off-site environmental impacts from the former PGDP. Radioisotopes specifically associated with PGDP include uranium (^{234}U, ^{235}U, ^{236}U, and ^{238}U), ^{99}Tc, ^{237}Np, ^{239}Pu, ^{240}Pu, and others; release pathways consisted of discharges to air, soil, surface water, and ground water.

The uranium enrichment facility at Piketon was designed to perform isotope separations by gaseous effusion of high-purity uranium hexafluoride. During the early Cold War era, as the US Government faced mounting demands for its uranium supplies, the PGDP operators commenced introducing what is commonly referred to by DOE staff as "reactor returns", namely, recycled uranium laden with varying amounts of ^{99}Tc, ^{237}Np, and Pu impurities remaining from the PUREX process (Moody, 1995). By the mid-1970's, the US Government's PGDP contractor, Goodyear Atomic Corporation (GAT) was aware of "transuranics", referring to findings of accumulations of Np and Pu at specific locations and material streams within the facility. A December 28, 1976 internal GAT memorandum **(Attachment 1)** expressed alarm over high Np and Pu concentrations in the MgF_2 solid sorbent traps used to scrub gases vented from bypassed sections of the cascade: *"one magnesium fluoride trap contained 190,000 d/m/g of neptunium-237 and 269,000 d/m/g of plutonium isotopes. Because of the grave dangers inherent in transuranics, both to human health and the environment, we request that all materials recovered from X-705 Oxide Conversion operations be sampled for transuranics before being processed for uranium recovery".*

Efforts by GAT commencing in the mid-1970's, aimed at constraining "transuranics" from being released into the environment, were only partially successful. To wit, the Department of Energy's PEGASIS database (DOE, 2024) reports 1.48 pCi/L ^{237}Np (2100 pg/L) in an August 8, 2018 groundwater sample from onsite Well X701-20W, indicating that a ^{237}Np groundwater plume is present (**Attachment 2**). This ^{237}Np plume is not discussed in DOE's ASER reports, however, based upon its location, it directly impacts Little Beaver Creek and its underlying shallow alluvial aquifer.

The Department of Energy (DOE) and its Federal predecessor agencies have known since no later than 1976 about a "transuranics" contamination problem at the Portsmouth gaseous diffusion plant, and its potential impact upon Little Beaver Creek. Nevertheless, there have been only limited studies of plant uptake of radionuclides near the PGDP, conducted for ^{99}Tc (Hoffman *et al.,* 1980; Acox, 1982; Hoffman *et al.,* 1982). These studies all point to transfer factors that exceed unity; Hoffman (1982) found a geometric mean of 7.4 and a geometric standard deviation of 2.8 for ^{99}Tc transfer factors in plants near collected in the perimeter zone of the PGDP site, indicating very high levels of Tc uptake into vegetation.

While the DOE has failed to address community members near PGDP about plant uptake of neptunium or other PGDP radionuclides, DOE-Basic Energy Sciences has long provided significant funding for research on the environmental behavior of neptunium, including its plant uptake properties (Peruski *et al*., 2018; Montgomery *et al*., 2023).

Methods. An August 2023 composite sample was collected of live and dead native annual grasses growing along the banks of Big Beaver Creek, from a location approximately 100 meters downstream of the confluence with Little Beaver Creek (**Attachment 3**). Thatch was cut with scissors at a level 5 cm above the ground. Care was taken to not include roots, nor any sediment adhering to the lower portion of the plants' stems. After collection, the grass was dried for several weeks in ambient air. The dry plant material was observed to be relatively free of adhering soil/sediment particles; it was shaken vigorously to remove any loosely adhering particles, although it was not subjected to any specific washing treatment. The dry grass was ground with a food processing mill, and was dry-ashed in an electric furnace at 450° C. The ash content of the ground plant material was 22 % of its dry mass.

Composite 500 gram samples of the top 10 cm of streambank soil/sediment, and humic floodplain soil were collected at different locations in the Little Beaver Creek - Big Beaver Creek confluence area. Composite samples of freshly deposited fine-grained sediments were also obtained from concrete bridge piers in the confluence area. Water samples ranging between one and three liters were collected in PET plastic containers (**Attachment 4**). Soil and sediment samples were oven-dried at 60° C and screened with a 0.4 mm brass sieve; five gram sub-samples were taken for analysis and dry-ashed at 550° C in 40 mL vials. Water sub-samples of 0.5 liters were used for ^{237}Np concentration measurements using the flow chart shown in **Attachment 5**.

Plant ash and sediment samples were similarly prepared for inductively coupled plasma mass spectrometry (ICPMS) analysis using the **Attachment 5** Pu separation process starting with Step 6. All sample types were spiked with an acid solution form of 28 picograms of ^{242}Pu, prepared using a stock solution purchased from the US National Institute of Standards and Technology (NIST 4334i). Soil, sediment and plant samples were leached with 20 mL of concentrated 70% nitric acid. Five gram sub-samples of a "negative control" consisting of powdered sandstone, as well as weapons-test positive control soils, known to contain ^{237}Np from global fallout, were also prepared. The results indicated reliable ^{237}Np measurement performance, and the ability to readily distinguish between "global fallout" and non-global fallout sources of neptunium, based on measurements of the ^{237}Np/^{239}Pu measured in control samples of known fallout or other provenance. A known "fallout" control soil, obtained from the Urseren Valley, Switzerland exhibited ^{237}Np/^{239}Pu = 0.47 $\pm$ 0.01 and ^{240}Pu/^{239}Pu = 0.193 $\pm$ 0.005. The atom ratios both agree very well with the Np - Pu atom ratios for global or "stratospheric" fallout described in Kelley *et al.* (1999). The agreement implies that Np and Pu are exhibiting parallel chemical behavior in this specific lab batch's chemical separations, enabling ^{237}Np measurements to be made using ^{242}Pu as a surrogate neptunium tracer.

To determine concentrations of ^{237}Np, ^{239}Pu and ^{240}Pu, a Thermo Scientific X2 quadrupole ICPMS was used at Northern Arizona University. The instrument was equipped with an APEX Model HF high-efficiency fluoropolymer sample introduction system and an APEX self-aspirating PFA Teflon concentric nebulizer. Ion count data were obtained in a peak-jump mode, using 10 millisecond dwell times at masses 237, 238, 239, 240, and 242. One "run" of 58 seconds consisted of 1000 sweeps through these five peaks; three to six "runs" were acquired for each sample solution. The ICPMS was tuned with a uranium-238 solution sensitivities were approximately 500,000 counts per second for the tuning solution; UH^+ yields were 0.000040 or less. The background count rates measured at mass 237 and 240 in "negative control" soil/sediment samples were less than one count per second, and less than two counts per second at mass 239.

Neptunium-237 was readily detected, at ion count rates well above ICPMS detection thresholds, for all samples of confluence zone water, soil, sediment and plant ash samples (e.g., detection of ^{237}Np in waters is shown in **Attachment 6**).

Results. **Attachment 7** shows results for ^{237}Np concentrations in picograms per gram, $^{239+240}$Pu activities, and the atom ratios ^{237}Np/^{239}Pu and ^{240}Pu/^{239}Pu for the confluence soil/sediment samples, the grass composite sample, and the Urseren control soil. The most striking feature of these results is the elevated ^{237}Np concentration in surface soil and freshly deposited sediments at the creek confluence. In comparison to the Urseren (stratospheric fallout) soil's concentration of 0.120 $\pm$ 0.002 picograms ^{237}Np per gram, concentrations of ^{237}Np in soils/sediments at the confluence area are about 100-fold elevated, ranging from 8 to 21 pg/g. It is apparent that the PGDP proximity represents an unusual "hotspot" of ^{237}Np environmental concentrations in ambient environments. At the same time, however, the $^{239+240}$Pu activities in confluence-area soil/sediment samples do not appear to be elevated in comparison to $^{239+240}$Pu activities normally associated with ubiquitous stratospheric fallout. However, the ^{240}Pu/^{239}Pu atom ratios in the soil/sediment confluence zone samples clearly reflect different Pu sources in addition to stratospheric fallout; the plutonium detected in the confluence area soil and sediment samples appears to be comprised of a mixture of stratospheric fallout, with a major added PGDP Pu component resembling the Moody (1995) signatures. The very high ^{237}Np/^{239}Pu ratios in the creek confluence soil and sediment samples underscore the unusual accumulation of non-fallout "transuranics" from PGDP in the Little Beaver-Big Beaver Creek riparian environment.

Attachment 7 also has results for the plant samples, with ^{237}Np and $^{239+240}$Pu being reported in terms of dry plant matter concentrations. The ^{237}Np concentrations indicate that significant uptake of neptunium into grasses is occurring in the riparian zone, just as one anticipates based upon the lab-measured transfer factors in Montgomery *et al.* (2023). The average of the plant-measured ^{237}Np/^{239}Pu atom ratios, 185 $\pm$ 20, is congruent with uptake of Np (and Pu) from the confluence-area soil/sediments (^{237}Np/^{239}Pu = 168 $\pm$ 20), rather than from ubiquitous stratospheric fallout sources (namely, ^{237}Np/^{239}Pu atom ratios are less than one, as reported in DOE-funded work by Kelley *et al.,* 1999).

The average ^{237}Np and $^{239+240}$Pu concentrations in the streambank grass are 2.05 $\pm$ 0.18 picograms per gram and and 0.034 $\pm$ 0.2 Becquerels per kilogram, respectively. For the confluence-area soils and sediments, the corresponding ^{237}Np average is 12.8 $\pm$ 4.8 picograms per gram, and $^{239+240}$Pu was found at 0.29 $\pm$ 0.21 Becquerels per kilogram. A resulting ^{237}Np transfer factor of (0.16 $\pm$ 0.06) is hence obtained. The magnitude of the transfer factor dictates that Np exhibits considerable plant uptake, and that portions of the soil column's Np inventory are being extracted from the soil, accumulating in plants, to be returned after the growing season to the top soil horizons, thereafter recycled and taken up into new plants repeatedly. The transfer factor for ^{237}Np is intermediate between factors of ~ 0.001 measured by Wallace *et al.* (1978) and the 1 to 10 range values reported in *Andropogon virginicus* grasses in greenhouse studies by Montgomery *et al.* (2023).

Neptunium-237, attributed to the PGDP, is present in Little Beaver and Big Beaver Creek waters at concentrations of 0.8-1.0 picograms of ^{237}Np per liter for samples collected on March 3, 2024. Neptunium could not, however, be detected in Big Beaver Creek waters, upstream of the entry of contaminated PGDP drainage in Little Beaver Creek. This confirms the nexus between the neptunium reported in onsite groundwaters by DOE in Pegasis, and dissolved neptunium in creek water. It is likely that the riparian plants are taking up neptunium through a combination of the fresh Np flux delivered directly in creek water, and uptake of Np from the large stored reservoir of this isotope in confluence zone soils/sediments.

Studies are needed of the species form of neptunium that is present in the water. The literature points to the stability of NpO_2^+ (aq), known as neptunyl cation, and suggests that this is the expected species of neptunium present in most natural waters under neutral or slightly acidic pH's where some dissolved oxygen is present.

Additional work is needed to measure a more extensive spatial/temporal set of soil, sediment, plant and plant samples, to better define the transfer factors for ^{237}Np, as well as other PGDP-sourced radionuclides, in field and greenhouse-based experiments. Previous DOE work in the early 1980's (Hoffman *et al.,* 1980; Acox, 1982; Hoffman *et al.,* 1982) examined the plant uptake of technetium-99 into plants near the then-operating PGDP, and measured very strong uptake with transfer factors exceeding unity. ***The public is urged to pay attention to soil quality when consuming local plants grown near PGDP.***

Attachment 1

INTERDEPARTMENTAL CORRESPONDENCE

TO: Listed Distribution

DATE: December 30, 1976
FROM DEPT: 922
CODE NO: GAT-922
REFERENCE:

DEPT:
LOCATION:

SUBJECT: CONTAMINATION FROM TRANSURANIC ELEMENTS

Traces of transuranic elements, such as plutonium and neptunium, are present in reactor-return materials processed in the X-705 Building, These elements recently have been detected in sludge from the X-701-B Holding Pond and in spent trapping materials stored in the X-744-G Warehouse. Because transuranics represent a health hazard, it is necessary to take appropriate steps to assure the safety of GAT personnel and the local environment.

Also, GAT analytical procedures for transuranics must be improved. The liquid effluents from X-701-B in the East Drainage Ditch have been monitored for transuranics since November 1976, but present GAT analytical procedures have a limit of detection that is equal, at best, to about 7 percent of the ERDA recommended. concentration guide (RCG) for neptunium-237. These detection limits should be lowered to below 1 percent of the RCG to increase the effectiveness' of the environmental monitoring program. The detection limits for airborne transuranics should be improved similarly.

To accomplish these purposes a committee is hereby formed comprising the following persons :

C. P. Blackledge, Chairman
R. I. Kaplan
C. F. Trivisonno
J. S. Murrell
W. E. Martin
J, C, Dikeman

This group is assigned the responsibility of studying all aspects of transuranic contamination problems. at GAT, including the following:

1. Developing more effective means of excluding transuranics from plant effluents and further assuring effectiveness of GAT health protection procedures;

2. Developing more sensitive analyses for transuranics

Trace quantities of transuranic elements.are probably being released to Little Beaver Creek; we estimate that the concentrations in water arc below the current analytical detection limits, Since neptunium and plutonium have been-detected in X-701-B, it' will be necessary to develop and implement a routine environmental monitoring program. Furthermore, we must take immediate action to prevent significant quantities of these radioisotopes from entering the environment from X-705.

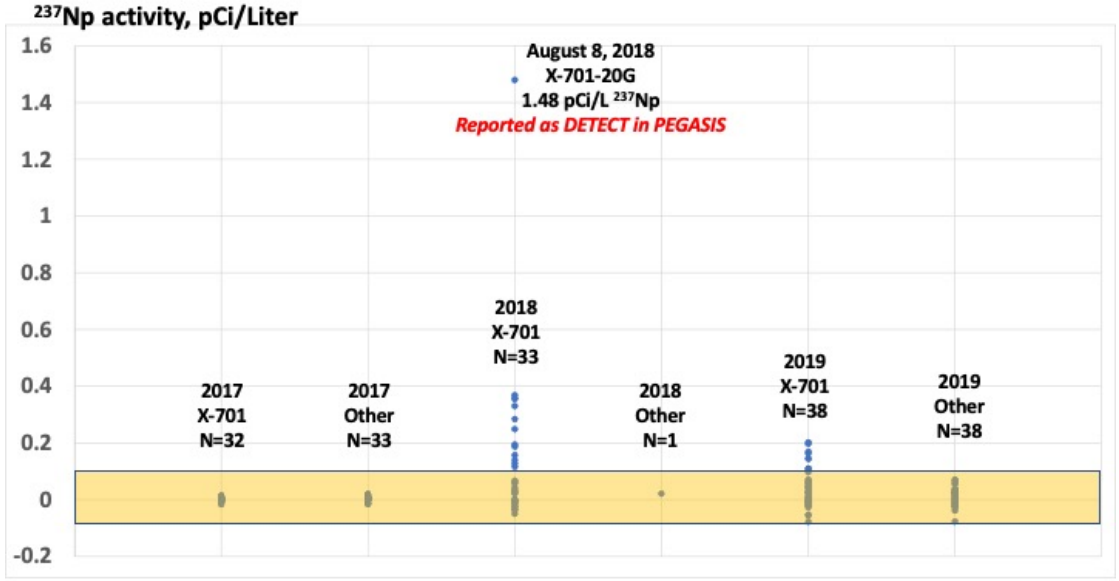
Since transuranic elements are generally considered to bc both more toxicologically and radiologically hazardous than uranium, it is imperative that we establish health protection measures to ensure that our employees are not inadvertently exposed to, these materials until we can' initiate adequate controls to assure their safety when handling *these* materials.
We need to :

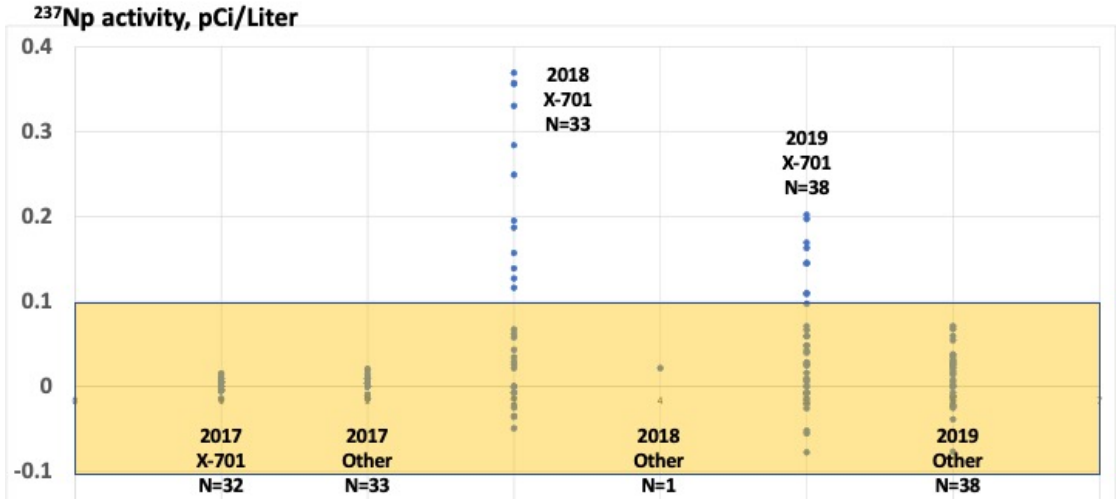
-identify the presence of transuranic elements (isotope , quantity, chemical form, and location) ;
-isolate transuranic elements from personnel and the environment ;
-develop interim handling procedures; and
-develop health protection, environmental: protection, and material control programs.

Since transuranic elements are generally considered to bc both more toxicologically and radiologically hazardous than uranium, it is imperative that we establish health protection measures to ensure that our employees are not inadvertently exposed to, these materials until we can' initiate adequate controls to assure their safety when handling *these* materials.
We need to :

-identify the presence of transuranic elements (isotope , quantity, chemical form, and location) ;
-isolate transuranic elements from personnel and the environment ;
-develop interim handling procedures; and
-develop health protection, environmental: protection, and material control programs.

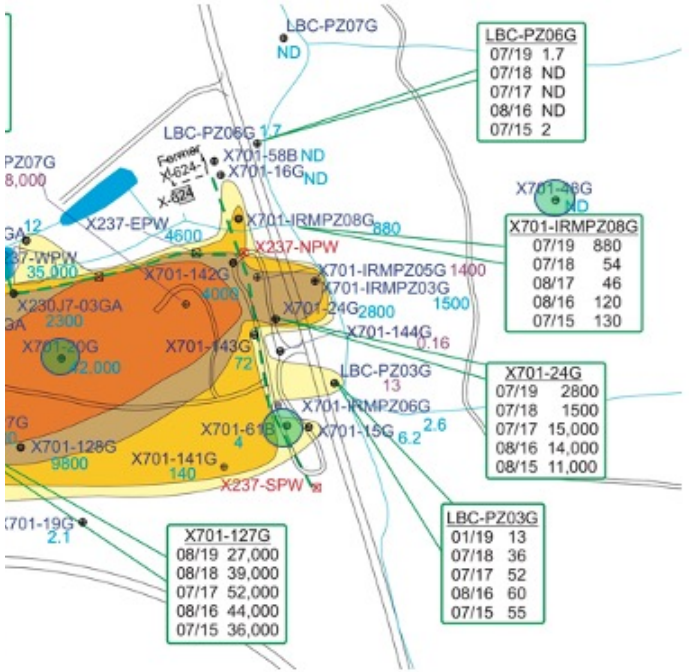
Attachment 2





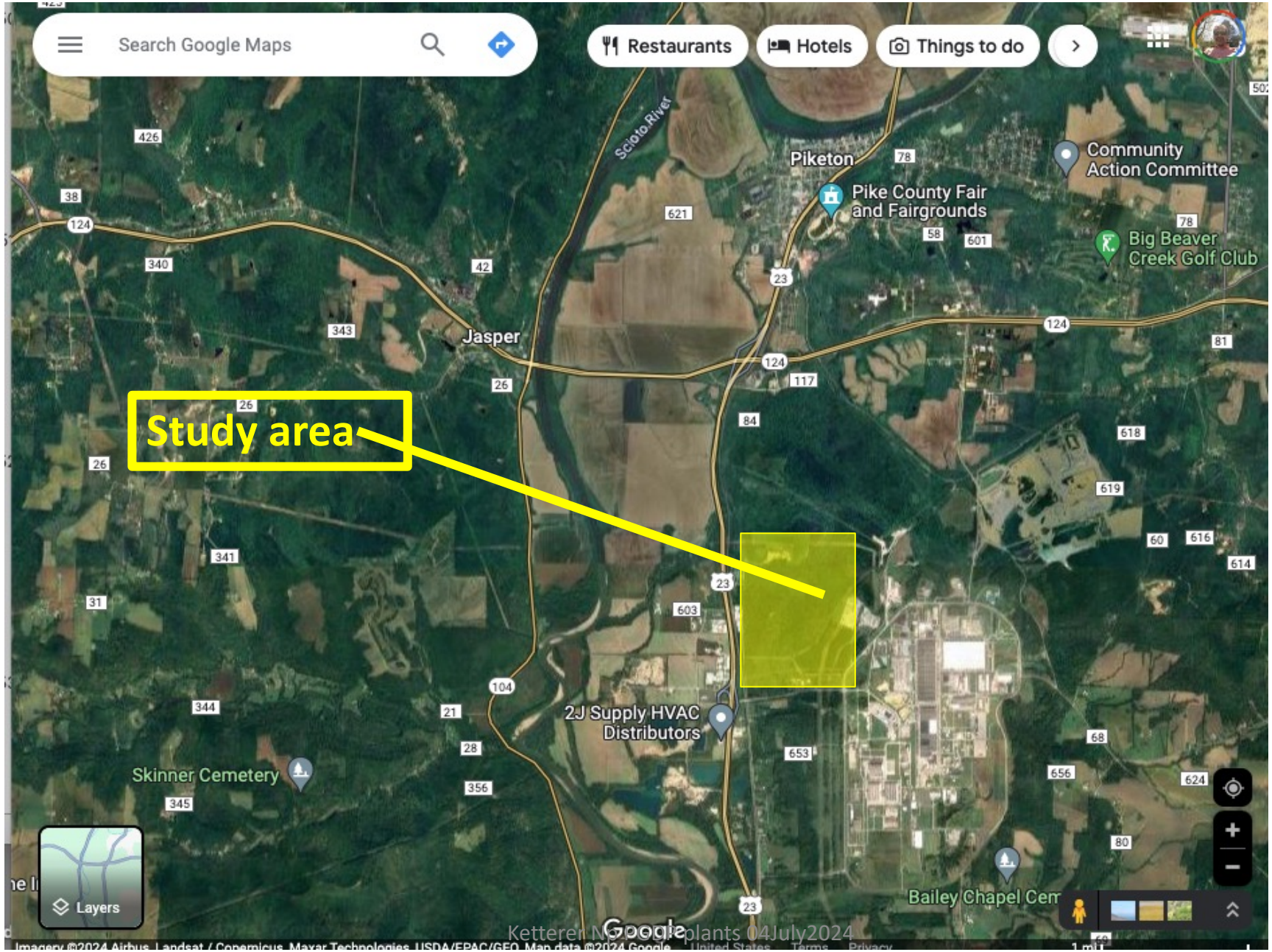
Station Name	Station Type	Date Collected	Media Type Description	Matrix	Results	Units	Chemical Name	Detect Flag
X701-20G	WL	8/8/18 0:00	Groundwater	WATER	1.48	pCi/L	Neptunium-237	DETECT
X701-48G	WL	8/7/18 0:00	Groundwater	WATER	0.369	pCi/L	Neptunium-237	NONDETECT
X701-TC54G	WL	8/13/18 0:00	Groundwater	WATER	0.357	pCi/L	Neptunium-237	NONDETECT
X701-TC10G	WL	8/8/18 0:00	Groundwater	WATER	0.356	pCi/L	Neptunium-237	NONDETECT
X701-TC17G	WL	8/8/18 0:00	Groundwater	WATER	0.33	pCi/L	Neptunium-237	NONDETECT
X701-TC05G	WL	8/13/18 0:00	Groundwater	WATER	0.284	pCi/L	Neptunium-237	NONDETECT
X701-127G	WL	8/8/18 0:00	Groundwater	WATER	0.249	pCi/L	Neptunium-237	NONDETECT
X701-TC01G	WL	8/13/18 0:00	Groundwater	WATER	0.195	pCi/L	Neptunium-237	NONDETECT
X701-TC03G	WL	8/13/18 0:00	Groundwater	WATER	0.187	pCi/L	Neptunium-237	NONDETECT
X701-TC48G	WL	8/8/18 0:00	Groundwater	WATER	0.157	pCi/L	Neptunium-237	NONDETECT
X701-TC22G	WL	8/13/18 0:00	Groundwater	WATER	0.139	pCi/L	Neptunium-237	NONDETECT
X701-66G	WL	8/14/18 0:00	Groundwater	WATER	0.127	pCi/L	Neptunium-237	NONDETECT
X701-TC61G	WL	8/13/18 0:00	Groundwater	WATER	0.116	pCi/L	Neptunium-237	NONDETECT

Station Name	Station Type	Date Collected	Media Type Description	Matrix	Results	Units	Chemical Name	Detect Flag
X701-TC28G	WL	8/14/19 0:00	Groundwater	WATER	0.202	pCi/L	Neptunium-237	NONDETECT
X701-TC10G	WL	8/14/19 0:00	Groundwater	WATER	0.197	pCi/L	Neptunium-237	NONDETECT
X701-20G	WL	8/7/19 0:00	Groundwater	WATER	0.169	pCi/L	Neptunium-237	NONDETECT
X701-TC03G	WL	8/12/19 0:00	Groundwater	WATER	0.163	pCi/L	Neptunium-237	NONDETECT
X701-TC05G	WL	8/12/19 0:00	Groundwater	WATER	0.145	pCi/L	Neptunium-237	NONDETECT
X701-TC61G	WL	8/12/19 0:00	Groundwater	WATER	0.145	pCi/L	Neptunium-237	NONDETECT
X701-TC67G	WL	8/12/19 0:00	Groundwater	WATER	0.11	pCi/L	Neptunium-237	NONDETECT
X701-20G	WL	8/7/19 0:00	Groundwater	WATER	0.109	pCi/L	Neptunium-237	NONDETECT

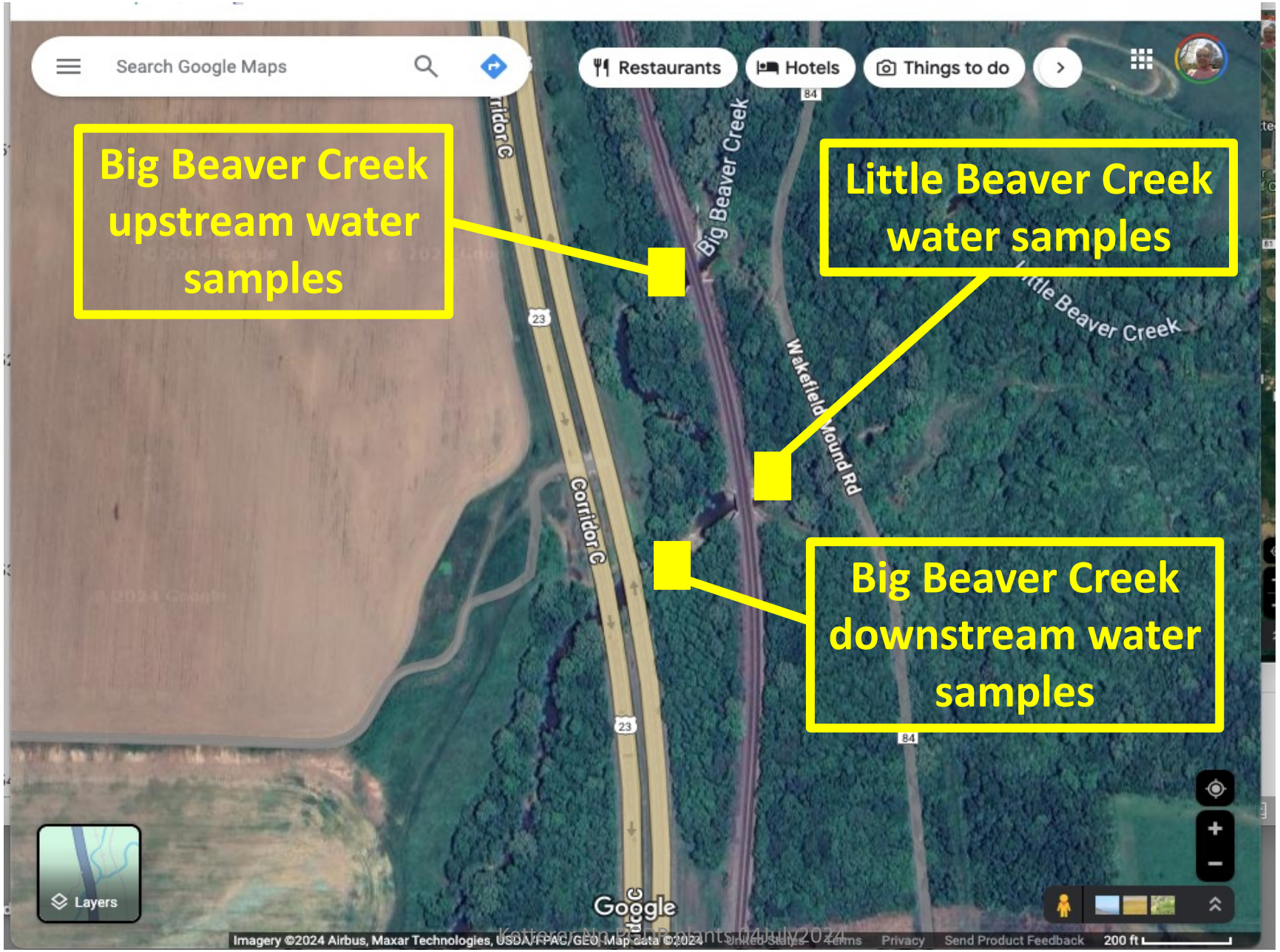


Examples of locations where ^{237}Np activity exceeds apparent detection threshold of 0.10 pCi/L in 2018 and/or 2019

Attachment 3

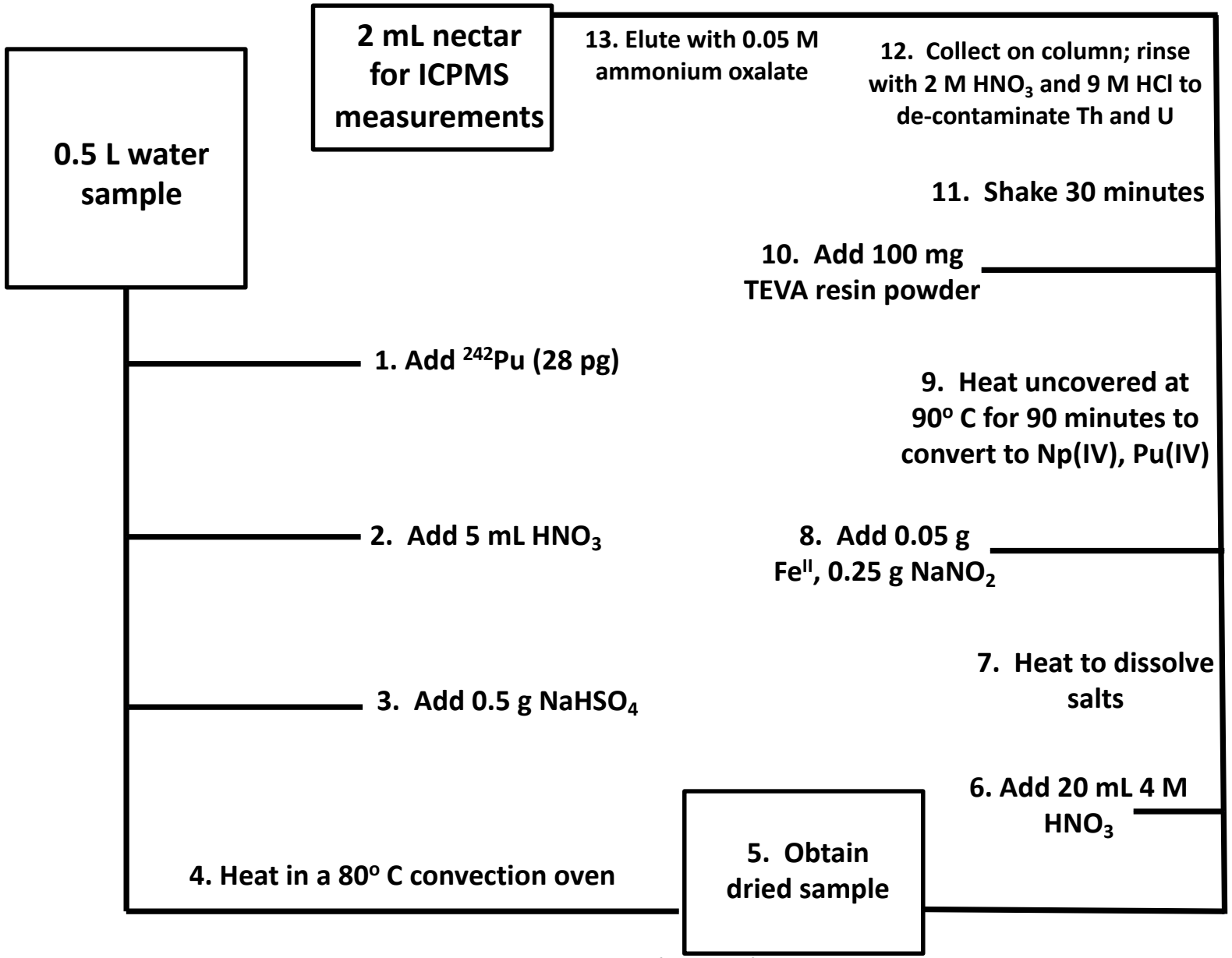


Attachment 3

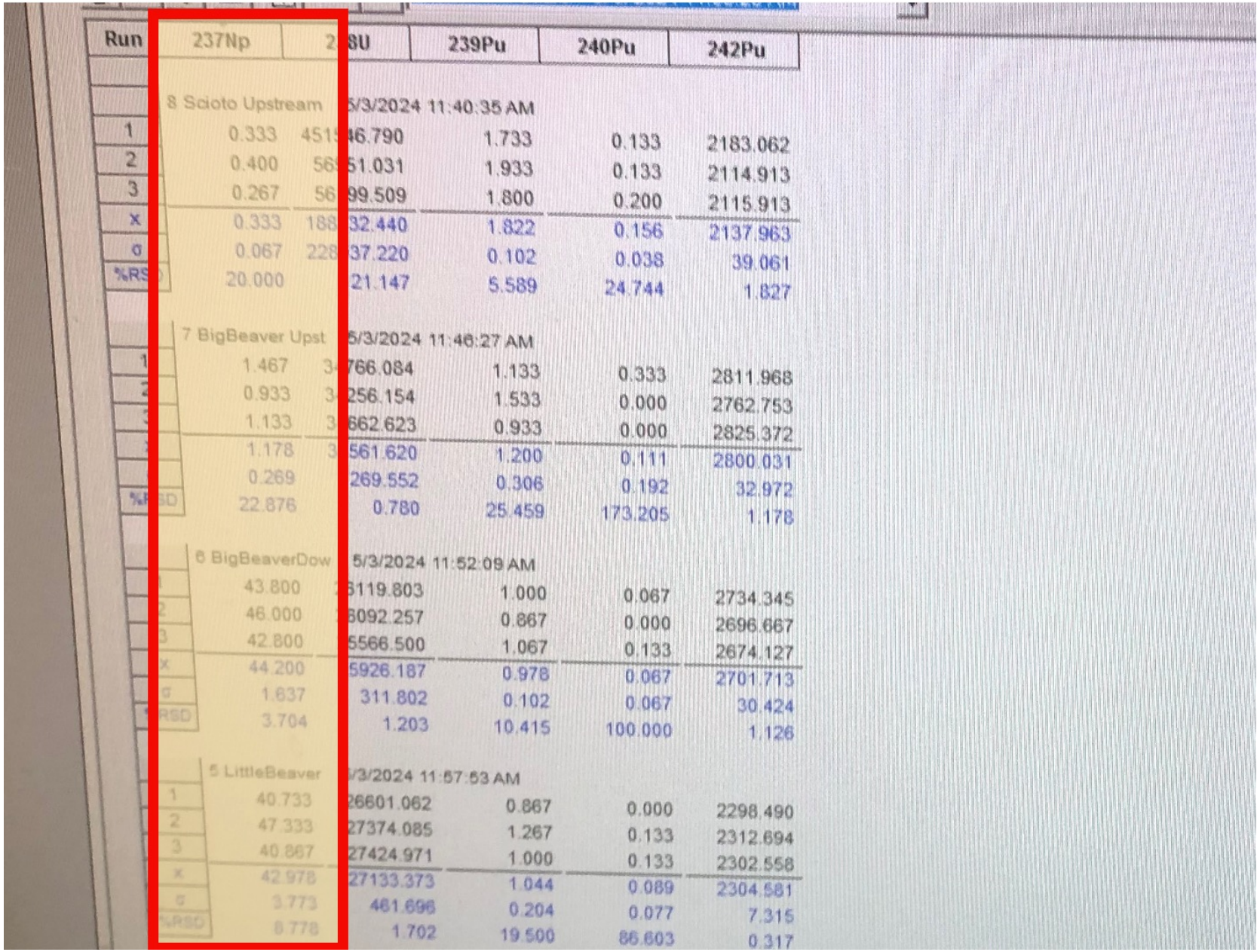


Attachment 4

Attachment 5



Attachment 6



Run	237Np	2	8U	239Pu	240Pu	242Pu
	8 Scioto Upstream		5/3/2024 11:40:35 AM			
1	0.333	451	46.790	1.733	0.133	2183.062
2	0.400	56	51.031	1.933	0.133	2114.913
3	0.267	56	99.509	1.800	0.200	2115.913
x	0.333	188	32.440	1.822	0.156	2137.963
σ	0.067	228	37.220	0.102	0.038	39.061
%RS	20.000		21.147	5.589	24.744	1.827
	7 BigBeaver Upst		5/3/2024 11:46:27 AM			
1	1.467	3	766.084	1.133	0.333	2811.968
2	0.933	3	256.154	1.533	0.000	2762.753
3	1.133	3	662.623	0.933	0.000	2825.372
x	1.178	3	561.620	1.200	0.111	2800.031
σ	0.269		269.552	0.306	0.192	32.972
%RSD	22.876		0.780	25.459	173.205	1.178
	6 BigBeaverDow		5/3/2024 11:52:09 AM			
1	43.800		6119.803	1.000	0.067	2734.345
2	46.000		6092.257	0.867	0.000	2696.667
3	42.800		5566.500	1.067	0.133	2674.127
x	44.200		5926.187	0.978	0.067	2701.713
σ	1.637		311.802	0.102	0.067	30.424
%RSD	3.704		1.203	10.415	100.000	1.126
	5 LittleBeaver		5/3/2024 11:57:53 AM			
1	40.733		26601.062	0.867	0.000	2298.490
2	47.333		27374.085	1.267	0.133	2312.694
3	40.867		27424.971	1.000	0.133	2302.558
x	42.978		27133.373	1.044	0.089	2304.581
σ	3.773		461.696	0.204	0.077	7.315
%RSD	8.778		1.702	19.500	86.603	0.317

Attachment 7

Sample/description	^{237}Np (pg/g)	$^{239+240}$Pu (Bq/kg)	^{237}Np/^{239}Pu	^{240}Pu/^{239}Pu
Fine-grained sediment + moss	8.0 ± 0.1[a]	0.19 ± 0.02	142 ± 13	0.111 ± 0.021
Fine-grained sediment + moss	12.3 ± 0.1	0.26 ± 0.02	146 ± 7	0.081 ± 0.015
Fine-grained sediment	15.5 ± 0.1	0.27 ± 0.01	170 ± 7	0.078 ± 0.017
Fine-grained sediment	21.3 ± 0.1	0.24 ± 0.02	298 ± 23	0.123 ± 0.023
Bank sediments, Big Beaver Creek	8.2 ± 0.1	0.16 ± 0.01	161 ± 4	0.096 ± 0.020
Bank sediments, Big Beaver Creek	9.9 ± 0.1	0.17 ± 0.03	190 ± 29	0.099 ± 0.007
Humic soil, floodplain	14.2 ± 0.1	0.75 ± 0.03	66 ± 4	0.133 ± 0.011
Streambank grass sub-sample #1[a]	1.79 ± 0.02	0.033 ± 0.007	159 ± 13	0.083 ± 0.035
Streambank grass sub-sample #2	2.12 ± 0.02	0.036 ± 0.003	183 ± 14	0.088 ± 0.026
Streambank grass sub-sample #3	2.05 ± 0.02	0.031 ± 0.004	194 ± 26	0.067 ± 0.011
Streambank grass sub-sample #4	2.22 ± 0.05	0.036 ± 0.009	205 ± 30	0.090 ± 0.043
Urseren control soil (fallout Np, Pu)	0.120 ± 0.002	1.00 ± 0.02	0.47 ± 0.01	0.193 ± 0.005

[a]All uncertainties are ± one experimental standard deviation of 3 to 6 sequential measurements. [b]Concentrations of ^{237}Np and $^{239+240}$Pu were measured by leaching dry-ashed samples, but are reported are on the basis of dry plant mass.

References

Acox, T.A., 1982. Study of technetium uptake in vegetation in the vicinity of the Portsmouth Gaseous Diffusion Plant, accessed June 6, 2024 at: https://inis.iaea.org/search/search.aspx?orig_q=RN:14723928.

Brookins, Douglas G., 2012. Eh-pH diagrams for geochemistry, Springer Science and Business Media.

Department of Energy, 2023. 2022 Annual Site Environmental Report (ASER), accessed June 6, 2024 at: https://www.energy.gov/sites/default/files/2023-12/2022%20ASER%20Presentation_SSAB%2011.2.23.pdf.

Department of Energy, 2024. PPPO Environmental Geographic Spatial Information System, accessed June 6, 2024 at https://pegasis.ports.pppo.gov/Pegasis/Default.aspx

Hoffman, F.O. *et al.*, 1980. Sampling of technetium-99 in vegetation and soils in the vicinity of operating gaseous diffusion facilities, accessed June 7, 2024 at https://www.osti.gov/biblio/5008976.

Hoffman, F.O.; Garten Jr., C.T.; Lucas, D.M.; Huckabee, J.W., 1982, Environmental behavior of technetium in soils and vegetation. Implications for radiological assessments, 10.1021/es00098a008.

Kelley, J.M.; Bond, L.A.; Beasley, T.M., 1999, Global distribution of Pu isotopes and ^{237}Np, https://doi.org/10.1016/S0048-9697(99)00160-6.

Montgomery, D.A.; Edayilam, N.; Page, H.; Sheriff, S.A.; Tharayil, N.; Powell, B.A.; Martinez, N.E., 2023, Coparative uptake, translocation and plant mediated transport of Tc-99, Cs-133, Np-237 and U-238 in Savannah River Site soil columns for the grass species *Andropogon virginicus*, http://dx.doi.org/10.1016/j.scitotenv.2022.159400.

Moody, K.J., 1995, Forensic radiochemistry of PUBLIC site inspection samples, accessed June 6, 2024 at https://www.osti.gov/biblio/26594.

Peruski, K.M., Maolubier, M., Kaplan, D.I., Almond, P.M, Powell. B.A., 2018. Mobility of aqueous and colloidal neptunium species in field lysimeter experiments, https://doi.org/10.1021/acs.est.7b05765