NORTHERN ARIZONA UNIVERSITY

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INTERIM REPORT: Results for Isotopic Studies of Uranium in Environmental Samples from the Vicinity of the Aerojet Ordnance Tennessee Facility, Jonesborough, TN

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Executive Summary: A study is being conducted to determine uranium "signatures" in environmental media (water, soil, aquatic sediments, and biota) near the Aerojet Ordnance Tennessee (AOT) facility in Jonesborough, Tennessee. The overall purpose of the work is to determine the extent to which uranium (U) has been dispersed at off-site locations near AOT. The study has involved collection of environmental samples from locations near AOT, followed by laboratory analyses of these samples. The AOT facility has a known history of fabricating materials with "depleted uranium" (DU). This report provides additional results that extend the base of information provided in an earlier report issued October 22, 2011.

The October 2011 results, as well as the additional results given herein, results clearly indicate the presence of DU, evidently originating from AOT, in water, soil, and sediment samples. Depleted uranium is distinguishable based upon the isotope ratios $^{234}U/^{238}U$, $^{235}U/^{238}U$, and $^{236}U/^{238}U$ measured in the samples. Mixing between DU and naturally occurring in the environment is evident from the results found herein.

Depleted uranium has been found in the following media: A) water in Little Limestone Creek downstream of the AOT facility; B) sediments in Little Limestone Creek downstream of the AOT facility; and C) soils containing atmospherically deposited DU at off-site locations near the AOT facility.

Purpose. A study is being conducted of uranium "signatures" in environmental media (water, soil, aquatic sediments, and biota) near the AOT facility in Jonesborough, Tennessee. The overall purpose of the work is to determine the extent to which uranium (U) has been dispersed off-site. The study has involved collection of environmental samples from locations near AOT, followed by laboratory analyses of these samples. Mass spectrometry, a well-established analytical technique, has been used to measure relative numbers of atoms for different isotopes (nuclear forms) of U. The results from mass spectrometry are used to compare U found in the environment vs. its known/expected isotopic composition in Nature, in order to evaluate whether naturally occurring U is being mixed with U from other sources that are not naturally occurring. This study is ongoing, given the open-ended scope/magnitude of the question, and the paucity of publicly available information regarding environmental contamination and releases from AOT.

Scope: The results presented in this report are intended to be of a *demonstrative* nature. This report emphasizes interpreting changes in U ratios as "signatures" of the presence of DU from AOT.

Background: Uranium isotopes. Uranium (U) has four different isotopes (nuclear forms) that occur in detectable quantities in Nature. The chemical behavior of each of these isotopes is essentially identical; the different isotopes are designated by their mass numbers. The mass number (a) is the sum of the number of protons (z) and neutrons (n) present in the nucleus of the U atom, and isotopes are designated by the element symbol with the mass number being written as a superscript on the left side of the element symbol: ^{a}U. All U atoms have z = 92. The four isotopes that occur in detectable quantities in Nature are ^{234}U, ^{235}U, ^{236}U, and ^{238}U. Each of these isotopes has different nuclear properties (half-life, decay energy, and susceptibility to fission).

Since U occurs in Nature, it is ubiquitous in water, soil, sediment, and the biosphere. The isotopes ^{235}U and ^{238}U are primordial; they have been present since the solar system was accreted. The isotope ^{236}U is present in very small amounts in Nature as a result of spontaneous fission processes of other U isotopes, followed by neutron capture of ^{235}U. The isotope ^{234}U is present in small amounts in Nature as a continuously produced decay product in the ^{238}U decay series. The relative proportions of ^{234}U, ^{235}U, ^{236}U, and ^{238}U present in Nature are fairly constant and predictable. ^{238}U is the most abundant isotope, and it is convenient to express the isotope composition of U as "ratios" or "isotope ratios", that is, as ratios of numbers of atoms relative to ^{238}U. The ratio ^{235}U/^{238}U is most often used to distinguish naturally occurring U vs. DU, and is discussed in this report. Additional information about sources of U can be garnered through analysis of ^{234}U/^{238}U and ^{236}U/^{238}U, as reported by Lloyd *et al.* (2009).

Processes related to the nuclear fuel cycle can introduce U into the environment with altered isotope compositions. "Enriched" U refers to U with a *higher* ^{235}U/^{238}U atom ratio than the naturally occurring ratio, and "depleted" refers to U with a *lower* ^{235}U/^{238}U atom ratio than the naturally occurring ratio. In the enrichment process, the lighter isotopes are selectively concentrated, with the objective of preparing a material of enhanced ^{235}U content for use as a nuclear reactor fuel or a fission weapon device. The enrichment process also enhances the ^{234}U content, and ^{234}U/^{238}U is higher in enriched U than the typical values found in Nature. Similarly, the enrichment process also produces "tails" from which most of the ^{235}U has been removed (depleted U). Depleted U also has a lower ^{234}U/^{238}U than is found in naturally occurring U.

Many samples of enriched or depleted U also contain readily detectable amounts of ^{236}U; this isotope signals the presence of U that has previously been irradiated by neutrons in a nuclear reactor or bomb. During the Cold War era, the US Government was concerned with an apparent shortage of U, and much of the U introduced into the nuclear fuel cycle during this timeframe had been recovered from plutonium production reactors, referred to as "recycled" U. Most samples of DU encountered in the US contain ^{236}U introduced from inclusion of this "recycled" U component.

The following compares the U ratios expected in Nature vs. "enriched" and "depleted" U:

Type of Uranium	^{234}U/^{238}U	^{235}U/^{238}U	^{236}U/^{238}U
Naturally occurring	~ 0.000055 **(a)**	0.0072527 **(b)**	~ 0.000000001 **(c)**
Enriched U	> 0.000055	> 0.0072527	Up to ~ 0.01
Depleted U	< 0.000055	< 0.0072527 **(d)**	~ 0.00003 typical

(a) The ratio between ^{234}U and ^{238}U is variable in Nature due to disequilibria in open systems present in the Earth surface environment. **(b)** This ratio is essentially constant in Nature, and has only been shown to vary by ~ 1-2 parts per thousand relative as a result of natural fractionation processes. A few exceptional naturally occurring situations where $^{235}U/^{238}U$ differs, such as the Oklo reactor, have also been identified. **(c)** The highest concentrations of ^{236}U in Nature are found in U ores, with $^{236}U/^{238}U \sim 10^{-10}$ being typical. Natural samples of non-ore materials are expected to contain these or lower levels of ^{236}U. **(d)** DU typically contains about 0.2% ^{235}U, with a $^{235}U/^{238}U$ atom ratio of about 0.002.

When U of different isotope compositions is mixed, the resulting sample exhibits a ratio that reflects the isotope compositions of the different pure components, and the respective total number of U atoms originating from each source. As a hypothetical example of this, when varying amounts of a DU sample having $^{235}U/^{238}U = 0.002$ are mixed with naturally occurring U ($^{235}U/^{238}U = 0.0072527$), the resulting samples exhibit ratios of $0.002 < {}^{235}U/^{238}U < 0.0072527$. In this situation, any detectable decrease in $^{235}U/^{238}U$ below 0.0072527, outside of the ~ 1-2 part per thousand relative deviation expected in Nature, is clear and incontrovertible evidence for the presence of some U in the sample being derived from DU. Similarly, mixing behavior occurs for the $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ratios. The detection of *any* measurable $^{236}U/^{238}U$ usually indicates the presence of enriched or depleted U, from a recycled U component (previously irradiated in a reactor), though small amounts of ^{236}U have also been produced *via* nuclear weapons testing.

Samples and Analyses. This interim report discusses U signature results from the following: A) surface water from Little Limestone Creek in the vicinity of the AOT facility; B) soil samples from locations near the AOT facility, and C) soil/sediment samples from floodplain areas in Little Limestone Creek downstream of the AOT facility.

Grab samples of water have been collected in 50 mL polypropylene test tubes. Soil samples have been collected with a trowel (surface composites of 0-5 cm), or as 4.1 or 5.2 cm i.d. cores collected in galvanized steel tubing. Sampling activities have been conducted in collaboration with members of the local community.

Samples were prepared by appropriate laboratory procedures, as required, and were analyzed by the technique of inductively coupled plasma mass spectrometry (ICPMS). The facilities at Northern Arizona University were used in this study. The author has 24 years experience in the use of ICPMS in environmental samples and ratio measurements, and has 16 years experience on using ICPMS in studies of U in environmental media.

October 2011 Results. Results are tabulated below. The numbers in parentheses adjacent to the reported ratios are the uncertainties in the measured ratios (± one standard deviation of 3 sequential measurements); thus, 0.0046(1) should be read as 0.0046 ± 0.0001.

It is evident that *all* of these samples, with the exception of the upstream water sample LL-6w, exhibit $^{235}U/^{238}U$ ratios lower than the naturally occurring value of 0.0072527. It is clear that this must stem from mixing between naturally occurring U and DU. AOT possesses an NPDES permit (TN0057983) for release treated discharges into Little Limestone Creek; therefore it

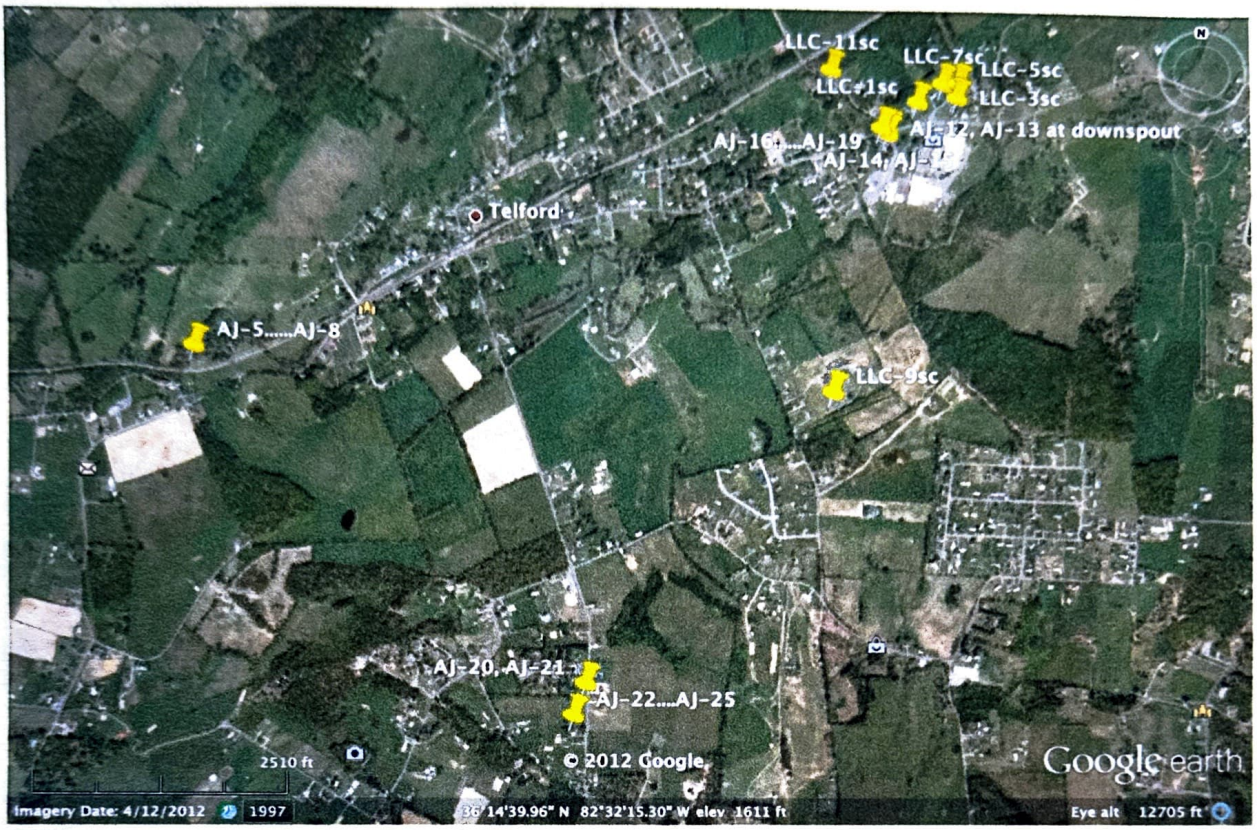
follows that AOT is the source of the DU being detected in the environment, as no other plausible sources exist.

The soil sample 18s was collected from a residence near the facility (the identity of this site is confidential). The $^{235}U/^{238}U$ of 0.0063 is outside of measureable error of the naturally occurring U value (0.00725) and hence it appears that DU is present in this soil. DU could be present in this location either as a result of atmospheric deposition or as a result of direct dumping of DU-containing residues at the property.

The soil core LL-5s was collected from a non-residential location in the immediate vicinity of the NFS facility (the identity of this site is also confidential). Analyses of the 0-5 and 5-10 cm depth intervals of this core demonstrate the presence of DU mixed with naturally occurring U, as evidenced by $^{235}U/^{238}U$ values systematically lower than naturally occurring U. It is believed that the DU in this location results from atmospheric deposition. Similar patterns of atmospheric deposition have also been found near DU facilities in Colonie, NY (Lloyd *et al.*, 2009) and near the Starmet U metallurgy in Concord, MA (Ketterer *et al.*, unpublished work).

Sample Name	Location	Date collected	$^{235}U/^{238}U$
26w	Water, Little Limestone Creek at residential location, Telford-New Victory Road, downstream of Aerojet	May 28, 2010	0.0046 (1)
27w	Water, Little Limestone Creek at residential location, Telford-New Victory Road, downstream of Aerojet	May 28, 2010	0.0046 (1)
32w	Water, Little Limestone Creek at the bridge over Clyde Miller Road, downstream of Aerojet	June 1, 2010	0.0046 (1)
33w	Water, Little Limestone Creek at the bridge over Clyde Miller Road, downstream of Aerojet	June 1, 2010	0.0045 (1)
LL-1w	Water, Little Limestone Creek at the bridge over Clyde Miller Road, downstream of Aerojet	May 11, 2011	0.0053 (1)
LL-2w	Water, Little Limestone Creek at residential location, Telford-New Victory Road, downstream of Aerojet	May 11, 2011	0.0056 (1)
LL-6w	Water, Little Limestone Creek ***upstream*** of Aerojet	Sept 29, 2011	0.0074 (1)
LL-10w	Water, Little Limestone Creek at residential location, Gravel Hill Road, downstream of Aerojet	Sept 29, 2011	0.0043 (1)
18s	Soil from residential property near Aerojet, possible site of dumping of Aerojet sludges	June 1, 2010	0.0063 (1)
LL-5s 0-5cm	Soil from non-residential property near Aerojet, possible location with accumulation of atmospheric deposition	May 11, 2011	0.0054 (1)
LL-5s 5-10cm	Soil from non-residential property near Aerojet, possible location with accumulation of atmospheric deposition	May 11, 2011	0.0063 (3)
	Natural U signature		**0.0072527**

June 2012 Results. Additional samples were collected in October 2011 to further delineate the presence of DU in the off-site environment near AOT. The samples reported herein consist of soil cores collected in 4.1 cm diameter galvanized steel tubing to a drive depth of 30 cm. The approximate location of the soil cores is as shown below.



Samples AJ-5 through AJ-8 consisted of soil/sediment obtained from a small mid-channel island in Little Limestone Creek downstream of AOT. The remaining samples reported herein consist of soil cores obtained at off-site locations in the proximity of AOT.

The cores were sectioned into 0-5 cm, 5-10 cm, 10-20 cm, and > 20 cm intervals. Material was dried, weighed, and pulverized; nominal 0.2 gram sub-samples were dry-ashed and then dissolved by treatment with 2.5 mL of concentrated HNO_3 and 1.5 mL of concentrated HF overnight at 80° C. After heating, 0.9 g of boric acid was added to consume the excess HF, and the sample solutions were diluted to 50 mL. Sample aliquots were diluted and an Ir internal standard was added to measure U concentrations by quadrupole ICPMS. An additional sample aliquot was separated by extraction chromatography with UTEVA resin to prepare a concentrated U fraction for isotope ratio measurements; the ratio measurements were also conducted by quadrupole ICPMS.

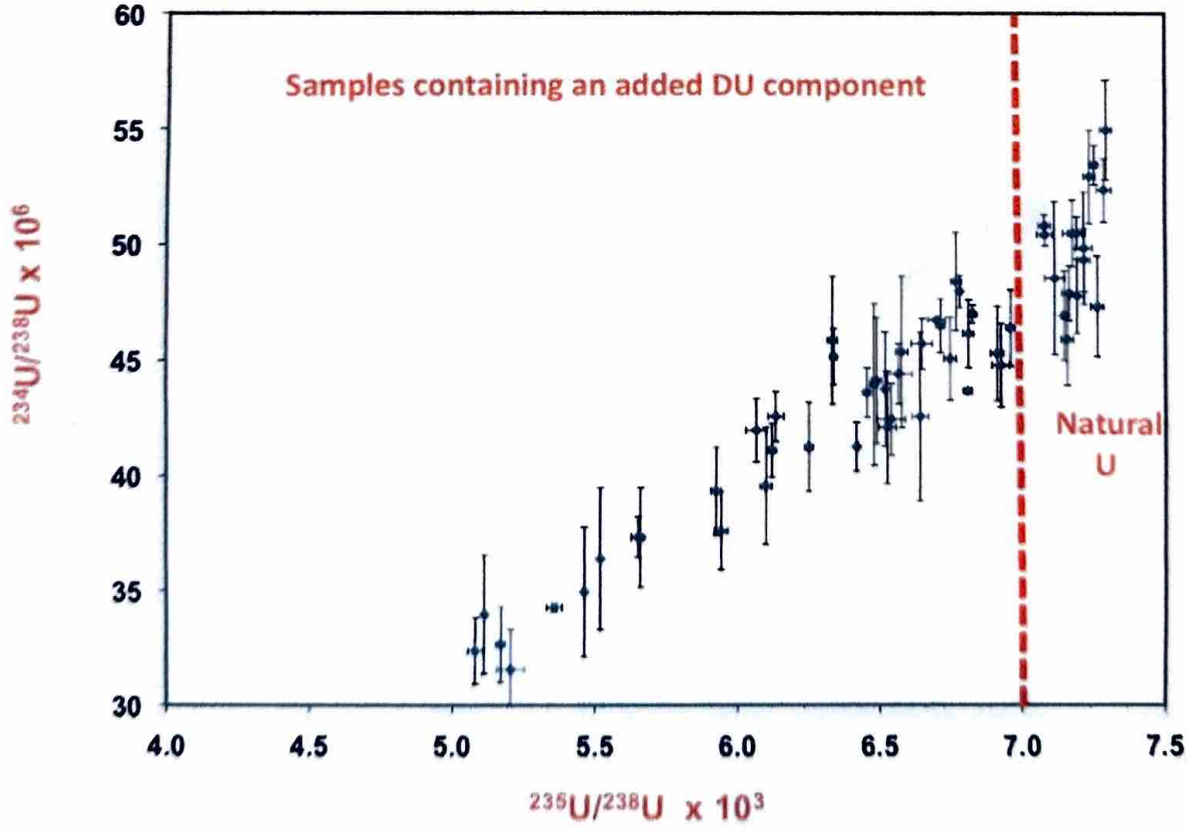
Results are given below. The tabulated values of the $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ratios represent the actual values multiplied by 10^6; for example, the tabulated $^{234}U/^{238}U$ value of 43.6 for Sample "AJ-5 0-5" should be read as 0.0000436. The "sd" refers to the measured standard deviation of n=3 or n=5 sequential measurements; namely, the $^{234}U/^{238}U$ value for Sample "AJ-5 0-5" can be read as 0.0000436 ± 0.0000025. The tabulated values of the $^{235}U/^{238}U$ ratio represents the actual values multiplied by 10^3; for example, the tabulated $^{235}U/^{238}U$ value of 6.517 for Sample "AJ-5 0-5" should be read as 0.006517 ± 0.000005. The depth intervals in units of cm (e.g., 0-5) follow the core's identity.

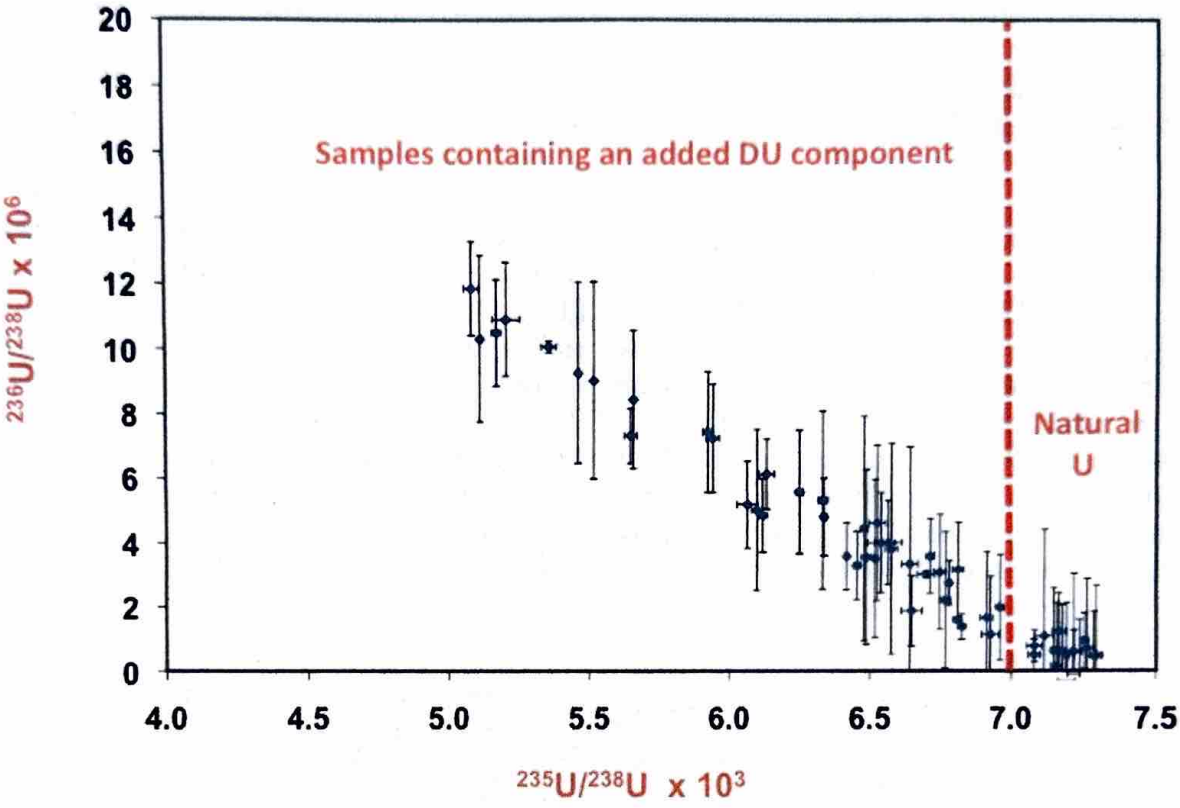
U Atom Ratio and U Concentration Results. Samples shaded contain an added DU component as determined by the criterion $^{235}U/^{238}U < 0.0070$ (refer to discussion in text).

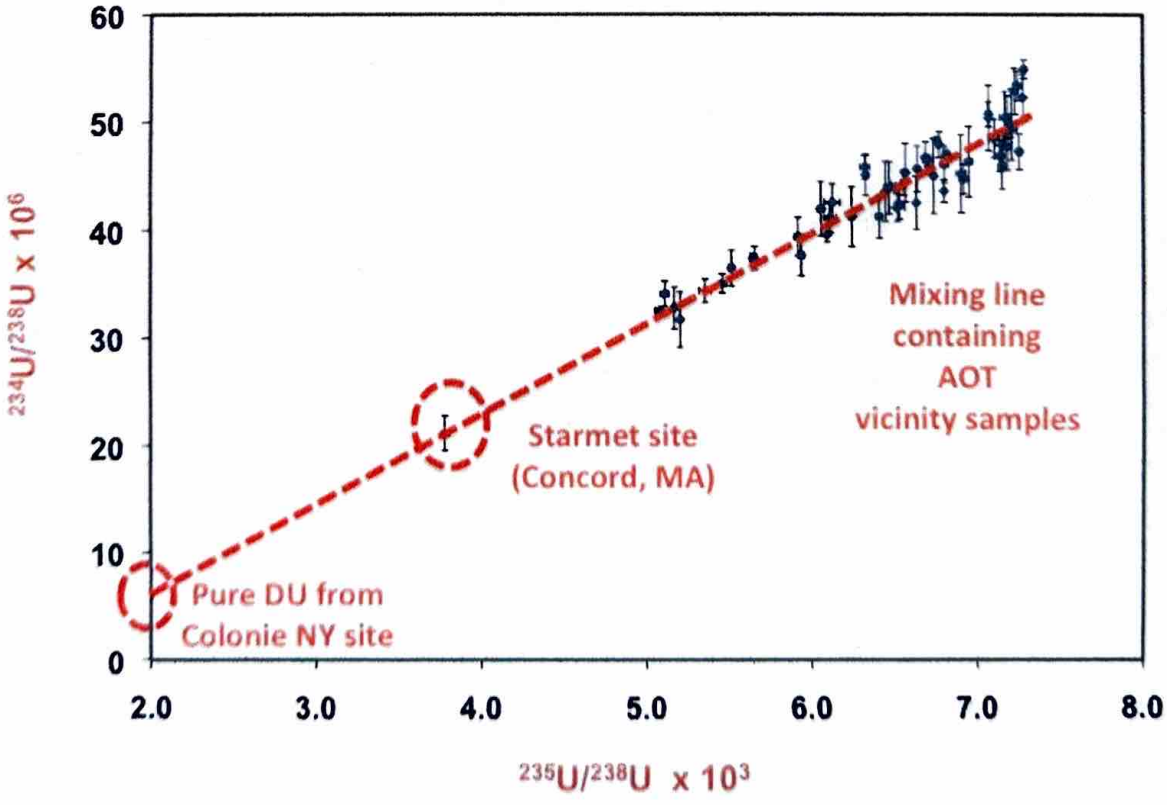
Lab ID	Sample ID	$^{234}U/^{238}U$	$^{234}U/^{238}U$ sd	$^{235}U/^{238}U$	$^{234}U/^{238}U$ sd	$^{236}U/^{238}U$	$^{236}U/^{238}U$ sd	U conc ppm	U conc sd
31	AJ-5 0-5	43.6	2.5	6.517	0.005	3.5	0.6	1.85	0.03
32	AJ-5 5-10	42.4	3.6	6.640	0.028	3.3	0.1	1.96	0.03
33	AJ-5 10-20	45.2	2.1	6.912	0.022	1.6	0.3	2.03	0.02
34	AJ-5 20-26	45.8	2.0	7.159	0.020	0.2	0.3	2.11	0.02
35	AJ-6 0-5	41.1	1.0	6.416	0.005	3.6	0.7	1.86	0.03
39	AJ-7 0-5	43.6	0.1	6.809	0.014	1.6	0.7	0.93	0.01
43	AJ-8 0-5	48.3	2.1	6.766	0.019	2.2	0.4	0.64	0.01
44	AJ-8 5-10	45.6	1.1	6.647	0.037	1.9	0.2	0.49	0.01
47	AJ-12 0-5	34.2	0.2	5.357	0.027	10.1	0.1	5.01	0.06
48	AJ-12 5-10	32.4	1.4	5.081	0.027	11.9	1.4	4.47	0.04
49	AJ-12 10-20	42.3	1.6	6.539	0.050	4.0	0.1	3.34	0.07
50	AJ-12 20-25.5	47.8	1.2	7.165	0.025	1.2	0.3	3.01	0.03
51	AJ-13 0-5	33.9	2.6	5.112	0.004	10.3	0.1	4.09	0.07
52	AJ-13 5-10	31.5	1.7	5.204	0.048	10.9	1.0	4.43	0.06
53	AJ-13 10-20	42.4	1.1	6.132	0.028	6.1	0.8	3.30	0.05
54	AJ-14 0-5	45.7	2.8	6.331	0.016	5.3	0.5	2.68	0.04
55	AJ-14 5-10	41.1	1.9	6.249	0.013	5.6	0.4	2.96	0.04
56	AJ-14 10-20	45.0	1.2	6.336	0.010	4.8	0.6	3.19	0.04
57	AJ-14 20-24	47.9	0.7	6.780	0.007	2.7	0.3	3.17	0.05
58	AJ-15 0-5	39.5	2.5	6.098	0.019	5.0	0.2	3.06	0.07
59	AJ-15 5-10	41.9	1.4	6.064	0.036	5.2	0.6	3.26	0.05
60	AJ-15 10-20	41.0	1.1	6.118	0.014	4.9	0.7	3.31	0.05
61	AJ-15 20-27.5	50.7	0.5	7.078	0.021	0.5	0.4	2.72	0.04
62	AJ-16 0-5	46.9	0.4	6.824	0.013	1.4	0.3	2.61	0.06
65	AJ-17 0-5	37.3	0.9	5.649	0.023	7.3	0.0	3.97	0.07
66	AJ-17 5-10	34.9	2.8	5.461	0.004	9.3	0.3	4.18	0.08
67	AJ-17 10-20	43.5	1.1	6.454	0.012	3.3	0.7	3.34	0.06
68	AJ-18 0-5	37.2	2.1	5.658	0.003	8.5	0.6	3.80	0.08
75	AJ-20 0-5	52.3	1.4	7.287	0.025	0.5	0.1	4.92	0.06
76	AJ-20 5-10	53.4	0.9	7.254	0.013	0.9	0.4	4.92	0.07
77	AJ-20 10-17	54.9	2.2	7.295	0.020	0.5	0.5	4.19	0.04
78	AJ-21 0-5	52.9	2.0	7.237	0.020	-0.4	0.0	5.11	0.05
81	AJ-22 0-5	50.4	0.7	7.193	0.030	-0.3	0.0	2.99	0.04
84	AJ-24 0-5	49.2	1.4	7.217	0.022	-0.1	0.2	3.19	0.05
88	LL9 0-5	44.7	1.8	6.925	0.031	1.1	0.2	1.76	0.02
89	LL9 5-10	48.4	3.3	7.114	0.034	1.1	0.1	2.06	0.04
90	LL9 10-25	49.7	2.4	7.217	0.029	0.6	0.1	2.03	0.03
91	LL8 0-5	50.4	1.5	7.177	0.033	0.6	0.2	2.82	0.05
95	AJ-25 0-5	46.8	1.9	7.147	0.010	0.6	0.2	3.04	0.04
99	LLC-3sc 0-5	32.6	1.6	5.170	0.015	10.5	0.1	4.99	0.07
100	LLC-3sc 5-10	36.3	3.0	5.517	0.004	9.1	0.6	4.81	0.10
101	LLC-3sc 10-20	50.3	0.5	7.080	0.027	0.8	0.2	3.47	0.06
102	LLC-3sc 20-29	46.4	1.2	6.712	0.014	3.6	0.4	3.53	0.05
103	LLC-5sc 0-5	44.3	1.3	6.564	0.048	4.0	0.4	4.06	0.08
104	LLC-5sc 5-10	42.0	2.4	6.525	0.029	4.6	0.4	4.49	0.07
105	LLC-5sc 10-20	43.8	3.5	6.478	0.012	4.4	0.3	4.50	0.06
106	LLC-5sc 20-28.5	44.9	1.8	6.746	0.021	3.1	0.6	4.26	0.06
107	LLC-1sc 0-5	39.2	1.9	5.923	0.017	7.5	0.4	3.68	0.05
108	LLC-1sc 5-10	37.5	1.7	5.941	0.022	7.3	0.7	4.04	0.06
111	LLC-11sc 0-5	46.0	1.5	6.811	0.019	3.2	0.8	4.28	0.09
112	LLC-11sc 5-10	46.6	0.1	6.698	0.031	3.0	0.3	4.16	0.08
115	LLC-7sc 0-5	44.0	2.7	6.486	0.018	3.6	0.3	4.15	0.07
116	LLC-7sc 5-10	45.2	3.3	6.575	0.022	3.8	0.6	4.24	0.10
149	AJ-23 0-5	47.2	2.2	7.264	0.022	0.7	0.3	2.48	0.03
153	LLC-9sc 0-5	47.7	1.6	7.193	0.026	0.5	0.1	2.98	0.06

Evidence for the Presence of DU in Off-Site Soils. The presence of U from other-than-naturally occurring sources can be detected using all three of the ratios tabulated above; as DU mixes with naturally occurring U, the result is a lower $^{234}U/^{238}U$ and $^{235}U/^{238}U$ along with a non-zero $^{236}U/^{238}U$. The precise distinction between "natural U only" samples and those containing an added DU component originating from AOT is best addressed by statistical treatments. Nevertheless, it is apparent that samples exhibiting $^{235}U/^{238}U < 0.0070$ all contain an added DU component of AOT origin. Secondary, co-varying criteria for detecting added DU are given by the $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ratios. However, the application of $^{234}U/^{238}U$ is hampered by the expected variation of this ratio in Nature that results from open-system disequilibria between ^{234}U and ^{238}U in surface soils. The detection of added DU based upon a non-zero $^{236}U/^{238}U$ is encumbered by the possible presence of small amounts of ^{236}U from nuclear weapons testing fallout as well as difficulties associated with detection of very low $^{236}U/^{238}U$ ratios.

The co-variance between the $^{235}U/^{238}U$ ratio and the ratios $^{234}U/^{238}U$ and $^{236}U/^{238}U$ provides a further indication that naturally occurring U is being mixed with DU. It is evident that the presence of DU is always accompanied not only by a decrease in $^{235}U/^{238}U$ but by a correlated decrease in $^{234}U/^{238}U$ as well as an increase in $^{236}U/^{238}U$. This behavior can be depicted in three-isotope mixing plots of $^{234}U/^{238}U$ vs. $^{235}U/^{238}U$ and $^{236}U/^{238}U$ vs. $^{235}U/^{238}U$:

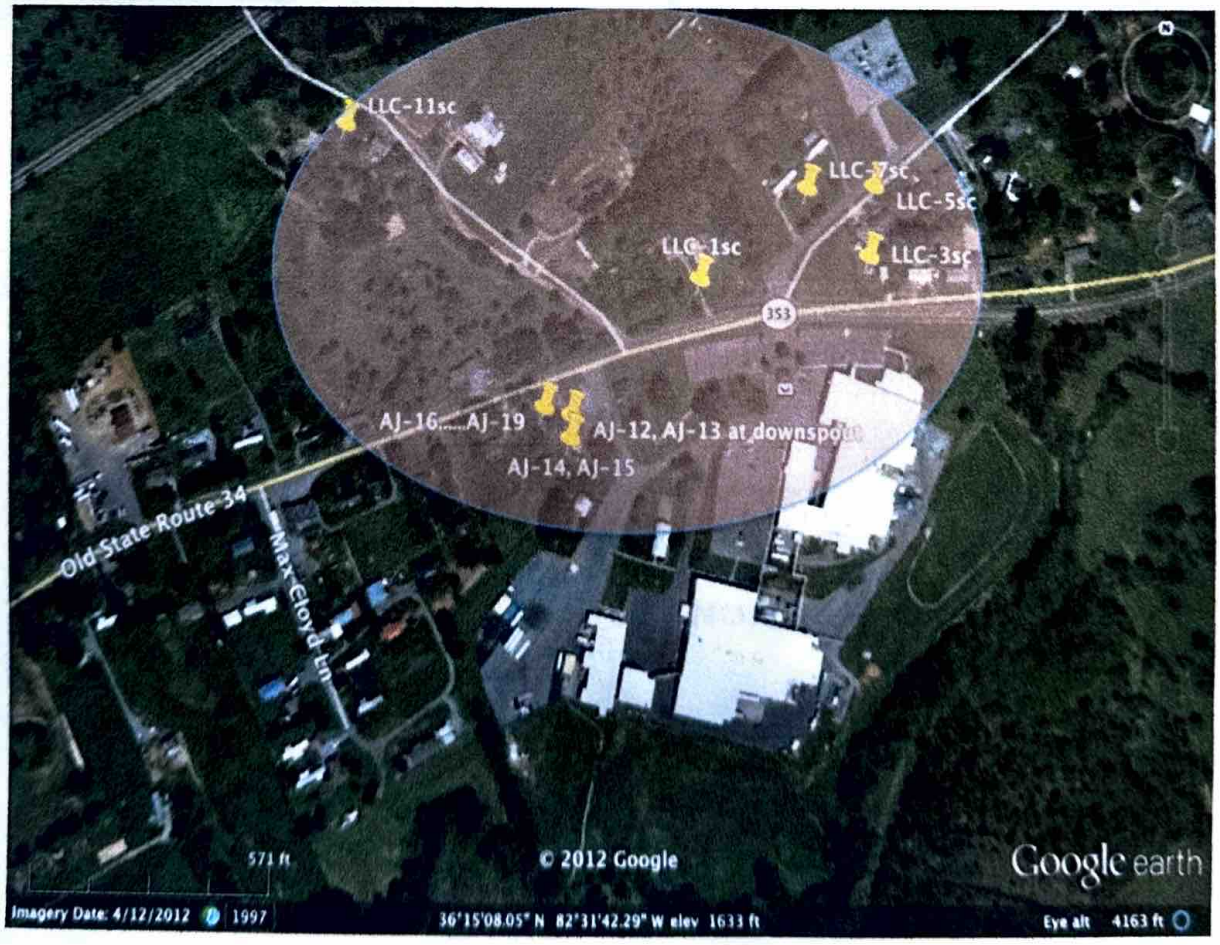



Samples containing an added DU component
Natural U
$^{236}U/^{238}U \times 10^6$
$^{235}U/^{238}U \times 10^3$

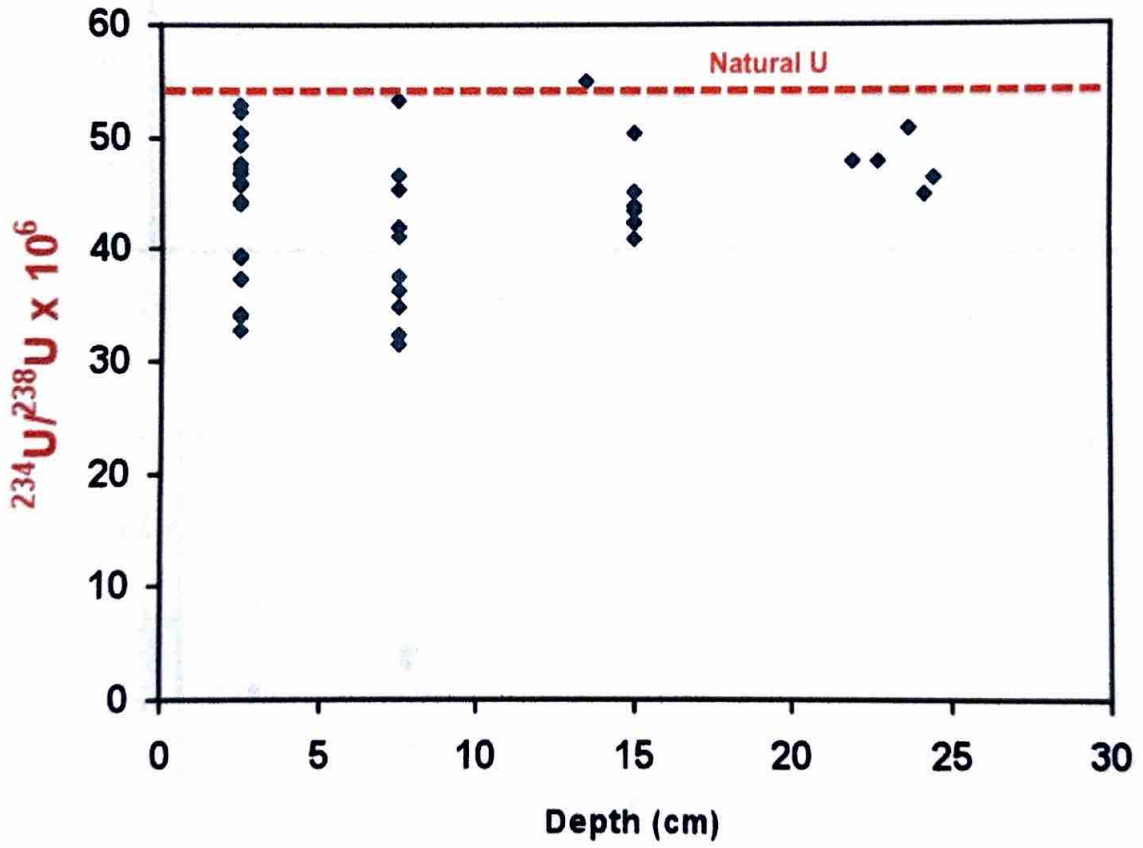

Mixing line containing AOT vicinity samples
Starmet site (Concord, MA)
Pure DU from Colonie NY site
$^{234}U/^{238}U \times 10^6$
$^{235}U/^{238}U \times 10^3$

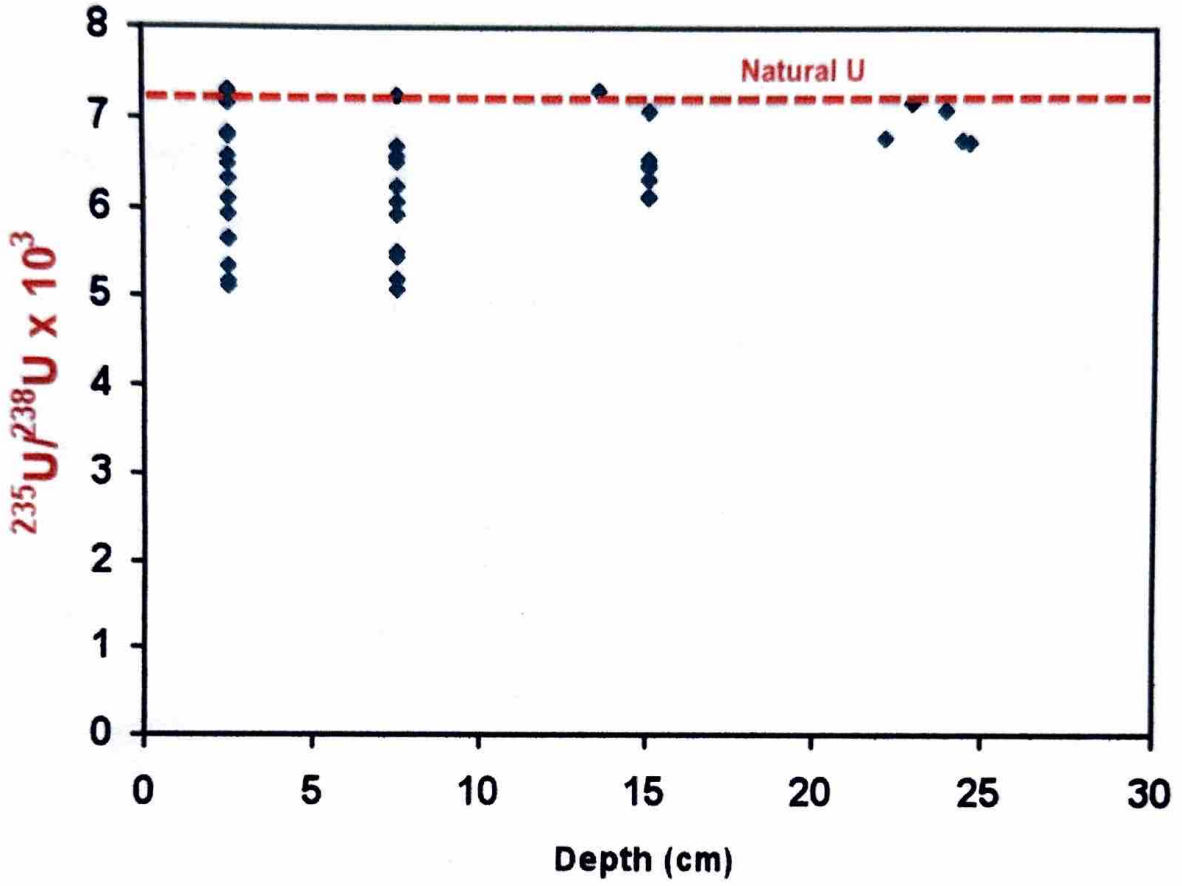
These plots indicate that naturally occurring U is being mixed with a single, characteristic isotopic blend of DU. Furthermore, it is apparent that the DU contamination in the environment near the AOT facility is isotopically identical to DU contamination found at similar US sites (Starmet site, Concord, MA, Ketterer, unpublished results; National Lead site, Colonie, NY, Lloyd *et al.*, 2009).

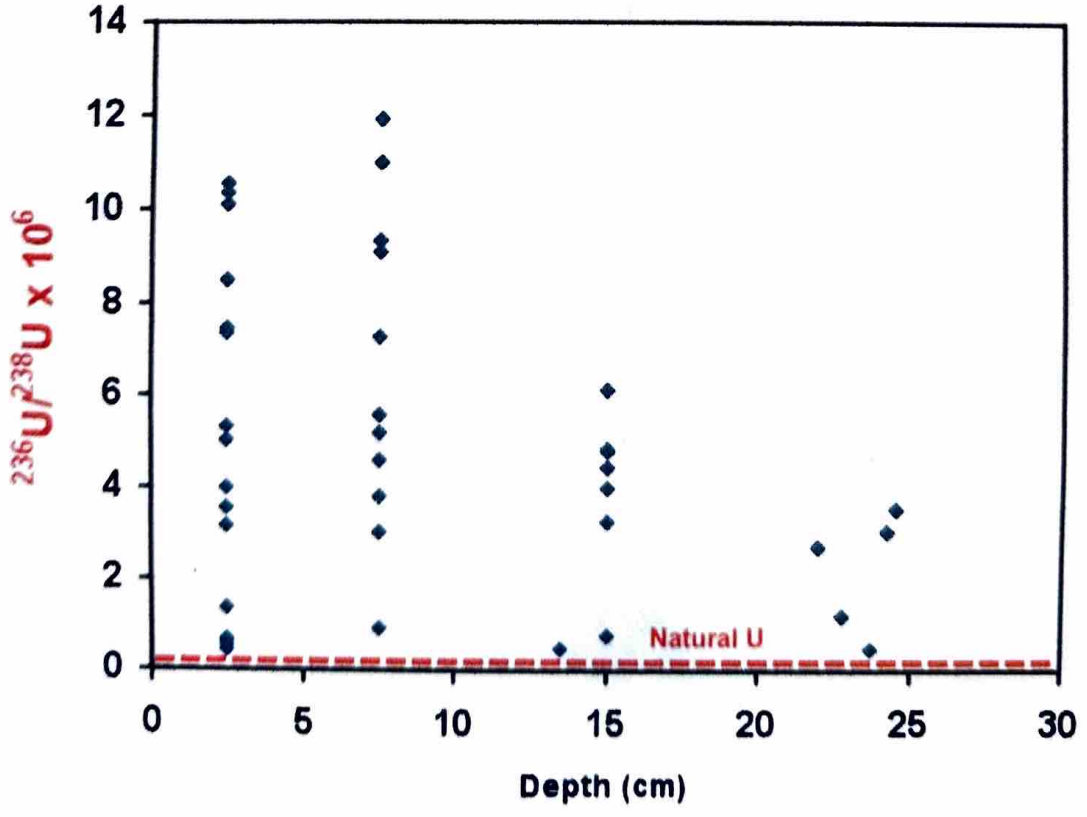
Geographic extent of contamination. Off-site environmental contamination with DU originating from the AOT facility is evident in soils from several locations close to AOT (AJ-12 through AJ-18 inclusive; LL9; LLC-1sc; LLC-3sc; LLC-5sc; LLC-7sc; LLC-11sc). DU contamination is also evident in the soil/sediment samples collected from a small island in Little Limestone Creek downstream of AOT (AJ-5 through AJ-8 inclusive). Soil samples obtained from more distant locations southwest of AOT do not exhibit any DU detectable by the measurements performed herein. An approximate delineation of the known plume is shown below. This delineation does not suggest that the plume is necessarily restricted to within this region; rather, it is quite likely that all undisturbed surface soils within this zone will exhibit some DU contamination. Additional work is warranted to better define the geospatial extent of the AOT contamination.



Atmospheric release pathway. Given below are plots of the ratios $^{234}U/^{238}U$, $^{235}U/^{238}U$ and $^{236}U/^{238}U$ as a function of depth. The results of the soil cores are fully consistent with the existence of an atmospheric deposition pathway for the dispersion of DU contamination from the AOT location into the surrounding off-site areas. This is evident through examination and comparison of the depth profiles of the U ratios in the soil cores, both individually and collectively. It is evident that the DU signature is most prominent in the 0-5 and 5-10 cm depth intervals of the soil cores exhibiting the contamination. In contrast, the locations where no DU deposition is evident, the U ratios are consistent as a function of depth, and are in agreement with the naturally occurring U isotope composition. The trends observed herein are in accordance with the depth behavior of atmospheric DU emissions released from facilities at Colonie, NY (Lloyd *et al.*, 2009) and Concord, MA (Ketterer *et al.*, unpublished studies).




Natural U
$^{235}U/^{238}U \times 10^{3}$
Depth (cm)
0
1
2
3
4
5
6
7
8
0
5
10
15
20
25
30


Natural U
$^{236}U/^{238}U \times 10^{6}$
Depth (cm)
0
2
4
6
8
10
12
14
0
5
10
15
20
25
30

Submitted by:

Michael E. Ketterer, PhD

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Reference Cited: Lloyd NS, Chenery SRN, Parrish RR. The distribution of depleted uranium contamination in Colonie, NY, USA. Sci Total Environ 2009; 408:397-407.