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Analysis of Savannah Survey Sediment Collections of 30 Sep 04

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Lawrence Livermore National Laboratory
Nonproliferation, Arms Control, and International Security Directorate

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Lawrence Livermore National Laboratory

30 Nov 04

From: LLNL / Nonproliferation, Arms Control, and International Security Directorate

To: Defense Threat Reduction Agency
6301 Telegraph Road
Alexandria, VA 22306

Subj: Analysis of Savannah Survey Sediment Collections of 30 Sep 04

Summary:

This report summarizes laboratory radiochemical analyses of sediment samples collected by the Defense Threat Reduction Agency during the DoD Savannah Survey operations of Sep 04.

The analytic goal was to determine if Wassaw Sound sediment collections of 30 Sep 04 display evidence for local anthropogenic uranium, as distinct from the recognized regional background stemming from the Savannah River Site (SRS). See References 1-9. Radiochemical methods were selected to maximize detection sensitivity for such anthropogenic uranium.

Within the suite of twelve collections, there would be evidence for a localized source if individual collections were to differ from the population as a whole. If in fact non-natural uranium were observed, definitive determination of whether the source was SRS effluent or a localized release would likely involve additional field sampling.

These collections were logged by the LLNL Forensics Science Center, photographed, and laboratory chain-of-custody was begun. The inventory received at LLNL is reported in Table 1. The separate collections were not assigned any relative priority among them.

LLNL has separately reviewed detailed records of the item in question, and determined what materials are involved and what radiochemical assays are of value. Attempting quantitative estimates of source-item material release, transport, and collection levels would be quite uncertain. Rather, present assays examine for departure from natural background isotopic compositions.

To summarize the findings, analyses for all collections displayed natural uranium isotopic composition – considering the $^{238}U/^{235}U$ and $^{236}U/^{235}U$ ratios – within measurement uncertainties, which were quite low. No one collection or set of collections stood apart from the others in its uranium isotopes. These $^{238}U/^{235}U$ data uncertainty levels also determined with 99.5% confidence (a three-standard-deviation determination) that no more than 0.46% of the total uranium present in the Savannah Survey collections could be depleted uranium. See Refs. 10-15.

Based on gamma spectroscopy assays of ^{137}Cs specific activity (Becquerels per kilogram), no collection displayed ^{137}Cs content levels above background levels present in the natural marine/riverine environment. Therefore, there is no measurable detection of Savannah River Site effluent in these collections based on ^{137}Cs assay. See Refs 1-9.

Assays of this particular collection suite give no ability to localize the USAF asset of interest.

Table 1. Savannah Survey Collection Suite of 30 Sep 04

LLNL Forensics ID	DTRA Serial Number*	Seal Number	Date	Location
04-16-1-A	A-5001	3215	30-Sep-04	N 31 55.6426 W 80 55.0664**
04-16-2-A	A-5002	3216	30-Sep-04	N 31 55.5887 W 80 55.0496
04-16-3-A	A-5003	3214	30-Sep-04	N 31 55.5740 W 80 55.2300
04-16-4-A	A-5004	3217	30-Sep-04	N 31 55.5962 W 80 55.0104
04-16-5-A	A-5005	3212	30-Sep-04	N 31 55.657 W 80 55.142
04-16-6-A	A-5006	3218	30-Sep-04	N 31 55.5043 W 80 55.0580
04-16-7-A	A-5007	3211	30-Sep-04	N 31 55.685 W 80 55.359
04-16-8-A	A-5008	3219	30-Sep-04	N 31 55.6168 W 80 55.1069
04-16-9-A	A-5009	3213	30-Sep-04	N 31 55.6318 W 80 55.0960
04-16-10-A	A-5010	3260	30-Sep-04	N 31 55.5345 W 80 55.0499
04-16-11-A	A-5031	3261	30-Sep-04	N 31 55.4920 W 80 55.1726
04-16-12-A	A-5032	3262	30-Sep-04	N 31 55.6034 W 80 55.1881

* Note the break in numerical sequence from #5010 to #5031, 5032.
** The location written for A-5001 was on the original collection label as W81 (Photo #2); corrected to W80 here with the belief that the collections were closer in proximity.

Photographs 1 and 2:
DTRA sediment collections upon receipt at LLNL Forensics Science Center.

Analytic Method:

Collection subdivision and sample preparation began on 8 Nov 04. Uranium purification chemistry and mass spectrometry analysis delivered preliminary results on all samples 10 Nov 04, and final results 12 Nov 04. Final gamma spectroscopy results were delivered 10 Nov 04, with one extended-duration assay completed on 15 Nov 04.

Mass spectrometry: The isotopic composition of uranium in the collections was determined by plasma-source mass spectrometry after radiochemical processing of both aqueous and solid collection fractions. Criteria for presence of anthropogenic uranium (depleted or enriched) were (1) a statistically-significant deviation in the $^{238}U/^{235}U$ atom ratio from the invariant natural ratio, or (2) the presence of ^{236}U, which does not occur naturally.

A portion of each sediment collection was centrifuged and the aqueous portion separated from the solid fraction. The solid portions were leached with acid to extract all adsorbed uranium content into solution (leachate). Leaching the solid fraction, as opposed to fully dissolving it in strong acids, will favor extracting any deposited anthropogenic uranium together with some indeterminate fraction of the natural uranium indigenous to the sediment. For present purposes, full dissolution would only raise the level of natural uranium against which we are looking for small deviations. Leaching optimizes the chance of discerning any local source signature and avoids time-consuming dissolutions. (Chemistry processing details are available in the Annex.)

Uranium separations and purification chemistries were performed on each collection's aqueous and leachate fractions. Aqueous fractions were processed first, and leachate fractions of solid sediment were processed second. The dozen samples in each sample suite were accompanied by a process blank sample.

Purified uranium samples were assayed by mass spectrometry for ratios of $^{234}U/^{235}U$, $^{236}U/^{235}U$, and $^{238}U/^{235}U$. Since the criteria involve departure from natural uranium isotopic composition, all radiochemical samples were processed as unspiked; therefore, the collections' total uranium content was not determined.

Four aqueous samples had replicate analyses performed, and these agreed within analytical uncertainty with the corrected values for the first analyses. Leachate samples and replicate analyses of aqueous samples were sufficient to determine that no ^{236}U was detected outside of measurement uncertainties (see below).

Uranium sample matrix issue: The initial suite of twelve aqueous samples displayed one- to two-sigma departures from the natural $^{238}U/^{235}U$ ratio (equal to 137.88 with some 50 ppm uncertainty). This was judged to reflect sample elemental impurities persisting in the processed uranium samples and affecting the mass spectrometer plasma ion source.

Four aqueous samples (#1, 2, 7, and 9) were chosen for replicate analysis with the uranium purification steps performed twice. These sediment samples were found to benefit slightly from double purification, since replicate results show an improved agreement with natural uranium isotopic composition.

Gamma spectroscopy: To determine any degree of SRS regional background present in the collections, gamma spectroscopy of solid collection fractions assayed for ^{137}Cs content. In this geographic area, elevations in ^{137}Cs beyond natural marine levels have been determined to be a reliable indicator of the regional background from the SRS (several such articles are available in published literature, see References 1-9).

Had there been any observed departures from natural uranium, ^{137}Cs assay levels would become important in determining whether the cause was the SRS regional background or an actual local source. Notionally, however, any SRS effluent present would be expected across the collection suite, whereas a localized source could stand out more apparently.

Analytic Results:

Uranium mass spectrometry results for aqueous and leachate portions of the sediment collections are available as final data (see Tables 2 and 3, and Charts 1 to 4). Gamma spectroscopy results are also available for all twelve collections (see Tables 4 and 5, and Charts 5 to 9).

Natural uranium isotopic variability: The ^{235}U/^{238}U atom ratio of natural uranium, with the exception of uranium associated with the Oklo natural reactor, is considered to be constant (see References 11 and 14).

Variability in ^{234}U/^{238}U has been long known and used to study geochemical processes. Two publications are devoted to uranium disequilibria, and discuss ^{234}U/^{238}U in rivers, estuaries and seawater (see References 10 and 13).

Savannah Survey Results: The observed ^{234}U/^{235}U, ^{238}U/^{235}U, and ^{236}U/^{235}U ratios for all twelve collections form a consistent suite of data, with no one collection or set of collections standing apart outside of uncertainties. The range of values observed for the aqueous fractions is consistent with natural seawater uranium composition with a ^{234}U/^{238}U activity ratio of ~ 1.14. The range of values observed for the leached fractions is consistent with natural ocean sediment uranium composition with ^{234}U/^{238}U activity ratio of ~ 1. No variations from natural uranium isotopic composition outside the limits of analytical uncertainty were detected for any sample for ^{234}U/^{235}U, ^{236}U/^{235}U or ^{238}U/^{235}U. In Tables 2 and 3, "not detected" for ^{236}U reflects that no mass spectrometer counts were detected above instrumental background. (See References 10-15 regarding natural uranium isotopic values and ranges.)

The absence of variation in the ^{238}U/^{235}U ratio can be used to set an upper limit on the possible presence of anthropogenic uranium in the collections. A simple, two-component mixing model incorporating natural uranium (^{238}U/^{235}U = 137.88) and depleted uranium (^{238}U/^{235}U = 500) end-members was used to evaluate the uranium isotope compositions measured in the collections. The only free parameter in the model is the concentration of the depleted uranium component. The results of this calculation constrain the amount of anthropogenic uranium contained in the collections to be less than from 0.31 – 0.46% of the total uranium content, depending on the specific sample. These results provide a conservative (three-standard-deviation) upper limit to the fraction of the total uranium in the collections that could be associated with an anthropogenic source. See References 10-15 for background information.

(Continues on Page 9.)

Table 2. Final results for Savannah Survey samples – Aqueous fractions – Uranium Mass Spectrometry

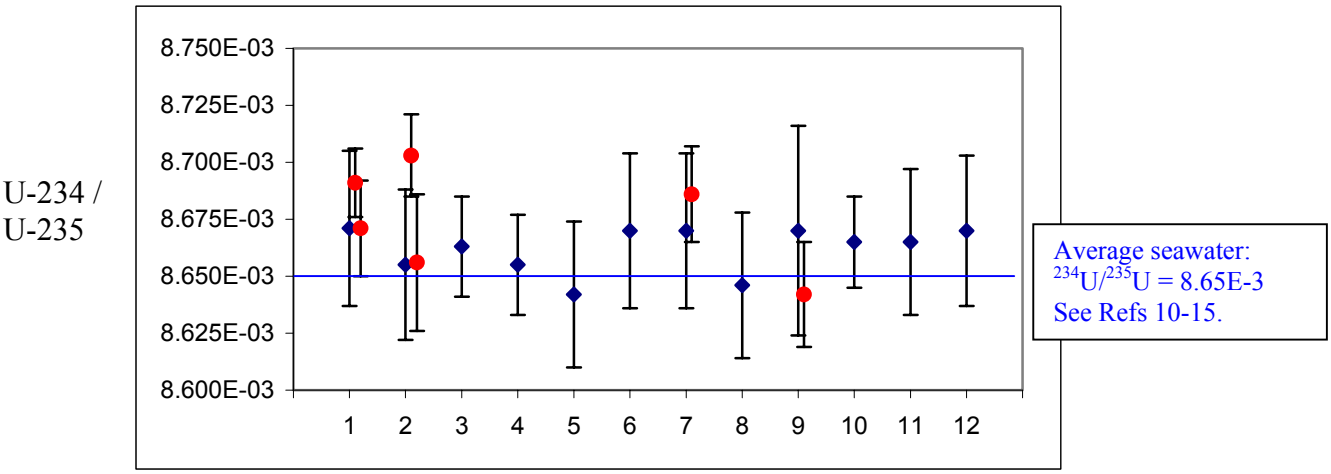
			Uranium Atomic Ratios						
LLNL Forensics ID	DTRA Serial Number	Run No.	**234/235**	**1-σ**	**236/235**	**1-σ**	**238/235**	**1-σ**	**Notes**
04-16-1-A-1	A-5001	1	8.671 E-03	3.4 E-05			1.3746 E+02	2.8 E-01	A
		2	8.691 E-03	1.5 E-05	Not Detected	-----	1.3774 E+02	1.0 E-01	
		3	8.671 E-03	2.1 E-05	Not Detected	-----	1.3789 E+02	1.0 E-01	B
04-16-2-A-1	A-5002	1	8.655 E-03	3.3 E-05			1.3763 E+02	2.8 E-01	A
		2	8.703 E-03	1.8 E-05	Not Detected	-----	1.3761 E+02	1.0 E-01	
		3	8.656 E-03	3.0 E-05	Not Detected	-----	1.3804 E+02	1.0 E-01	B
04-16-3-A-1	A-5003	1	8.663 E-03	2.2 E-05	Not Detected	-----	1.3770 E+02	1.0 E-01	
04-16-4-A-1	A-5004	1	8.655 E-03	2.2 E-05	Not Detected	-----	1.3780 E+02	1.0 E-01	
04-16-5-A-1	A-5005	1	8.642 E-03	3.2 E-05			1.3799 E+02	2.7 E-01	A
04-16-6-A-1	A-5006	1	8.670 E-03	3.4 E-05			1.3764 E+02	2.8 E-01	A
04-16-7-A-1	A-5007	1	8.670 E-03	3.4 E-05			1.3782 E+02	2.7 E-01	A
		2	8.686 E-03	2.1 E-05	Not Detected	-----	1.3791 E+02	1.0 E-01	B
04-16-8-A-1	A-5008	1	8.646 E-03	3.2 E-05			1.3766 E+02	2.7 E-01	A
04-16-9-A-1	A-5009	1	8.670 E-03	4.6 E-05			1.3785 E+02	4.0 E-01	A
		2	8.642 E-03	2.3 E-05	Not Detected	-----	1.3781 E+02	1.0 E-01	B
04-16-10-A-1	A-5010	1	8.665 E-03	2.0 E-05	Not Detected	-----	1.3769 E+02	1.0 E-01	
04-16-11-A-1	A-5031	1	8.665 E-03	3.2 E-05			1.3769 E+02	2.7 E-01	A
04-16-12-A-1	A-5032	1	8.670 E-03	3.3 E-05			1.3765 E+02	2.7 E-01	A

Notes

A: Process blank correction was required for these analyses, based on $^{236}U/^{235}U$, and no ^{236}U assay was possible. Collections #1, 2, 7, and 9 had replicate analyses displaying no ^{236}U content.

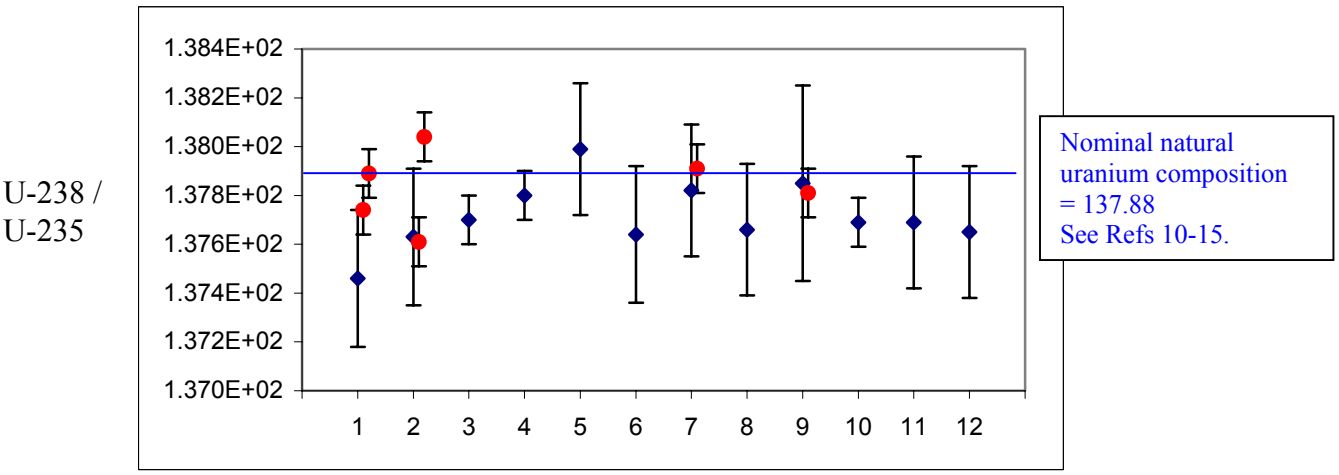
B: Uranium purification was performed twice, to reduce observed effects from the sediment matrix on the mass spectrometer assay.

Chart 1. Savannah Survey samples – Aqueous fractions – ^{234}U / ^{235}U by Mass Spec



* Data points shown in red reflect re-analyses as described in the text.

Chart 2. Savannah Survey samples – Aqueous fractions – ^{238}U / ^{235}U by Mass Spec



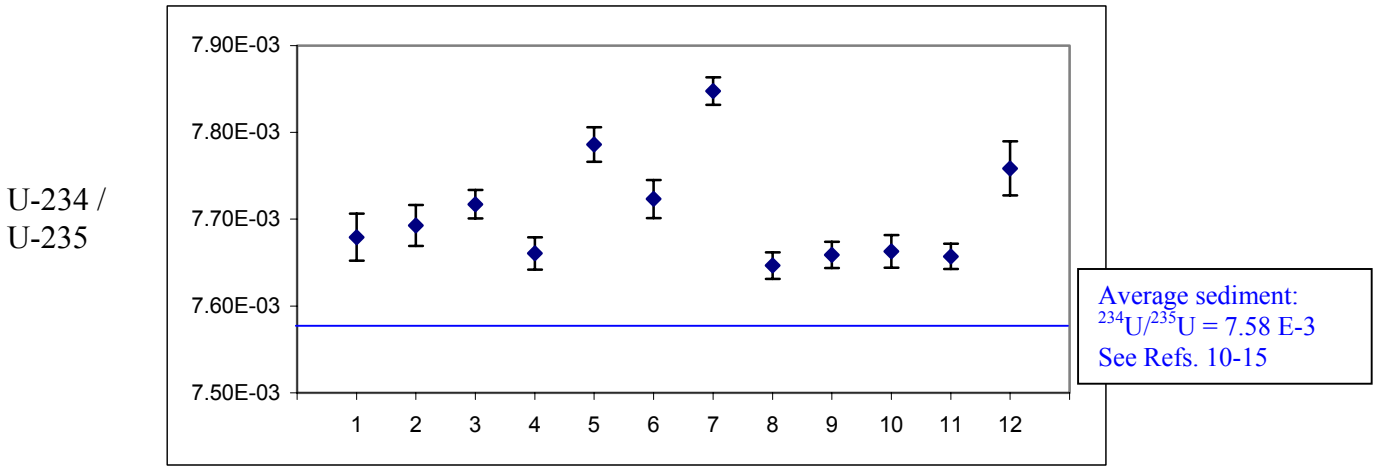
* Data points shown in red reflect re-analyses as described in the text.

Table 3. Final results for Savannah Survey samples – Leachate fractions – Uranium Mass Spectrometry

			Uranium Atomic Ratios						
LLNL Forensics ID	**DTRA Serial Number**	**Run No.**	**234/235**	**1-σ**	**236/235**	**1-σ**	**238/235**	**1-σ**	**Notes**
04-16-1-A-3-A	A-5001	3215	7.679 E-03	2.7 E-05	Not Detected	-----	1.3797E+02	1.0E-01	
04-16-2-A-3-A	A-5002	3216	7.693 E-03	2.4 E-05	Not Detected	-----	1.3801E+02	1.0E-01	
04-16-3-A-3-A	A-5003	3214	7.717 E-03	1.6 E-05	Not Detected	-----	1.3790E+02	1.0E-01	
04-16-4-A-3-A	A-5004	3217	7.661 E-03	1.9 E-05	Not Detected	-----	1.3794E+02	1.0E-01	
04-16-5-A-3-A	A-5005	3212	7.786 E-03	2.0 E-05	Not Detected	-----	1.3787E+02	1.0E-01	
04-16-6-A-3-A	A-5006	3218	7.723 E-03	2.2 E-05	Not Detected	-----	1.3776E+02	1.0E-01	
04-16-7-A-3-A	A-5007	3211	7.848 E-03	1.6 E-05	Not Detected	-----	1.3785E+02	1.0E-01	
04-16-8-A-3-A	A-5008	3219	7.647 E-03	1.5 E-05	Not Detected	-----	1.3797E+02	1.0E-01	
04-16-9-A-3-A	A-5009	3213	7.659 E-03	1.5 E-05	Not Detected	-----	1.3781E+02	1.0E-01	
04-16-10-A-3-A	A-5010	3260	7.663 E-03	1.9 E-05	Not Detected	-----	1.3783E+02	1.0E-01	
04-16-11-A-3-A	A-5031	3261	7.657 E-03	1.5 E-05	Not Detected	-----	1.3784E+02	1.0E-01	
04-16-12-A-3-A	A-5032	3262	7.758 E-03	3.1 E-05	Not Detected	-----	1.3805E+02	1.0E-01	

* All leachate uranium purifications were performed with single purification steps.

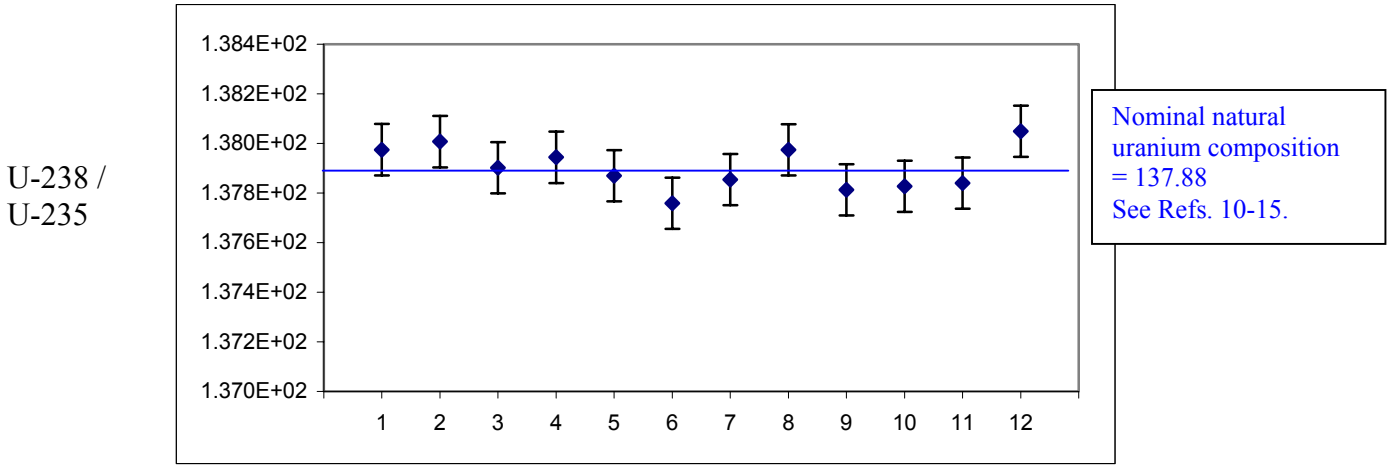
Chart 3. Savannah Survey samples – Leachate fractions – ^{234}U / ^{235}U by Mass Spec



Comment: These leachate fractions are somewhat higher in ^{234}U / ^{238}U content than average sediment because they contain an indeterminate fraction of seawater, with its higher ^{234}U / ^{238}U ratio of 8.65 E-3. See Chart 1 above. This is entirely anticipated and does not reflect any local source of non-natural uranium.

Overall, ^{234}U content was not adopted as a criterion for detection of non-natural uranium for these exact reasons of its natural variability.

Chart 4. Savannah Survey samples – Leachate fractions – ^{238}U / ^{235}U by Mass Spec



(Continues from Page 4.)

Gamma spectroscopy results for solid-fraction portions of all twelve collections are displayed in Tables 4 and 5, and Charts 5 through 9. All detected nuclides are those commonly found in natural soil decay chains (Bi-213, Ra-226, and Th-228) and the naturally occurring K-40. The observed variation is expected for sediment collections such as these. See Refs 1-9.

No evidence was found for detection of any fission products, including ^{137}Cs. Minimum detection limits of ^{137}Cs were estimated for the spectra in hand, and these limits were all found to be lower than 10 Bq/kg, which is the threshold for non-natural ^{137}Cs content in the Savannah River estuary (see References 1-9).

Chart 10 displays the longest-duration gamma spectrum, taken of Collection #A-5001.

Table 4. Final results for Savannah Survey – Solid fractions – Gamma Spectroscopy

LLNL Forensics ID	DTRA Sample ID	Weight (Grams)	NUCLIDE	Bq/Kg AT ZERO TIME	PCT ERROR
04-16-1-A-2	1	19.05	BI X	4.966E+00	22.1
			K 40	1.264E+02	30.9
			RA 226	2.732E+01	17.1
			TH 228	9.676E+00	39.8
04-16-2-A-2	2	23.42	K 40	1.320E+02	44.5
			RA 226	1.282E+01	38.3
04-16-3-A-2	3	21.32	BI X	8.412E+00	19.3
			RA 226	4.076E+01	13.0
			TH 228	1.882E+01	23.7
04-16-4-A-2	4	21.57	BI X	2.225E+00	32.0
			K 40	1.109E+02	19.3
			PO X	8.221E-01	82.1
			RA 226	1.613E+01	8.7
			TH 228	8.329E+00	15.3
04-16-5-A-2	5	16.14	BI X	1.077E+01	7.8
			K 40	1.145E+02	18.7
			RA 226	3.327E+01	5.4
			RA 228	2.181E+01	16.2
			TH 228	2.439E+01	6.2
04-16-6-A-2	6	19.90	BI X	6.220E+00	9.4
			K 40	1.914E+02	17.6
			RA 226	1.997E+01	8.9
			RA 228	1.859E+01	16.6
			TH 228	2.389E+01	8.1
04-16-7-A-2	7	19.77	BI X	2.003E+00	21.1
			RA 226	8.265E+00	19.3
			TH 228	4.196E+00	36.5
04-16-8-A-2	8	21.82	K 40	1.363E+02	18.3
			RA 226	1.056E+01	14.2
			TH 228	1.092E+01	11.5
04-16-9-A-2	9	20.34	RA 226	1.510E+01	28.5
			TH 228	1.013E+01	34.6
04-16-10-A-2	10	20.85	RA 226	1.489E+01	29.2
			TH 228	1.532E+01	22.7
04-16-11-A-2	11	16.33	RA 226	2.917E+01	25.7
04-16-12-A-2	12	13.16	TH 228	2.205E+01	28.9

Chart 5. Savannah Survey samples – Solid fractions – Bi-213 by Gamma Spec

Bismuth-213
(Bq/kg)

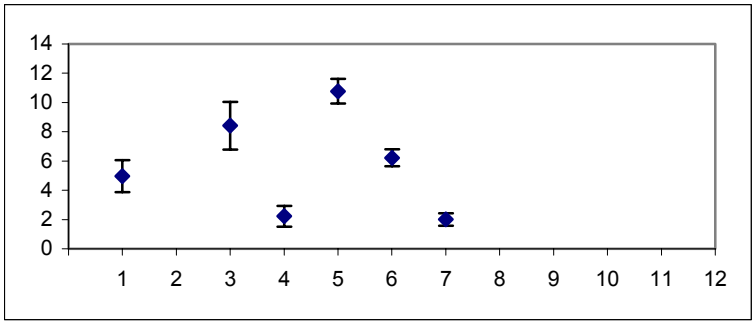


Chart 6. Savannah Survey samples – Solid fractions – K-40 by Gamma Spec

Potassium-40
(Bq/kg)

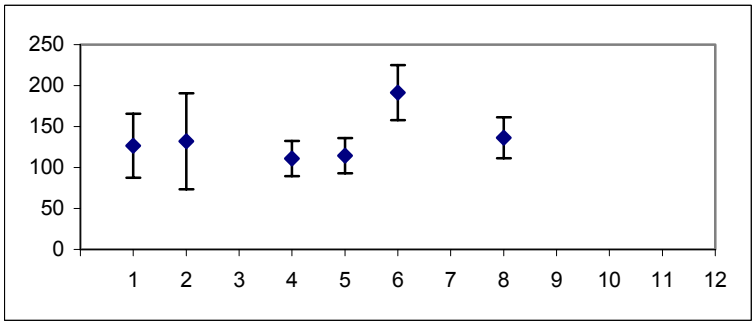


Chart 7. Savannah Survey samples – Solid fractions – Ra-226 by Gamma Spec

Radium-226
(Bq/kg)

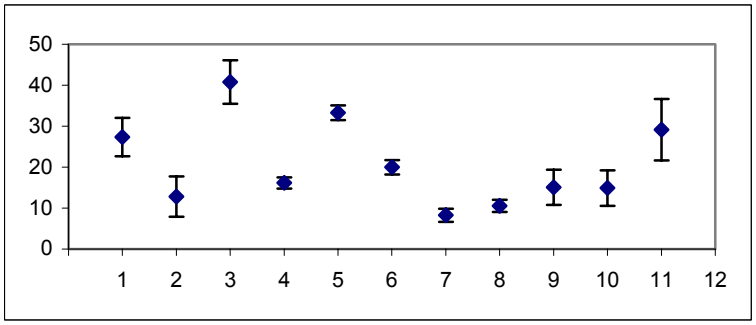


Chart 8. Savannah Survey samples – Solid fractions – Th-228 by Gamma Spec

Thorium-228
(Bq/kg)

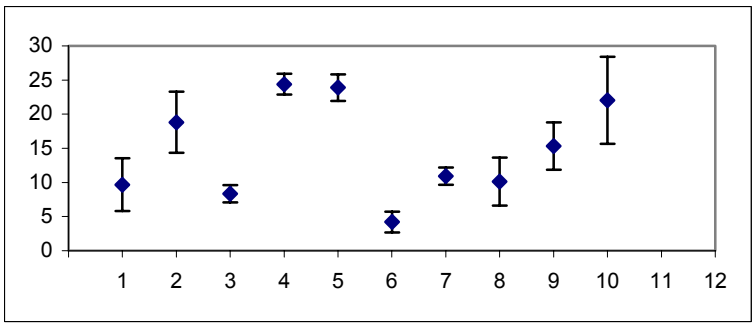
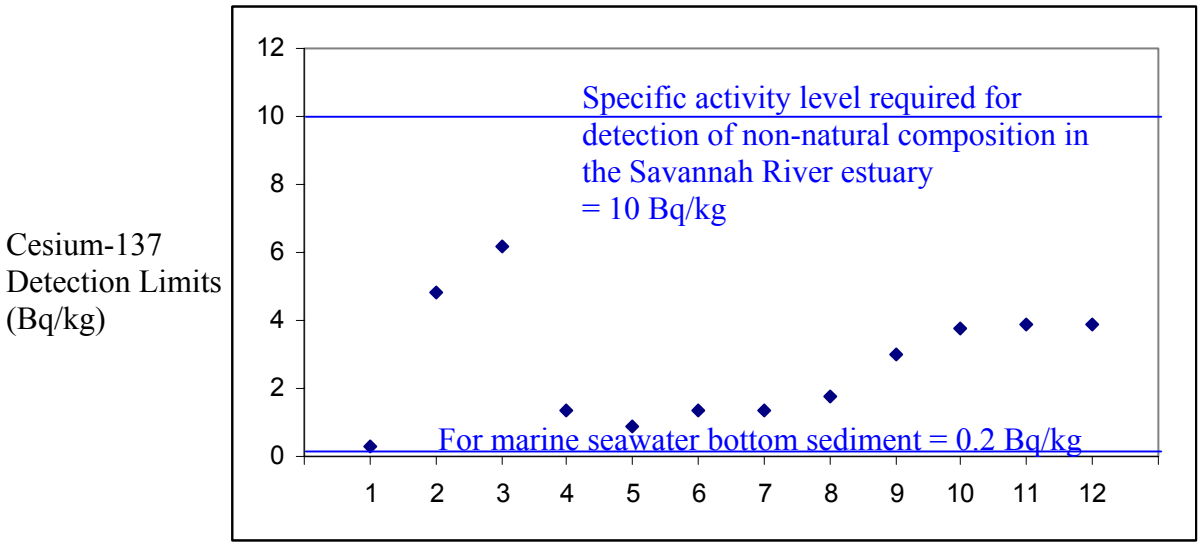


Table 5. Savannah Survey Samples – ^{137}Cs detection limits – no actual detection.

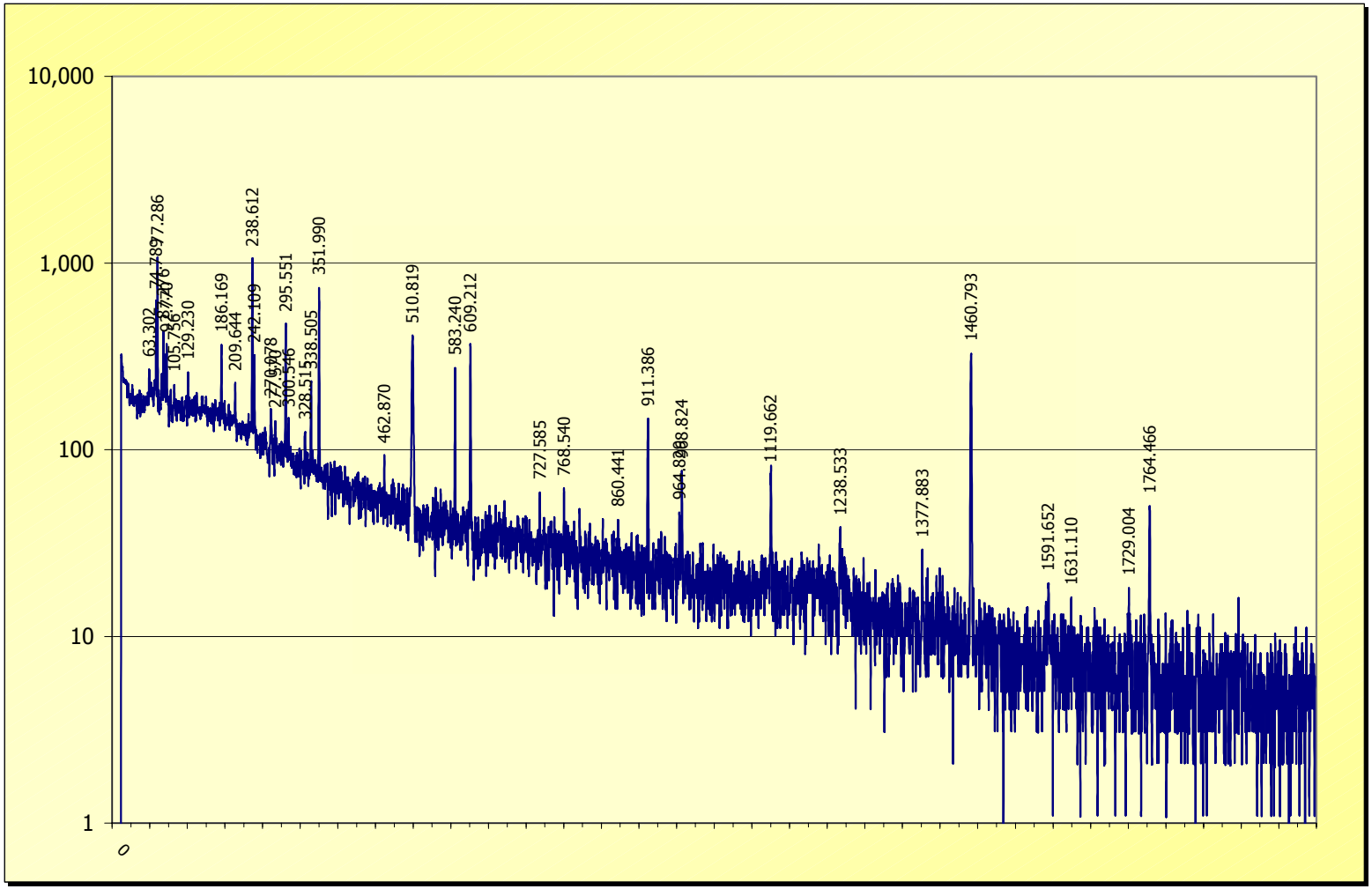
LLNL Forensics ID	DTRA Serial Number	Cs-137 Detection Limit (Bq/kg)	Comments
04-16-1-A	A-5001	0.3	Longest duration count
04-16-2-A	A-5002	4.8	
04-16-3-A	A-5003	6.2	
04-16-4-A	A-5004	1.3	
04-16-5-A	A-5005	0.9	
04-16-6-A	A-5006	1.4	
04-16-7-A	A-5007	1.3	
04-16-8-A	A-5008	1.8	
04-16-9-A	A-5009	3.0	
04-16-10-A	A-5010	3.8	
04-16-11-A	A-5031	3.9	
04-16-12-A	A-5032	3.9	

Chart 9. Savannah Survey Samples – ^{137}Cs detection limits – no actual detection.



Comment: In References 1-9, ^{137}Cs assays are reported for environmental levels in marine seawater. Bottom sediment content is approximately 0.2 Bq / kg, and suspended sediment is up to approximately 10 Bq / kg for the Savannah River marine environment. The difference is due to natural physical processes. Significantly, the SRS effluent content of ^{137}Cs observed farther up the estuary ranges from 50-100 Bq / kg. Since all these Savannah Survey collections are well below the action level of natural ^{137}Cs in suspended marine sediment, there is no evidence for SRS effluent in the collections on this basis.

Chart 10. Intrinsic gamma emission spectrum of Collection A-5001. Long duration spectrum with high-purity germanium spectroscopy, displays the presence of anticipated natural sediment nuclides: Radium-226 and daughters, Thorium-228 and daughters, and Potassium-40. No evidence for any SRS effluent signatures, either fission products or actinides, and the absence of detectable Cesium-137 at 661.62 keV was especially noted.



Annex: Radiochemical Sample Preparation for Savannah Survey Samples

Samples were submitted to the LLNL Forensic Science Center for photographic documentation and the initiation of chain of custody. Approximately 20 cc of sediment and at least 5 mL of overlying water was transferred to a 50 mL centrifuge tube and spun (04-16-n-A). The overlying water (04-16-n-A-1) was decanted into new, clean Teflon vials and passed through uranium purification chemistry prior to analysis by MCICP-MS (IsoProbe). Any excess water left in the 20 cc tube was archived for future use (04-16-n-A-4). In order to reduce the reported uncertainty in the results, the archived overlying water from samples A-5001 and A-5002 (04-16-n-A-4-A) was prepared and analyzed a second time and the results are included in the report. Due to a possible matrix effect on the isotopic measurements, a third sample was taken from the archived overlying water from jars A-5001, A-5002, A-5007, and A-5009 (04-16-n-A-4-B) and purified twice prior to analysis. These results are included in the report.

Approximately 10 cc of the sediment remaining after the water fraction was removed from the 50 mL centrifuge tube was transferred to a Prindle vial (04-16-n-A-2) and submitted for gamma spectroscopy analysis. The results of this analysis have been reported.

The remaining sediment in the 50 mL centrifuge tube was leached with 10 mL of 8M HNO_3, spun, and the leach solution removed. The sediment was then leached a second time with 10 mL of 8M HNO_3, spun and the leach solution consolidated with the first (04-16-n-A-3). A 5 mL aliquot of the leach solution (04-16-n-A-3-A) was then transferred to new, clean Teflon and passed once through uranium purification chemistry prior to analysis by MCICP-MS (IsoProbe). The results of this analysis are included.

References on Savannah River Site Effluent in the Savannah River and Estuary:

1. Beals D. M. and Hayes D. W. (1995) Technetium-99, iodine-129 and tritium in the waters of the Savannah River Site. *Science of the Total Environment* **173**(1-6), 101-115.
2. Beasley T. M., Elmore D., Kubik P. W., and Sharma P. (1992) Chlorine-36 releases from the Savannah River Site nuclear fuel reprocessing facilities. *Ground Water* **30**(4), 539–548.
3. Carlton W. H., Murphy C. E., and Evans A. G. (1996) Plutonium in the Savannah River Site environment. *Health Physics* **71**(3), 290-299.
4. Carlton W. H., Murphy C. E., Jannik G. T., and Simpkins A. A. (1999) Radiostrontium in the Savannah River Site environment. *Health Physics* **77**(6), 677-685.
5. Carlton W. H., Murphy J., C. E., and Evans A. G. (1994) Radiocesium in the Savannah River Site environment. *Health Physics* **67**(September), 233–244.
6. Goldberg E. D., Griffin J. J., Hodge V., Koide M., and Windom H. (1979) Pollution history of the Savannah River estuary. *Environmental Science and Technology* **13**, 588–594.
7. Hayes D. W. and Sackett W. M. (1987) Plutonium and Cesium Radionuclides in Sediments of the Savannah River Estuary. *Estuarine Coastal and Shelf Science* **25**(2), 169-174.
8. Mulholland P. J. and Olsen C. R. (1992) Marine origin of Savannah River Estuary sediments: evidence from radioactive and stable isotope tracers. *Estuarine Coastal and Shelf Science* **34**(1), 95-107.
9. Olsen C. R., Thein M., Larsen I. L., Lowry P. D., Mulholland P. J., Cutshall N. H., Byrd J. T., and Windom H. L. (1989) Plutonium, lead-210, and carbon isotopes in the Savannah Estuary: riverborne versus marine sources. *Environmental Science & Technology* **23**(12), 1475-1481.

References on Natural Uranium Composition:

10. Bourdon, B., G. M. Henderson, et al., Eds. (2003). Uranium-Series Geochemistry. Reviews in Mineralogy & Geochemistry. Wash DC, Mineral. Society of America.
11. Cowan, G. A. and H. A. Adler (1976). "The variability of the natural abundance of ^{238}U." Geochimica et Cosmochimica Acta 40: 1487-1490.
12. Dai, M., J. M. Kelley, et al. (2002). "Sources and migration of plutonium in groundwater at the Savannah River Site." Environmental Science & Technology 36(17): 3690-3699.
13. Ivanovich, M. and R. S. Harmon, Eds. (1992). Uranium-Series Disequilibrium: Applications to Earth, Marine, and Environ. Sciences. Oxford, Clarendon Press.
14. Steiger, R. H. and E. Jager (1977). "Subcommission on geochronology: Convention on the use of decay constants in geo- and cosmochronology." Earth and Planetary Science Letters 36(359-362).
15. Windom, H., R. Smith, et al. (2000). "Uranium in rivers and estuaries of globally diverse, smaller watersheds." Marine Chemistry 68(4): 307-321.