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Accelerator Mass Spectrometric Measurements of Uranium-236 Associated with Potential Workplace Intakes of Anthropogenic Uranium

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Background

Why Interest in uranium isotopes!

- **Emerging needs in a number of different fields**
  - **Human health**
  - **Nuclear forensics**
  - **Environmental assessments**
- **Advances in measurement technologies, especially in relation to technologies based on mass spectrometry**

Analytical & Nuclear Chemistry Division MC ICP-MS

Accelerator Mass Spectrometry

Applications:

- Routine measurements of rare/ stable isotope ratios and rare isotope sensitivities atoms; e.g., very well developed for $^{14}C/^{12}C$.
- More recently demonstrated for long-lived radionuclides such as the actinides, ^{129}I, and ^{99}Tc
- AMS heavy-element line at LLNL designed specifically for low-level detection of actinide elements–now used routinely for Pu (and U) isotope measurements at sensitivities of 10^5–10^6 atoms.

Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratory

Features of the heavy element AMS system at CAMS;

- Rapid electrostatic switching between masses of interest
- High abundance sensitivity and a very wide dynamic range
- Simple chemistry and relatively high sample through-put (cost efficiencies)
- Very robust measurement technology

HVEC Model FN Tandem Van de Graaff accelerator operated at voltages up to 9 MV: gas or foil stripping

Uranium isotopes in the environment

Isotope	Atom%	Half-life (years)	Origin
^{234}U	0.005	4.468×10^5	^{238}U decay chain
^{235}U	0.72	2.34×10^7	Primordial
^{238}U	99.27	7.038×10^8	Primordial
^{236}U	ca. 10^{-16}	2.446×10^5	$^{235}U + 1n \rightarrow {}^{236}U$

^{236}U measurements in bioassay samples

Reagent Blank Data (N= 20)

Mean = 4.9×10^{-7} +/- 2.4×10^{-7} ng

Limit of Detection ~ 7×10^{-7} ng

c/w ^{238}U ~ 0.2 ng; ^{235}U 0.04 ng
based on MC ICP-MS

Sample ID	^{236}U, ng	1-σ
Reagent Blank_1	6.7E-07	2.9E-07
Reagent Blank_2	7.9E-07	2.8E-07
Reagent Blank_3	1.0E-07	1.1E-07
Reagent Blank_4	7.5E-07	2.7E-07
Reagent Blank_5	4.2E-07	1.9E-07
Reagent Blank_6	3.5E-07	1.8E-07
Reagent Blank_7	2.4E-07	1.7E-07
Reagent Blank_8	9.5E-07	2.9E-07
Reagent Blank_9	7.9E-07	3.0E-07
Reagent Blank_10	6.4E-07	2.9E-07
Reagent Blank_11	6.1E-07	2.9E-07
Reagent Blank_12	7.1E-07	2.9E-07
Reagent Blank_13	3.6E-07	2.2E-07
Reagent Blank_14	2.7E-07	2.4E-07
Reagent Blank_15	2.8E-07	2.4E-07
Reagent Blank_16	4.5E-07	2.4E-07
Reagent Blank_17	6.1E-07	2.5E-07
Reagent Blank_18	1.7E-07	2.2E-07
Reagent Blank_19	2.7E-07	2.1E-07
Reagent Blank_20	4.6E-07	2.4E-07

Baseline ^{236}U in LLNL worker cohort

Sample ID	ng ^{236}U per sample	1-σ	ng ^{236}U per gram	1-σ
70250	1.7E-07	3.0E-07	1.8E-09	3.3E-09
70252	4.6E-07	3.4E-07	8.2E-09	5.9E-09
70254	3.6E-07	3.6E-07	6.7E-09	6.8E-09
70256	3.5E-07	3.7E-07	3.8E-09	4.0E-09
70293	1.2E-07	3.0E-07	1.2E-09	3.0E-09
70386	2.6E-07	3.7E-07	4.6E-09	6.6E-09
70392	1.3E-07	3.0E-07	2.4E-09	5.7E-09
70396	3.9E-07	3.7E-07	5.0E-09	4.7E-09

LLNL rad worker cohort with a known or suspected workplace intake of uranium

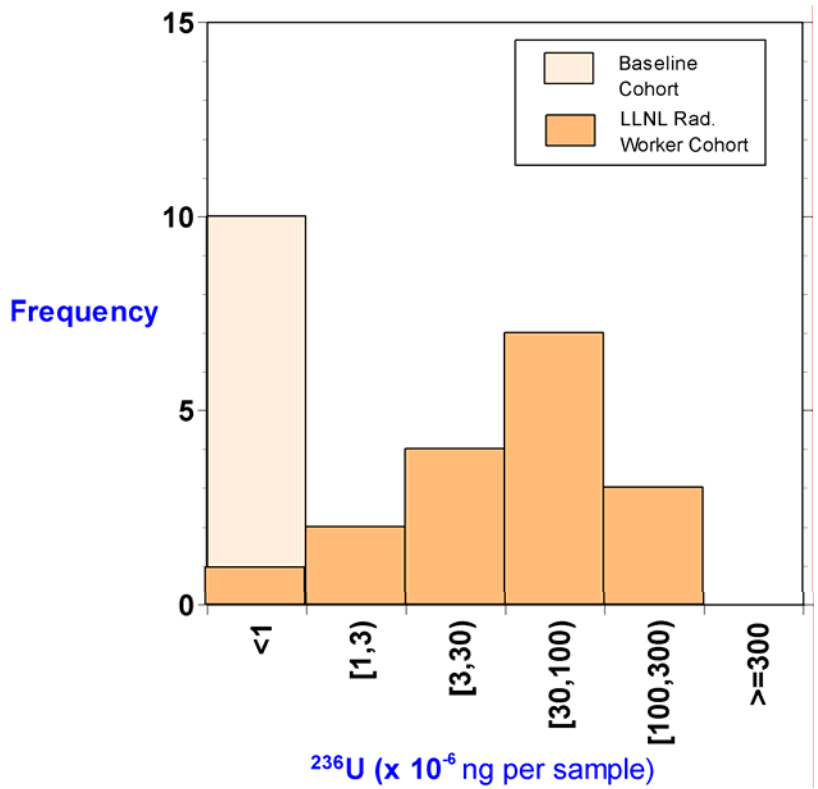
Sample ID	ng ^{236}U per sample	1-σ	ng ^{236}U per gram	1-σ
66479	1.7E-04	6.7E-06	3.2E-06	1.3E-07
66480	1.5E-06	5.5E-07	2.0E-08	7.4E-09
66783	4.6E-06	7.5E-07	5.9E-08	9.6E-09
66834	8.4E-05	3.9E-06	1.1E-06	4.9E-08
66242	4.5E-06	7.5E-07	5.0E-08	8.5E-09
66241	8.0E-05	5.1E-06	3.9E-07	2.5E-08
68860	3.4E-05	2.7E-06	1.6E-07	1.3E-08
69360	2.5E-04	9.1E-06	1.5E-06	5.3E-08
68236	9.8E-06	1.2E-06	7.5E-08	9.0E-09
69763	5.4E-05	3.0E-06	4.3E-07	2.3E-08
68611	<7E-07		<4E-08	
67608	6.3E-06	9.5E-07	3.1E-08	4.6E-09
67982	1.6E-05	1.5E-06	5.8E-08	5.6E-09
69489	2.7E-06	6.7E-07	3.2E-08	7.9E-09
69838	1.3E-04	6.2E-06	1.6E-06	8.0E-08
69840	3.8E-05	2.6E-06	1.9E-07	1.3E-08
70022	4.9E-05	3.0E-06	6.1E-07	3.7E-08

Total U versus ^{236}U in LLNL worker cohort with a known or suspected workplace intake

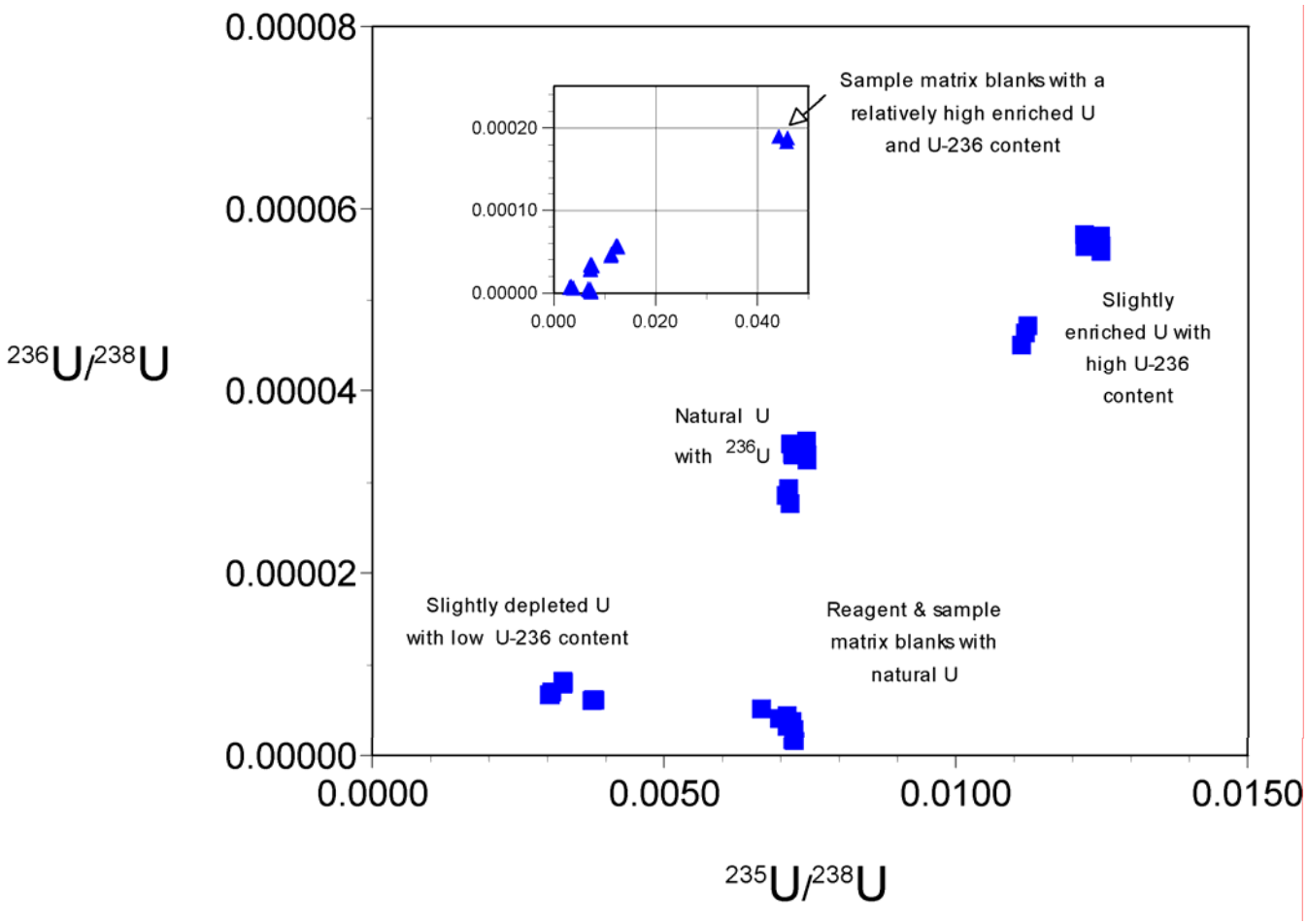
Sample ID	ng ^{235}U per sample (ICP-MS)	1-σ	ng ^{238}U ng per sample (ICP-MS)	1-σ	Total ng U per sample (ICP-MS)	1-σ	ng ^{236}U per sample (AMS)	1-σ
66479	<0.0007		7.2	0.1	7.2	0.1	1.7E-04	6.7E-06
66480	<0.0007		0.0	0.1	0.0	0.1	1.5E-06	5.5E-07
66783	0.0196	0.0004	0.2	0.1	0.3	0.1	4.6E-06	7.5E-07
66834	<0.0007		3.8	0.1	3.8	0.1	8.4E-05	3.9E-06
66242	0.0008	0.0002	0.3	0.1	0.3	0.1	4.5E-06	7.5E-07
66241	0.0125	0.0003	5.8	0.1	5.8	0.1	8.0E-05	5.1E-06
68860	0.0009	0.0002	3.0	0.1	3.0	0.1	3.4E-05	2.7E-06
69360	0.0277	0.0005	9.5	0.2	9.6	0.2	2.5E-04	9.1E-06
68236	0.0159	0.0004	2.4	0.1	2.4	0.1	9.8E-06	1.2E-06
69763	0.0233	0.0005	3.4	0.1	3.4	0.1	5.4E-05	3.0E-06
68611	0.0159	0.0004	0.0	0.1	0.0	0.1	<7E-07	
67608	0.0146	0.0004	16.0	0.3	16.0	0.3	6.3E-06	9.5E-07

What do the results show?

- The ^{236}U content of bioassay samples collected from the LLNL rad worker cohort appears to be clearly elevated over those of the baseline cohort
- Data confirms that worker(s) has been previously exposed to an anthropogenic source of uranium containing ^{236}U



Results of intercomparison study on U isotopes in bioassay samples (unpublished)



Conclusions

- AMS offers a robust measurement technique for detection of ^{236}U in bioassay samples at sensitivities of 10^5-10^6 atoms
- The application of ^{236}U in bioassay studies appears to offer new opportunities for improving the standard of occupational safety and risk management at LLNL, and elsewhere around the DOE complex
- ^{236}U is a potentially useful ‘fingerprint’ for assessing the presence of anthropogenic sources of uranium either inside the human body or in the environment