

Title: Comparative Measurements of ^{238}U , ^{235}U , ^{234}U (and ^{236}U) in Soil Samples from the Marshall Islands Using Inductively Coupled Plasma Mass Spectrometry.

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Oral Presentaiton

Abstract:

COMPARATIVE MEASUREMENTS OF ^{238}U , ^{235}U , ^{234}U (AND ^{236}U) IN SOIL SAMPLES FROM THE MARSHALL ISLANDS USING INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY. Terry Hamilton, Yoko Fujikawa¹, Ross Williams², Roger Martinelli, Masahiro Saito¹, Jim Brunk, Kiyoshi Shizuma³, Steven Kehl, and Emi Ikeda¹

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There is growing interest in high precision measurements of U isotopes for applications in human health monitoring, environmental surveillance and nuclear forensics. Isotopic data allows for a more accurate assessment of the origin of U by comparing $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ atom ratios with known natural abundances. The purpose of this study was to develop a measurement technique for U isotope determination in carbonate soils collected from a former nuclear tests site in the Marshall Islands using a multi-collector inductively coupled plasma mass spectrometry (MCICP-MS). The results of an intercomparison exercise were then used to provide independent confirmatory measurements to assess the accuracy and precision of the method compared with that obtainable on a quadrupole instrument (ICP-QMS) previously used for U isotope determination on soils containing residues from nuclear weapons fallout. The intercomparison was conducted with no prior consultation on sample processing and analytical procedures between the two laboratories. A total of 14 soil samples containing an average concentration of 1600 ng g⁻¹ total U were used for the intercomparison. For MCICP-MS, U isotopes were isolated from single 1 gram aliquots of dry soil after total dissolution in HNO₃/H₂O₂/HF. The average internal precision (2 σ) obtained on $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ atom ratios in soils by MCICP-MS was 0.06% and 0.5%, respectively. For ICP-QMS, U isotopes were isolated from 2 to 3 aliquots obtained by total dissolution of 100 mg of soil in HNO₃/HClO₄/HF. Sample consumption was 1.3 ng of U per analysis. The comparative level of precision obtained by ICP-QMS on 16 to 34 replicate analyses was 0.6 and 5 %, respectively. Even so, the average relative bias on $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ atom ratios between instruments was only 0.07% and 0.6 %, respectively, and was less than the internal precision of ICP-QMS itself. Anomalous

$^{235}\text{U}/^{238}\text{U}$ atom ratios were easily distinguishable in soil samples collected from the northern islands of Enewetak Atoll on both instruments. The latter soils also contained detectable concentrations of ^{236}U using MCICP-MS and confirmed the presence of low concentrations of residual anthropogenic U in atoll soils at the test site.

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