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Measurement of Pu Isotopes at Ultra-trace Levels Using Multi-collector ICP-MS

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The discrimination of sources of anthropogenic plutonium in environmental materials requires precise and accurate isotope ratio determination at very low levels of absolute concentration. In this study plutonium isotope ratios have been determined on solutions with concentrations ranging from 50 fg ml^{-1} to 500 pg ml^{-1} using multi-collector ICP-MS (*Micromass IsoProbe*). At these levels it has been possible to determine Pu isotope ratios with a precision of better than 1%. This has been achieved by adding an equal atom ^{236}U - ^{233}U double spike to Pu solutions to correct for drift in signal intensity between peak jump sequences and instrument mass bias during each ratio determination. Analyses were made using less than 1 ml of Pu solution. In the case of the 50 fg ml^{-1} sample (where $^{240}Pu/^{239}Pu$ ~ 0.18) this corresponds to a 239Pu content of ~40 femtograms. $^{240}Pu/^{239}Pu$ can be reproducibly measured to within 0.9% 2SD at 50 fg ml^{-1}, better than 0.35% at 5 pg ml^{-1}, and 0.15% at 100 pg ml^{-1}. Using the same analytical technique we have also measured $^{242}Pu/^{239}Pu$ with a precision better than 2.5% on solutions containing 10 femtograms ^{242}Pu.

Pu concentration	$^{240}Pu/^{239}Pu$	+/-	% 2 SD	n =	Average in-run % 2SE
50 - 100 fg ml^{-1}	0.2232	0.0018	0.79	11	0.58
1 – 4 pg ml^{-1}	0.2250	0.0015	0.65	25	0.19
5 – 10 pg ml^{-1}	0.2256	0.0008	0.35	19	0.11
100 – 600 pg ml^{-1}	0.2258	0.0003	0.12	8	0.05

Table 1: Pu isotope data in relation to total Pu