Methods for ^{137}Cs and ^{239,240}Pu analysis

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Sampling

Artificial radionuclides to be measured (Plutonium isotopes and Cesium-137) are present at ultra trace levels, which makes difficult their measurement and thus forces to collect large volume samples. 100 L water samples were transferred into a plastic barrel and acidified with 32% HCl. 242Pu and stable Be spikes as chemical yield tracers, and an iron carrier solution (FeCl3) were added under constant stirring. After waiting 12 hours in order to get a chemical equilibrium, the pH was then made 9 with NaOH in order to precipitate the iron as $Fe(OH)_3$. This scavenges beryllium and plutonium onto the precipitate while cesium stays in solution. After that the supernatant was then transferred into a second barrel while the precipitate was stored in small plastic bottles for further processing at laboratory. By addition of 65% HNO3, the pH was lowered again and subsequently stable Cs was added as a chemical yield tracer. Then, a pre-weighed sample of ammonium molybdophosphate was added while stirring thoroughly. This produced a yellow precipitate that scavenges Cs. The precipitate was left to settle and then transferred into smaller bottles.

Cs-137

A pre-weighed sample of ammonium molybdophosphate (AMP) was added while stirring thoroughly. This produced a yellow precipitate that scavenges Cs. The precipitate was left to settle and then transferred into vials for gamma spectrometry.

Pu-239,240

The method used to quantify Pu isotopes in sediment samples by ICP-MS at LDEO was modified after Maxwell [1997] and Kenna [2002]. About 2 g of dry sediment were spiked with 242Pu as a yield monitor. Sediments were completely digested using a mixture of HNO3 (65%) and HF (40%) under heating at 180°C in a hot plate. After matrix dissolution, HClO4 (1 mL) was added in order to remove HF by fuming. This step was repeated three times and the solution was evaporated to dryness. Sample was redissolved with 1mL HCl (%) and 5mL d.i. H2O. After that, Pu was co-precipitated with Fe(OH)3, adding concentrated NH4OH. After cooling, the resulting precipitate was centrifuged and the supernate removed. The precipitate was rinsed and re-centrifuged three additional times with deionized water. The final iron precipitate was dissolved in concentrated 1mL HNO3 (65%) and taken to dryness. This step was done 3 times. After that the iron precipitate was dissolved in 40mL 3M HNO3. Prior to ion exchange purification, the solution was prepared to contain Pu in the IV oxidation state by adding 1.3 g of FeSO4·7 H2O and 2.2 mL 4.2 M NaNO2. Ion exchange separation was carried out with TEVA- resins and TEVA-pre-filter resin placed on a vacuum system. The 30 mL 3M HNO3 solution containing the digested sample is loaded to the TEVA resin after its preconditioning with 25 mL of 3M HNO3. The Pu fraction was isolated from the other actinides by washing the column with 9 M HCl (2.5 mL) to remove Th and 8 M HNO3 (15 mL) to remove U. Before Pu was eluted, a pre-filter resin was placed between column and solution

collector to minimize the contribution of other components and then Pu was eluted with a mixture of 0.13 M HF and 0.02 M HNO3 (25 mL). The eluate was evaporated to dryness and redissolved with 7 mL HNO3 (65%) and 2 mL 3M HNO3. Then, it was prepared for a second purification adding 0.65 g of FeSO4·7H2O and 2 ml 4.2M NaNO2 to ensure that Pu was in IV oxidation stage. The radiochemical separation with TEVA-resins was identical to the first step. After collecting the final Pu fraction, three drops of concentrated HClO4 was added to remove the rest of resin, and then the solution was evaporated to almost dryness. The drop was dissolved in 1mL HNO3 (65%) and transferred to small Teflon vial and heated to dryness at 200°C. Finally, 2 mL of an acid mixture 1% HNO3/0.1% HF was added to adjust and transfer the solution to the ICP-MS vial and solution was heated few seconds.

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