Evaluation of a transuranic component in reactor derived releases from Fukushima Dai-ichi to the marine environment

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Abstract

The incident at Fukushima Dai-ichi on March 11, 2011 was the consequence of several events (earthquake, tsunami, equipment failures, etc.) and led to the release of radioactive contaminants into the atmosphere and ocean. Although specific details regarding the mechanism and/or the extent of radioactivity released to the environment are lacking, significant levels of fission/activation products as well as Pu-239, Pu-240, and Np-237 have been measured in the vicinity of Fukushima. As part of a research cruise in June, 2011 to study the problem, we received water samples collected between 30 and 600 km offshore. The main objective of our work is to determine the presence or character a transuranic component in the reactor releases. Here we report results from Pu isotopic analysis of selected samples from the cruise. The Pu isotopic composition is diagnostic in resolving reactor-derived contamination from background levels associated with global fallout. Based on the relationship between the Fukushima derived Pu, 239Pu and 240Pu we estimate the total amount input to the study site from direct release.

RESULTS

Pu isotopes and concentrations in seawater samples

Fig. 2 Map showing the sampling stations of KOK-1108 in relation to Fukushima (red star) and the Kuroshio Current.

Pu isotopes compared to 239Pu and 241Am

Fig. 4 (A) 239Pu vs. 239,240Pu activity. (B) 239,240Pu vs. 137Cs activity. Linear regression indicates a strong correlation between increasing levels of 239Pu and 237Cs and increases in the 239Pu/240Pu atom ratio.

Pu end-member mixing calculations

Based on the measured 237Cs/137Cs activity ratio (~1) the average burn-up of the fuel rods can be estimated to be ~25 gigawatt-days/metric ton of uranium (GWDMU), which in turn corresponds to a 239Pu/240Pu atom ratio ~0.46 (R. Web, personal communication). Using this value as the Fukushima end-member and 0.22 as the global fallout 239Pu/240Pu end-member, we can estimate the relative contributions for each source using a simple two end-member mixing model (eq. 1). Subscripts m, G, and F indicate the measured 239Pu/240Pu value and the global fallout and Fukushima end-member values, respectively.

Pre-Fukushima (2009 NE Pacific data)

1. Determine whether or not there is a transuranic component in the reactor releases.
2. Characterize the isotopic composition (239Pu, 240Pu, and 237Np) separating Pu and Np.
3. Develop a sampling strategy for future studies.

Methods

• Samples collected for UAB Sr/Ca analysis (see map right)
• Modified method of Warpee, J.T., Orlando, K.A. (2010) to separate Pu and Np
• Column fractions sent to LDEO for additional purification and analysis of Pu-239, Pu-240, Np-237 by ICP-MS (modified after Kenna 2002)

CONCLUSIONS

1. Pu isotopic analysis provides evidence of input from a source that is enriched in 239Pu relative to fallout from nuclear weapons tests and consistent with being derived from a nuclear reactor.
2. Application of a two end-member mixing model indicates that Fukushima derived Pu ranges between 0-50% of the total Pu measured in our samples.
3. There is some evidence of subsidence is observed. (C) Enhanced 239+240Pu isotopes attached to large particles through the water column to deep ocean sediments and re-suspension by near-bottom currents.
4. This is likely a lower limit as it does not account for Pu loss due to particle scavenging in the water column.

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