A20150531-9341

Detection of Plutonium-239 as an Alternative Age Constraint in the Antarctic Plateau Shallow Snowpack

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To determine Pu records related to nuclear weapons tests carried out since the 1950s in snow pit samples, we used an Inductively Coupled Plasma-Sector Field Mass Spectrometry (ICP-SFMS), coupled with an Apex high efficiency sample introduction system (Apex HF). A snow pit was dug at a site (77°18′S, 39°47′E, 3785 m a.s.l.) near Dome Fuji in East Antarctica, dating from 1957 to 2007. To avoid contamination during sample preparation, sample handling was carried out under ultraclean conditions; class 10 clean benches in class 1000 clean laboratories at the Korea Polar Research Institute (KOPRI) (Hong et al., 2012). The main advantages of ICP-SFMS technique are rapidity of analysis and simple sample preparation method for ^{239}Pu at femtogram levels in small-volume samples from snow/ice. However, this technique is prone to spectral interferences. The existence of high content of uranium in sample could lead to significant interferences with ^{239}Pu owing to uranium hydride ($^{238}UH^{+}$) formation. We found that the interference effect of $^{238}UH^{+}$ was negligible when the ^{238}U concentrations were lower than 10 pg g^{-1}. It was checked in various concentrations of multi-standard solutions such as 0.1, 0.2, 0.5, 1, 2, 5, 10, 25, 50, and 100 pg g^{-1}. When the ^{238}U concentrations were higher than 10 pg g^{-1}, significant counts for $^{238}UH^{+}$ were measured. In the snow pit samples, the ^{238}U concentrations were lower than 0.5 pg g^{-1}. Accordingly, ^{239}Pu signals were detected without $^{238}UH^{+}$ interference. For the calculation of ^{239}Pu concentration, semi-quantitative method was used. It is based on assumption that the first ionization energy for Pu and U are very close (6.06 eV and 6.19 eV, respectively) and therefore they should have a similar behavior when ionized in the plasma (Gabrieli et al., 2011). An external calibration method was applied for the semi-quantification of ^{239}Pu with ^{238}U in the samples. The concentrations of the standard solution used for the calibration curve were 0.1, 0.2, 0.5, 1, 2, and 10 pg g^{-1}. Detection limits obtained for U and Pu are 0.005 pg g^{-1} and 0.042 fg g^{-1}, respectively. The results from the dilute solutions of riverine water certified reference materials (CRM) (SLRS-5) showed good agreement with the compiled value (Heimburger et al., 2012) (in pg g^{-1}): 101±6 (n=10) versus 93±6(n=25) for U. Consequently, the ^{239}Pu records were reconstructed at femtogram levels in the Antarctic snow pit sample by the application of a semi-quantitative method. These suggest that this method can be widely used for the reconstruction of the fallout history of ^{239}Pu and for the age constraint in other Antarctic Plateau sites.

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