# An alternative age constraint of snowpit at Dome Fuji, Antarctica by plutonium concentrations

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Plutonium is a useful time marker for dating ice cores and snow pits because it in the environment mainly originates from atmospheric nuclear weapons tests carried out since the 1950s. We present temporal history of plutonium-239 deposition on the Dome Fuji and compare that with the nuclear test record and previous results from Antarctica and other regions.

To determine $^{239}Pu$ in snow pit samples, covering ~50 years (1956-2007), at Dome Fuji in East Antarctica (Hong et al., 2012), we used an inductively coupled plasma-sector field mass spectrometry (ICP-SFMS) (Element2, Thermo Finnigan MAT, Germany), coupled with an Apex high efficiency sample introduction system (Apex HF, ESI, USA). The ICP-SFMS 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 $g^{-1}$. 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.

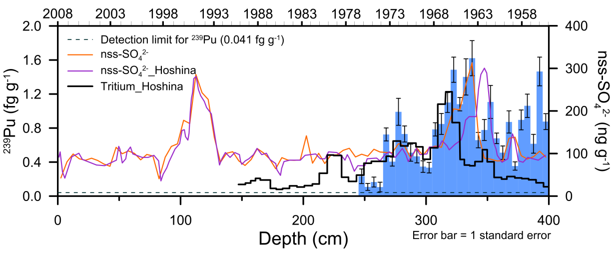


Figure 1. Profiles of Pu concentration and comparison with other chemical signatures. (This study vs Hoshina et al., 2014)

Depth profile of $^{239}Pu$ concentrations is shown in fig. 1. The three major $^{239}Pu$ concentration peaks were found at depths of 390-395 cm, 335-340 cm and 275-280 cm. The $^{239}Pu$ peaks are related to thermonuclear weapon tests by the US (1952-1958), USSR (1961-1962), France (1966-1974) and China (1964-1980), respectively. The $^{240}Pu/^{239}Pu$ ratios which are commonly used as indicators for the source of Pu determined for the pre-moratorium samples are about 0.3 and for the post-moratorium are about 0.2. Between 1970 and 1973 (±1), the $^{240}Pu/^{239}Pu$ ratios are rapidly decreased from 0.2 to 0.07 (± 0.02) and 0.08 (± 0.03). It was related to the French tests which took place at Mururoa and Fantataufa, Tuamotu Archipelago (21°S, 137°W) between 1966 and 1974 (Koide et al., 1985). The profile of $^{239}Pu$ was compared with those of nss-$SO_4^{2-}$ and tritium to examine the compatibility of the different age markers. In the nss-$SO_4^{2-}$ profile, two pronounced peaks were found at depths of 110-115 cm and 335-340 cm; around the shallower peak, attributed previously to the 1991 Pinatubo eruption and the deeper peak appeared together with the $^{239}Pu$ peak dated to the year 1964±1, it was related to the Mt. Agung eruption of March 1963 (Hong et al., 2012). Tritium was a coproduct of the nuclear test and its fallout over the Antarctic Plateau was also enhanced during 1950s-70s. However, its maximum fallout had been observed in 1966 (Hoshina et al., 2014), about 2 (±1) years later than that of $^{239}Pu$. The shift in their peak deposition was attributed to the different phases between tritium in the form of HTO (gas) and plutonium present in the particulates (Jouzel et al., 1979). In Fig. 1, the tritium peak was found at the depth ~10 cm shallower than the $^{239}Pu$ and Agung nss-$SO_4^{2-}$ peaks. Such difference in the peak depth is compatible with the 2-year lag when the surface snow accumulation rate (~8 cm $yr^{-1}$) and its compaction at 3 m depth (~30%) are taken into consideration.

The Antarctic Plateau represents an ideal place to detect $^{239}Pu$ fallout using ICP-SFMS. The low snow accumulation thereon is responsible for the detectable level of $^{239}Pu$ in snowmelt without pre-concentration. In addition, the low U contents in snow minimize the potential isobaric interference of $^{238}UH$ on $^{239}Pu$. 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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