Geochemical Fractionation of ^{239}Np in Fresh Nuclear Debris through the Atmosphere

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Highly radioactive particles were detected at Niigata within 36 hr or less of the third Chinese nuclear test on May 9, 1966. The individual particles ranged in total β-activity over a factor of about 100 and their particle size, from 5 to 24μ in diameter.

The debris particles were highly fractionated; concerning short-lived fission products, the collected particles were all enriched with refractory fission products ^{97}Zr, ^{99}Mo and ^{143}Ce, and depleted in ^{132}Te and ^{133}I, as would be expected by their chemical and nuclear properties. Most of the particles were impoverished with ^{239}Np, but some of them were markedly rich in ^{239}Np with no detectable amount of ^{132}Te and ^{133}I in them. Figure 1 shows two

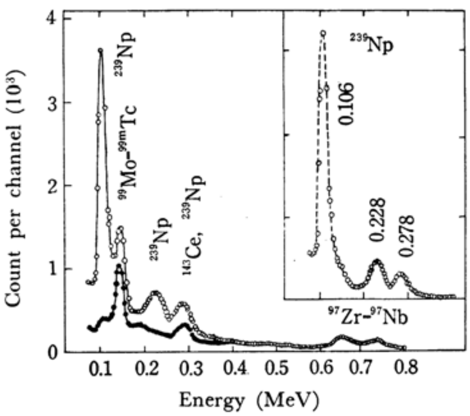

Fig. 1. Normalized γ-ray spectra of two particles (A and B), measured 44 hr after the formation ○, Particle A; ●, Particle B.

γ-ray spectra of such two extreme type particles; the two spectra are normalized to equal activity level of radiozirconium. A γ-ray spectrum obtained by ploting the difference in counts per channel between the two equalled a γ-ray spectrum of ^{239}Np, as shown in the inset. Figure 1 seems to suggest quite similar fractionation behavior among ^{97}Zr, ^{99}Mo and ^{143}Ce in spite of the difference in the ^{239}Np content.

According to extensive studies by Freiling[1] and Crocker[2] on the radionuclide fractionation, ^{239}Np seems to behave refractorily in the case of a surface burst. While, in the present observation, ^{239}Np seemed to behave like a refractory group only in a small number of the total particles collected. The particle samples studied were collected at ground level and at a particular location far away from the place of their formation. Taking this point into consideration, the present data would suggest that the gravitational and meteorological forces and the differences in size and density among debris particles would be combined to cause particle separation during a travel of the atomic cloud through the atmosphere. Thus, the fractionation behavior of ^{239}Np in this case seemed to be somewhat modified as compared with that derived from an averaged sample collected by a large-scale sampling procedure.

We have previously pointed out that a volatilely behaving nuclide ^{89}Sr found itself much more enriched in a rain sample than in a ground-air one and that ^{140}Ba of less volatile character behaved in a reverse way.[3] Further, it is of interest to refer to the fact that a rain sample (20 *l*) collected before highly radioactive particles appeared in ground air, had a unique radiochemical composition, consisting mainly of ^{239}Np, ^{132}Te and ^{133}I, but containing no detectable amount of refractory fission products. This finding would suggest that upper-air-borne particles rich in ^{239}Np, perhaps fine in size, first appeared in the upper atmosphere over Japan and a part of them fell to the ground by wash-out in rain. The observation of such rain-water activities would also give an indication that the particle separation may have taken place to a considerable extent in the course of a 1.5-day travel of the atomic cloud from the Chinese test site to Japan.

1) E. C. Freiling, *Science*, **133**, 1991 (1961).
2) G. R. Crocker, *Nature*, **210**, 1028 (1966).
3) S. Koyama, T. Sotobayashi and T. Suzuki, *Nature*, **209**, 239 (1966).