

Reassessment of the dispersion of uranium originating from nuclear test explosions across East Asia through the atmosphere

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Background and Methodology

We have already reported that some Japanese atmospheric deposits have been contaminated with the uranium whose isotope ratio ($^{235}\text{U}/^{238}\text{U}$) had been fractionated from the natural ratio [1, 2]. For example, the uranium with a high $^{235}\text{U}/^{238}\text{U}$ ratio relative to the natural ratio (enriched uranium) has continuously been transported to the western side of the Japanese Islands since the 1960s until today. This suggests that the debris of nuclear test explosions conducted in Asia before the 1980s have continuously drifted east through the atmosphere. These debris must have been transported to the east end of Asia not only directly but also through several resuspension processes from their hypocenter region. However, the actual source region and transportation processes of such nuclear debris have never been identified clearly. In this context, we preliminarily measured the uranium isotope ratios of the dry deposits from Asian mineral dust events at Nagasaki, Japan and of surface soils across East Asia to clarify the mechanisms for the transportation of nuclear debris. The dry deposits from dust events had been collected at Nagasaki, which is located at western Japan near the Korean Peninsula, in March 2001, just at the time of a huge Asian mineral dust (Kosa, yellow-sand) event.

Results and Remarks

The $^{235}\text{U}/^{238}\text{U}$ ratios in the dry deposits samples from Nagasaki are obviously higher than the natural ratio (maximally ca. +20%). Contrastively, no obvious fractionation in the uranium isotope ratio is found for the surface soils in the East Asian region served to this study, although they tend to have slightly higher $^{235}\text{U}/^{238}\text{U}$ ratio than the natural one. Our results undoubtedly reveal that Asian mineral dust has been transported with nuclear debris which contains enriched uranium. The contribution of secondary processes like repeated resuspension of contaminated surface soils, which had been overlaid with nuclear debris through fallout processes, to the anomalous uranium isotope ratios found in Japanese atmospheric deposits may not be so substantial.

[1] Kikawada *et al.* (2009) *J. Nucl. Sci. Technol.* **46**, 1094-1098.

[2] Kikawada *et al.* (2011) *Mineral. Mag.* **75**, 1182.

Decrease of Arsenate Adsorption onto Bacteriogenic Iron Oxides by the Presence of Organic Materials

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The adsorption behaviour of trace elements onto iron (Fe) oxides has been well demonstrated for their importance in water chemistry. Especially, bacterial-induced iron oxides (Bacteriogenic iron oxides: BIOS) are of common interest because of their ubiquity and characteristics of adsorption of various ions. In this study, arsenate adsorption behaviour onto (A) synthetic ferrihydrite, (B) natural BIOS collected from Okinawa hydrothermal vent, (C) synthetic BIOS obtained by incubation of iron-oxidizing bacterium (*Mariprofundus ferroxydans*) were compared. BIOS synthesis was performed using a set of diffusion cells by which can obtain pure BIOS free from other inorganic and organic materials which are abundant in natural BIOS (e.g., silica, clay mineral, and other ions adsorbed onto BIOS). Adsorption experiments were performed under sea water condition (I: 0.7 M; initial arsenate concentration: 70 mg/L; adsorbent: approx. 0.5 mg) as a function of pH 4-10. Iron mineral species of iron oxides were specified by Fe K-edge X-ray adsorption fine structure (XAFS) [1] and adsorption structure of arsenate was examined by As K-edge XAFS analysis.

Iron K-edge XAFS analysis revealed that both natural and synthetic BIOS consisted mainly of ferrihydrite with 45-55 % of highly amorphous Fe hydroxides that is characterized by the primitive Fe hydrolysis stages. The crystal size was nano-scale which was smaller than the synthetic ferrihydrite. Thus, It was expected that BIOS should have more arsenate adsorption capacity than synthetic ferrihydrite. However, the amount of arsenate adsorbed onto each iron oxides decreased in the order of synthetic ferrihydrite > natural BIOS = synthetic BIOS with a same adsorption trend as a function of pH. XAFS and μ -XRF analysis indicated that arsenate was mainly adsorbed onto Fe phase within natural and synthetic BIOS forming inner-sphere complexation to the Fe oxides. These results were consistent with previous results on arsenate adsorbed onto synthetic ferrihydrite. Contrary to their reduced crystal particle size, specific surface areas of synthetic BIOS was decreased by 25% from synthetic ferrihydrite, which is possibly caused by the coprecipitation of Fe oxides with organic materials [2]. Thus, it is suggested that strong aggregation of Fe particles by the presence of organic material reduces (i) the surface area and/or (ii) the active adsorption site within BIOS, which ultimately result in the decrease of the arsenate adsorption onto BIOS. These results also suggests that direct and indirect effects of organic material should be taken into account to evaluate the anion adsorption onto BIOS.

[1] Kikuchi *et al.* (2011) *Chem. Lett.* **40**, 680-681

[2] Mikkuta *et al.* (2008) *Geochim. Cosmochim. Acta.* **72**, 1111-1127.