Investigation of isotope fractionation of Fe in anthropogenic aerosols to determine its contribution to the surface ocean

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In high nutrient-low chlorophyl (HNLC) regions, iron (Fe) is a limiting factor for the primary production [1]. Natural (lithogenic) aerosol is one of the main sources of Fe to the regions, whereas recently it has also been reported that Fe in anthropogenic aerosols is another important source due to its higher solubility to the seawater than that of natural Fe [2]. However, relative contribution of natural and anthropogenic Fe to the HNLC regions is still unknown. In our previous study, it was found that Fe isotope ratio (δ^{56}Fe (‰) = $1000\times[(^{56}Fe/^{54}Fe)_{sample}/(^{56}Fe/^{54}Fe)_{IRMM\text{-}014}]-1$) of anthropogenic Fe is much lower than that of natural Fe, indicating that Fe isotope ratio can be used as a tracer to know their contributions [3]. In this study, we aimed to understand the reason why anthropogenic Fe yields low δ^{56}Fe, and to know contribution of anthropogenic Fe to the ocean. Aerosols collected near some anthropogenic Fe sources and marine aerosols were used for analysis. Isotope analysis was conducted using multicollector ICP-MS. X-ray absorption fine structure (XAFS) analysis was also conducted to know their species which can be related to the formation process.

We found that most fine aerosols containing Fe were spherical and in the form of (hydr)oxides, indicating that they were emitted through evaporation under high-temperature condition. They showed δ^{56}Fe values 1~4‰ lower than their source materials through the evaporation processes. Following the Rayleigh equation, we suggested that low volatility of Fe is important for the large fractionation and that degree of the fractioation can be different by the evaporating conditions such as temperature or species.

It was found that fine particles in the marine aerosols had lower δ^{56}Fe values than those of coarse particles, especially near the coast. Using endmenber of natural and anthropogenic Fe (0.0‰ and -3.9‰, respectively), up to half of dissolved Fe in the marine aerosols were suggested to be of anthropogenic origin, indicating that anthropogenic Fe can be an important source to the surface ocean.

[1] Martin & Fitzwater (1988), *Nature* **331**, 341–343. [2] Sedwick *et al*. (2007), *Geochem. Geophys. Geosyst.* **8**, 1–41. [3] Kurisu et al. (2016), *J. Geophys. Res. Atmos.* **121**, 11119–11136