Estimation of emission source of sulfate aerosol collected in the northwestern region in Japan

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Sulfate aerosol has large impacts on climate and environment. To estimate the effects, it is important to know sulfate speciation, sources, and transport. In this study, we show chemical characterization of size-fractionated sulfate aerosol collected in Noto peninsula, in the northwestern region of Japan, which is a remote background site sensitive to air mass transported from mega cities in East Asia. In addition, sulfur isotope ratio ($\delta^{34}S$) and trace element abundances were also analyzed to discuss emission sources. Here, we reported results of fine particles (< 1.3 µm) collected in summer when air mass mainly came over Japan Sea and winter when it passed through Asian continent.

Speciation of sulfate measured by X-ray absorption fine structure spectroscopy was almost same between both seasons: ammonium sulfate and hydrated sulfate were dominant. Meanwhile, δ^{34} S values in summer sample was higher form 2.5 to 7.0 ‰ than that in winter sample depending on particle size. δ^{34} S values of aerosol can change depending on emission source and isotope fractionation [1]. However, the latter effect was small to explain variation of δ^{34} S values between two seasons. Therefore, difference in the δ^{34} S was plausibly caused by the different emission sources of sulfate aerosols. Low NO_3^{-}/SO_4^{2-} mass ratio (< 0.34) in aerosol, which is used as an indicator of sulfate source [2], suggested contribution form gasoline and diesel was negligible. δ^{34} S value of heavy oil, which is often used to ship fuel, is generally higher than that of coal [1]. Additionally, in summer, enrichment factor of vanadium (V) and nickel (Ni) relative to crust was significantly higher in summer than that in winter, and V and Ni are positively correlated with sulfate. V and Ni are useful indicator for aerosol emitted from heavy oil combustion [3]. Our results indicated that source of sulfate aerosol was different between summer and winter, though sulfate speciation was similar. Contribution of sulfur originated from heavy oil can be significant in summer season.

[1] Han et al. (2017) *Sci. Rep.* **6**, 29958. [2] Yao et al. (2002) *Atmos. Environ.* **36**, 4223-4234. [3] Becagli et al. (2012) *Atmos. Chem. Phys.* **12**, 3479-3492.