Temporal and spatial variation of anthropogenic sulfur deposition in Japan by using sulfur isotopic ratio

*Yayoi Inomata[1], Tatsuyoshi Saito[1], Yukimasa Morohashi[1], Naoyuki Yamashita[1,2], Hiroyuki Sase[1], Tsuyoshi Ohizumi[1,3], Katsuyuki Takahashi[4], Naoki Kaneyasu[5], Daisuke Funaki[6], Aya Iwasaki[7], Kazunori Nakagomi[8], Tomoaki Shiroma[7], Takashi Yamaguchi[9]

1.Asia Center for Air Pollution Research, 2.Forestry and Forest Products Research Institute, 3.Niigata Prefectural Institute of Public Health and Environmental Sciences, 4.Japan Environmental Sanitation Center, 5.National Institute of Advanced Industrial Science and Technology, 6.Shimane Prefectural Institute of Public Health and Environmental Sciences, 7.Okinawa Prefectural Institute of Health and Environment, 8.Nagano Environmental Conservation Research Institute, 9.Environmental and Geological Research Department

[Introduction]
It is well recognized that non-sea salt (nss) sulfur isotopic ratios are useful to identify the sources because the sulfur isotopic ratio have source specific values. We have measured sulfur isotopic ratio in precipitation by using Japanese monitoring sites for the Acid Deposition Monitoring Network in East Asia (EANET). In this study, we investigated the spatial and temporal distribution of anthropogenic sulfur deposition and evaluated the contribution from transboundary transport from the Asian continent to Japan,
[Observation and analysis]
We collected precipitation samples at 12 sites (Rishiri, Tappi, Ochiishi, Sadoseki-misaki, Niigata-Maki, Niigata-Kajikawa, Oki, Happo, Tokyo, Ijira, Hedo, Ebino, Ogasawara) after 2014. The sampling interval was 2 weeks, 1 month, and seasonally depending on the sampling schedule at each site. The sulfur isotopic ratios were measured by stable isotope mass spectrometer. Canyon Diablo Troilite (CDT) was used as the standard material. The analytical precision was ±0.16‰. The nss-sulfur isotopic ratio was calculated assuming that Na^+ is originated from sea water.
[Results and discussion]
At the monitoring sites located in the coast of the Sea of Japan, temporal variations of nss-sulfur isotopic ratio were characterized by seasonal variation with high in winter and low in summer season (e.g. Niigata-Kajikawa, +2-+4.9‰). Similar seasonal variations were also found in Tokyo and Ijira, where locates the Pacific coast sites. However, nss-sulfur isotopic ratio in Tokyo and Ijira was lower with smaller seasonal variation in comparison with those at the coastal site of the Sea of Japan (e.g. Tokyo, -0.73-+4.0‰). It was reported that sulphate in the air mass in China were strongly affected by coal combustion (0-15‰, Xiao et al., 2011), whereas that in Japan were derived from oil combustion, which was negative values (-2.7‰, Ohizumi et al.,1997). Furthermore, it was reported that nss-sulfur isotopic ratio in aerosol in central China is 4.5±1.3‰ (Li et al., 2013). The difference of nss-sulfur isotopic ratio and these seasonal amplitude between coastal site of Sea of Japan and Pacific coast suggests that transboundary transported sulphate were mixed with the emitted sulphate from domestic (Japan) sources.
At Happo, mountain monitoring site (1850 m asl), nss-sulfur isotopic ratio were almost constant value (4.7±1.2‰) except for several samples in spring. Contribution of Asian dust will cause the higher values. Almost constant value at Happo suggests that nss-sulfur isotopic ratio is minor contribution from local anthropogenic sources, indicating the transboundary transport from the Asian continent.
At Ogasawara, remote marine site, nss-sulfur isotopic ratios were high in spring-summer and low in winter, which were opposite seasonal variation. The higher nss-sulfur isotopic ratio in

spring-summer was possibly the biogenic emissions.
Relative contributions of sulfur with different sources have been evaluated with mass balance model. It was clear that the transboundary transported sulphur deposition in the coastal site of Sea of Japan (e.g. Niigata-Kajikawa 1.1-20 mg m^{-2} day^{-1}) is two-three times larger than those in the Pacific coast (e.g. Tokyo 0.03-6.7 mg m^{-2} day^{-1}).
[References]
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