Estimation of air pollutant sources from the seasonal variation of Sr and Pb isotope ratios of aerosols

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[Introduction] Transboundary air pollution remains as a major environmental concern in Japan since it is located downwind of the Asian continental outflow. Due to the prevailing high pressure system in the Pacific, it has been generally accepted that the air quality near the ground surface of central Japan is relatively unaffected by the polluted air-mass originating from the Asian continent. However, our recent observations and increasing number of studies have seen episodes of high aerosol concentrations even in summer periods, indicating the significance of the polluted air-mass not only spring and winter but also in summer. However, as there are a variety of aerosol sources (e.g. refuse incineration, volcanic eruption) within Japan, it is difficult to distinguish domestic aerosols from foreign ones. In order to identify the sources of air pollutants in Japan, we determined the Sr and Pb isotopic ratios and elemental compositions of aerosols collected at Noto peninsula, Japan, in different seasons.
[Samples and analytical procedure] The sampling site is NOTO Ground-based Research Observatory (NOTOGRO) at the tip of Noto peninsula (37°45'N, 137°36'E). All of the samples were collected weekly on the roof of a building (15 m a.g.l.) using a High-volume air sampler (AH-600F, SHIBATA) with a constant flow rate of 700 L/min. Only the coarse fraction exceeding 2.5 μm in diameter was collected on a 12.6 ×16.6 cm^2 SiO_2 filter through an impactor. Filters were extracted with 5% HOAc (acetic acid) solution. Residual fraction after extraction with HOAc was digested in HF-HCl-HNO_3 solution. Isotopic ratios of Sr and Pb were measured using a Thermo Scientific Triton thermal ionization mass spectrometer (TIMS) and Thermo Scientific Neptune multicollector-inductively coupled plasma-mass spectrometer (MC-ICP-MS), respectively.
[Results and discussion] The HOAc-leachate materials are characterized by low $^{87}Sr/^{86}Sr$ ratios and approach to a marine value of 0.70918, indicating the dominant contribution of sea salt aerosol. In contrast, the HOAc-residue materials have high $^{87}Sr/^{86}Sr$ ratios, and sample collected from 9 to 16 July show the highest $^{87}Sr/^{86}Sr$ ratios similar to surface soil silicates in the north China (0.71432 in average; Nakano et al., 2004). This result supports a view that the continental air-mass may reach to Japan even in summer season. About Pb isotope ratios of the HOAc-leachate materials, samples with low V/Mn showed similar Pb isotopic ratios reported in China. Major sources of V and Mn are from oil combustion and coal combustion, respectively. This result suggests that the air-mass over NOTOGRO in spring to early summer of 2015 are affected by the coal-derived aerosols in China.

Keywords: aerosol, Sr-Pb isotopes, transboundary air pollution

