Investigation on the possibility of the identification of air pollutant sources using stable isotope ratios of lead and strontium

*Shinji Sakurai[1], Takao Nakagiri[1], Kosuke Tanaka[1], Haruhiko Horino[1], Ki-Cheol Shin[2], Ichiro Tayasu[2], Shiho Yabusaki[2]

1. Osaka Prefecture University, 2. Research Institute for Humanity and Nature

In Japan, transboundary air pollution coming from neighboring countries is concerned as one of the serious threads on the health. However, it is not easy to assess the actual impact because of difficulty to identifying the detailed transport path and the source of pollutants. Although it is expected that stable isotope ratios (hereafter, isotope ratios) of lead (Pb) and strontium (Sr) can be used as environment tracers to identify the pollutant source, even the basic information on their characteristics such as chronological and/or seasonal variations at resolution of rainfall event as well as the relationship between the ratios and other water quality parameters is still not enough to consider the possibility.
In our study, rainwater was collected in a polyethylene bottle, on the top of which a polypropylene funnel was set, separately in every rainfall event from June 2016 to August 2018 at the campus of Osaka Prefecture University in Sakai, Osaka, Japan. The isotope ratios of Pb and Sr ($^{207}Pb/^{206}Pb$, $^{208}Pb/^{206}Pb$, and $^{87}Sr/^{86}Sr$) in the rainwater samples were measured using a multi-collector inductively coupled plasma mass spectrometer. Concentrations of heavy metals (Fe, Mn, Cd, Cu, Ni, and Pb), inorganic ions, pH, EC, TOC, COD, T-N, and T-P were measured as water quality parameters. Rainfall amount was measured with a rain gauge set near the rainwater sampling point. Other meteorological data needed to investigate and estimate the pollutant sources, such as air temperature, wind direction, and duration of drought, were obtained from the Japan Meteorological Agency.
There was no obvious trend either chronologically or seasonally for $^{207}Pb/^{206}Pb$ or $^{87}Sr/^{86}Sr$. The values of the $^{207}Pb/^{206}Pb$ ratio declined slightly during the observation period, but the $^{87}Sr/^{86}Sr$ ratio did not show this chronological variation.
There was no obvious correspondence between most of water quality parameters and each isotope ratio. The $^{207}Pb/^{206}Pb$ varied widely during events in which Cr and Fe concentrations were low, but was close to 0.871 as the concentrations of these metals were getting higher. There is similar tendency in $^{87}Sr/^{86}Sr$ for Fe and Cr. It was suggested that these metals might be derived from the same source.
For correspondence with meteorological factors, the monthly rainfall amount weighted average of $^{207}Pb/^{206}Pb$ fluctuated when the wind from the west or the west-southwest was dominant, while it took almost a unique value for the south-southwest. It was suggested that Pb was originated from various sources with the west wind.
Plots of the obtained data on the $^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$ were distributed upside and parallel to the lead isotope growth curve. When comparing our plots with the previous global studies, we found that ours were distributed in a manner similar to those from China and Russia. It was indicated that it is difficult to identify pollutant sources from these countries, which are thought to cause transboundary pollution in Japan.
Further research, including the collection of data on isotope ratios as environmental tracers, is necessary to identify pollutant sources using isotope ratio of Pb and/or Sr.

Keywords: stable isotope ratio, air pollution, heavy metal, lead, strontium, rainwater

