大気中水銀同位体分析：手法開発および実大気への適応
Investigetion of atmospheric mercury isotopic compositions: technical development and applications

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Mercury (Hg) is a toxic heavy metal, which exists in various chemical forms in the environmental system. In the atmosphere, Hg exists in three forms ($Hg^0_{(g)}$, gaseous elemental Hg (GEM); $Hg^{2+}_{(g)}$, gaseous oxidized Hg compounds; and $Hg_{(p)}$, particulate/aerosol bound Hg). $Hg^0_{(g)}$ is the dominant species of atmospheric Hg, accounting for >95% of the total Hg in the atmosphere. Because $Hg^0_{(g)}$ is highly volatile and has limited solubility in water, it cannot be easily removed by wet or dry deposition processes. Therefore, the residence time of $Hg^0_{(g)}$ in the atmosphere is relatively long (0.5 to 1 years), which allows long-range transport from mercury emission source(s). Conversely, $Hg^{+2}_{(g)}$ and $Hg_{(p)}$ are effectively removed from the atmosphere through wet and dry depositions. Because $Hg^0_{(g)}$ deposit upon oxidation to $Hg^{2+}_{(g)}$ far from its emission source(s), deposition on local/regional scales represents a combination of multiple sources of Hg (global, regional, or local; natural or anthropogenic).

Over the last decade, the development of analytical methods of highly precise Hg isotopic measurements demonstrated mass dependent fractionation (MDF) and mass independent fractionation (MIF) of Hg isotopes in environmental samples. MDF of Hg isotopes occur during various natural and industrial Hg transformations. MIF of Hg isotopes is observed during abiotic reduction, photochemical and non-photochemical, and physical and chemical processes. Such processes lead to differences in the Hg isotopic composition of different emission sources, both natural and anthropogenic, and atmospheric processes (i.e., transportation, oxidation/reduction, deposition, and reemission). Therefore, Hg isotopic compositions could be used as a tracer of the sources and processes of atmospheric Hg. In this study, we aimed to develop the methods of collection and pretreatment for isotopic measurement of $Hg^0_{(g)}$, and then isotopic composition of $Hg^0_{(g)}$ was investigated for various regions in Japan.

To identify potential mercury sources, air mass back trajectories were calculated for each sample using the NOAA HYSPLIT model. We divided the back trajectory patterns observed in the results into three groups of air masses predominantly derived from (1) a marine source derived from the Pacific Ocean, (2) coastal and land sources that probably contain anthropogenic mercury emitted from urban-industrial regions, and (3) continental sources associated with northwesterly flow at higher altitude (>1500 m) and long-range transportation. Although multiple sources were possibly impacted during the 24 hr ambient sampling, we were able to observe a correlation between the back-trajectory types and Hg isotopic composition.

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