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Mass-Independent Fractionation Reveals the Sources and Transport of Atmospheric Particulate Bound Mercury

Xuechao Qin[1,2], Xinyuan Dong[3], Congqiang Liu[3], Rongfei Wei[1], Zhenghua Tao[1,2], Hua Zhang[4], and Qingjun Guo[1,5]

[1]Center for Environmental Remediation, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

[2]University of Chinese Academy of Sciences, Beijing 100049, China

[3]Institute of Surface-Earth System Science, School of Earth System Science, Tianjin University, Tianjin 300072, China

[4]State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China

[5]College of Resources and Environment, University of Chinese Academy of Sciences, Beijing 100190, China

Mercury (Hg) is highly toxic and the only heavy metal that can exist in the atmosphere in gaseous form. When atmospheric Hg mixes with aerosols, it forms particulate-bound mercury (PBM). PBM can be transported and settle down quickly across regions, posing serious threats to ecosystems globally. Despite these concerns, tracking the sources and transport of atmospheric Hg remains challenging due to its global dispersal nature. However, the three-dimensional fractionation of Hg isotopes provides a feasible approach for addressing this issue. In this study, $PBM_{2.5}$ and PBM_{TSP}samples were collected simultaneously in rural, suburban, urban, industrial, and coastal areas of the Beijing-Tianjin-Hebei (BTH) region, which is influenced by severe atmospheric pollution and the East Asian monsoon. Due to the significant influence of anthropogenic sources, the concentrations of $PBM_{2.5}$ and PBM_{TSP} were highest in the industrial and coastal areas, followed by the urban, suburban, and rural areas. The δ^{202}Hg values of $PBM_{2.5}$ and PBM_{TSP} at the five sites were negative, overlapping with the values of most anthropogenic sources. However, most $PBM_{2.5}$ and PBM_{TSP} samples showed significantly positive Δ^{199}Hg, significantly higher than the values of emission sources,especially for $PBM_{2.5}$. The mass-independent fractionation (MIF) of Hg and sulfur isotopes showed that strong photochemical reduction happened during long-distance transport, making Δ^{199}Hg have a positive shift. The positive changes in Δ^{200}Hg may be due to ozone-mediated oxidation during the transport process, as shown by the interesting relationships between O_3, Δ^{199}Hg, and Δ^{200}Hg in $PBM_{2.5}$. Additionally, the analysis of backward trajectories unveiled the influence of air masses originating northwest of the BTH region through high-altitude transport. The cross-border transport of PBM, influenced by westerly and northwesterly air masses from Central Asia and Russia, markedly impacted PBM pollution in the BTH region. Furthermore, these air masses, upon reaching the BTH area, would transport heightened PBM concentrations to the ocean through the winter monsoon. Conversely, during the summer, southeastward air masses transported from the ocean by the summer monsoon acted to mitigate the inland PBM pollution. The study results show that significant positive odd-MIF of PBM can occur in places with intensive

anthropogenic emissions rather than being limited to remote areas. It implies that the odd-MIF resulting from atmospheric transport has likely been significantly undervalued. Our research offers valuable perspectives on the transport, transformation, and circulation of Hg in the environment.