

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-493

Anonymous Referee #1

Referee comment on "Mercury isotopic compositions in fine particles and offshore surface seawater in a coastal area of East China: Implication for Hg sources and atmospheric transformations" by Lingling Xu et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-493-RC1>, 2021

The Manuscript entitled '**Mercury isotopic compositions in fine particles and offshore surface seawater in a coastal area of East China: Implication for Hg sources and atmospheric transformations**' investigated the Hg isotopic composition of fine aerosols (PM_{2.5}) sampled from industrial and mountain sites in a coastal area of East China. In addition, the authors also evaluated the Hg isotopes in surface seawater close to the Industrial area. The authors aimed to obtain the roles of anthropogenic sources and atmospheric transformations in particulate Hg isotopic compositions. Stable Hg isotopes have become a useful proxy for the identification of Hg sources, particularly as a result of improvements in high-precision analytical methods. Limited data are available on the stable isotopes of Hg or their application in source apportionment in atmospheric aerosols. Therefore, studies on atmospheric Hg and its isotopic compositions are important for understanding the atmospheric concentrations, sources, transport mechanisms, and fate of particulate Hg and the data are important to the broad scientific community. The manuscript is well written and the results are discussed in detail, although, some of the latest studies are not reviewed. Hence, I suggest the acceptance of this manuscript in ACP after minor suggestions below are addressed.

A little more on atmospheric particulate mercury (PBM) and its scenario (literature review) is needed in the introduction section.

The motivation to carry out this study must be made clear with more gaps identified.

Line 98-112: The literature review missed some of the recent works on PBM isotopic ratios of atmospheric samples (e.g., Source identification of atmospheric particle-bound mercury in the Himalayan foothills through non-isotopic and isotope analyses; Atmospheric particle-bound mercury in the northern Indo-Gangetic Plain region: Insights into sources from mercury isotope analysis and influencing factors).

Line 246-249: The authors presented the Hg mass in PM_{2.5}, however I did not find the PBM concentrations presented and discussed. The Hg mass can also suggest the source is from natural or anthropogenic. When assessing Hg enrichment and sources, the PBM/PM ratio may be useful if we have Hg concentrations for natural and anthropogenic

components (e.g., soil and coal) in the region of interest? Please check it for the two studied sites.

Line 258: Spearson correlation? Should be Spearman?

The Hg isotope data presented here does not seem to be able to distinguish between different sources. For example, Hg isotopes (Figure 2) show urban, remote and near sources, however, the clear sources e.g., coal, industrial emission, traffic and soils are all possible source of particulate Hg? This is not clear and not discussed clearly. Distinguishing between these sources seems difficult based on isotope alone. Thus I am not sure why the authors conclude anthropogenic sources (what are the sources) is not clear.

Line 343: Why the authors directly start with numbering 1. Coal combustion, this may break the flow and so on?

Similarities or differences in Hg isotope ratios at the two sites need to be described and the different seasons of their collection reported. The authors should see if their results plotted on a coherent mixing line on an inverse Hg concentration plot (i.e. $\delta^{202}\text{Hg}$ vs $1/\text{HgP}$). Soils and values for PM from other locations in China might also be informative on such a plot. More broadly, Hg isotope ratios in aerosols from coastal sites should be compared with those in aerosols from other locations in Asia. This may be placed in Supplementary document.

Plot of $\Delta^{199}\text{Hg}$ (‰) vs. $\delta^{202}\text{Hg}$ (‰) is not presented. Hg-MIF ($\Delta^{199}\text{Hg}$) signatures are also valuable for distinguishing Hg contamination pathways because Hg^{2+} photo-reduction in aerosols. The authors discussed on the slope, however, it is important to show the figure to clearly understand the atmospheric transformation and photochemical process.

Line 421-424: This statement needs more thought. Photo-reduction of Hg^{2+} mostly results in positive $\Delta^{199}\text{Hg}$ in reactant Hg.

Line 530-532: Please show in figure as suggested previously.

The detailed revisions are needed before publications.