

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-493

Anonymous Referee #2

Referee comment on "Mercury isotopic compositions in fine particles and offshore surface seawater in a coastal area of East China: implications for Hg sources and atmospheric transformations" by Lingling Xu et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-493-RC2>, 2021

The proposed paper described Hg isotope variation of PM_{2.5} sample collected from urban and mountain area of East China to test Hg isotope as the tracer of source and process of particulate Hg in atmosphere. Since Hg isotope of particulate Hg is still scarce, the data provided by this study surely contribute to better understanding of Hg chemistry in the atmosphere. The authors well cover the previous monitoring and experimental studies, and they tried to interpret their data set through comparing the relevant works. Nevertheless, two points have to be considered to evaluate this work correctly. Firstly, description of methodology section is insufficient. As authors mentioned, the technical difficult is accurate measurement of trace amount of particulate Hg in PM_{2.5} sample. I cannot validate quality of the data only from the provided information in methodology section (see specific comments). Secondly, missing of Hg⁰ data makes all interpretation rather speculative. Gaseous elemental Hg is the predominant form of Hg in atmosphere, while gaseous oxidizing Hg and particulate Hg (likely contribution of Hg(II) is high) occupy minor pool. Conversion of Hg species from large to minor pool potentially causes large isotope fractionation. I think authors should mention the isotopic variation of GEM in China more carefully to interpret their data. The specific comments are as below.

L66. Despite Hg_{PM} level expressed here being volume based, their own results are expressed as mass basis. It makes comparison difficult.

L92. ~1.0 for photo-reduction of Hg(II)

L93. ~1.6 for photo-oxidation of Hg(0)

L144. What is "regional emission"? It should be specified.

L147. Although I thought seawater data is rather minor focus in this paper, more oceanographic background should be provided to help data interpretation, such as temperature and primary productivity.

L200. This means, authors pooled 10 samples to be one? If so, it should be written accordingly.

L202~. Recovery through this combustion process should be given at the last of this paragraph. Careful operation is often required for complete recovery using dual combustion furnace.

L214 Concentration of SnCl_2 should be given.

L220-227. The description of MC-ICP-MS analysis is poor although they cited one reference paper. The method here is CV-MC-ICP-MS? If so, it should be noted. In which aqueous Hg concentration did author choose to the isotope analysis? Did author match the Hg signal of sample and standard? The UM-Almaden values were obtained by exactly same level to the sample? Since sample measurements were made only one time, these information are important to validate data quality.

L246. Again, why the author showed only mass-based concentration. Besides, there are no data of total mass of particle on the filter. Without this value, readers cannot calculate concentration of Hg in final solution used for the isotope analysis. If the author used hydride generation system such as CETAC HGX-100, >2.5 ng is required for the precise data analysis.

L249. Again, what is the regional emission?

L300. remarkably positive odd-MIF

L302. L299-302. I couldn't understand the reasoning here. What is the enhanced photo-reaction? Hg^0 reduction? Or MMHg demethylation? The $\delta^{202}\text{Hg}$ vs $\Delta^{199}\text{Hg}$ of DMS in Fig. 2 seems positively correlated with slope being ca. 0.4. Does this trend support author's interpretation?

L349. A prior study estimated that...of coal feeds based on the mass balance model (Sun et al., 2014).

Figure 4. I am afraid poor data quality from the rather scattered correlation of $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$.