

## ***Interactive comment on “Isotopic composition for source identification of mercury in atmospheric fine particles” by Q. Huang et al.***

**Anonymous Referee #2**

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Huang et al. report mercury isotope signatures on atmospheric particles in a urban environment, as well as in potential particulate Hg sources. Seasonal variations are observed, and discussed as differences in particle emission sources (Biomass burning, coal combustion, smelting, long-range transport). The approach used makes sense and the overall discussion is of good quality. I have a few comments and questions that could help to improve the discussion:

1) First, as mentioned in the text, atmospheric processes can affect Hg isotope composition. The authors only discussed the variations in terms of different sources, with mainly local emission sources as well as long range transport of PM<sub>2.5</sub>-Hg. I wonder if the presence of PM<sub>2.5</sub> can modify the atmospheric Hg speciation, enhancing oxidation of GEM and/or RGM binding on particles. In other words, does PM<sub>2.5</sub> and Hg necessarily have the same source? I expected more discussion on atmospheric Hg

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dynamics.

2) I was also surprised to see that the most extreme Hg isotope signatures (at least for  $\Delta^{199}\text{Hg}$ ) are found for PM<sub>2.5</sub> samples, and not for potential sources (Figure 3). The high  $\Delta^{199}\text{Hg}$  are discussed as deriving from long-range transport, which could make sense although it is not proven. The lowest  $\Delta^{199}\text{Hg}$  signatures are however discussed as an impact of local coal combustion, while coal samples analyzed here do not display such low  $\Delta^{199}\text{Hg}$ .

3) Lines 337-340: The principal component analysis on element concentrations indicates that biomass burning is only a minor source of PM<sub>2.5</sub>-Hg. Later in the discussion (paragraph starting line 365), biomass burning is evaluated as an important parameter driving PM<sub>2.5</sub>-Hg concentration and isotope signatures, especially in Autumn. Could you comment on these contrasting conclusions?

4) Lines 463-468: Here is a discussion about the potential effect of photochemical reduction of Hg. It is true that Hg photoreduction usually induces enrichment in heavy and odd Hg isotopes in the remaining fraction (as observed by Bergquist and Blum, 2007). However, the inverse effect (enrichment in heavy and even Hg isotopes) was observed experimentally in presence of sulfur ligands (Zheng and Hintelmann, 2010). Finally, could this explain the Hg isotope variations in PM<sub>2.5</sub>?

5) Regarding the objective of the study, which was to evaluate “the effectiveness of the Hg isotope technique for tracking the sources of the PM<sub>2.5</sub>-Hg”, would you say that Hg isotope signatures were necessary? It seems to me that the main conclusions are made based on EC and Zn/Al ratio (shown in Figure 7). If only Hg concentrations and isotope ratios were known, would you be able to address PM<sub>2.5</sub>-Hg sources?

Minor comment: On line 70, The range given for GEM  $\delta^{202}\text{Hg}$  (-3.88 to 0.43 ‰) is incorrect. Demers et al. (2015) found values up to 1.4 ‰