

Shi et al., reported air Hg measurements in Jan and Jul of 2012-2020, and tried to quantify the potential sources for these measured data annual trends. Generally, these long-term data are beneficial to understand the Hg emissions and air Hg variations in China, since a series of air cleaning actions taken in recent years by the government the subject is of interest; the methodology is robust; the results and discussion are presented well in the most sections. Several issues need the authors further to identify:

Response: We appreciate Prof. Feng for providing valuable comments. We have studied these comments carefully and revised the manuscript accordingly.

Section 2.2

From the authors' description, just using the Tekran 2537 without an annular denuder, the data should be mixed the signals of GOM and some PBM which with the small particle size. I suggest the authors should state clearly about their measurements. If the ratio of GOM and PBM to TGM are <5%, the authors can use the GEM to represent the TGM.

Response: We added the details of atmospheric Hg measurement in the revised manuscript (**Line 87-92**). The Tekran 2537B/1135/1130 ambient Hg speciation system was simultaneously running in this study. During the 1 h sampling period, the GOM and PBM were first collected onto the KCl-coated quartz annular denuder and the quartz filter respectively, and GEM concentrations were measured by a Tekran 2537B Hg vapor analyzer every 5 min at a sampling flow rate of 1 L min⁻¹. The detection limit of GEM measurement was 0.06 ng m⁻³. In the following 1 h desorbing phase, the PBM and GOM were sequentially desorbed and then quantified by the Tekran 2537B. For most of the data (70%) during the study period, the proportion of GOM and PBM in TGM was less than 5%. Thus, GEM concentrations in this study were directly compared to TGM in the following discussion.

Section 2.5

This section is very important in the whole methodology section, but the authors' description was not very clearly. Several issues need the author further confirm: one is 24h-Latitude and 24h-Longitude? What's the detail representation of these terms, the back-trajectory endpoint location during last 24 h? Another one is the air transmission. I would like to say it is the air transportation.

Response: Yes, the 24h-Latitude and 24h-Longitude interaction represents the backward trajectory endpoint location during last 24 h. We replaced the expression "air transmission" with "air transportation" in the revised manuscript. In addition, we quite agree with the importance of this section and have added more details about variable selection and factor interpretation in the manuscript (**Section 2.5 Model development**).

Section 3.1.1

Line 220-230 Given the authors only measured the GEM concentrations in Jan and Jul in each year, the authors should compare their data with the references mentioned the same month data, not the annual average data.

Response: According to our previous study, the GEM concentrations in Xiamen showed a significant seasonal variation and, January and July were typical months representing winter

and summer (Xu et al., 2015). In the revised manuscript, we added an explanation of seasonal representation (Line 106-114), and compared the Jan. and Jul. data with corresponding winter and summer data from the references rather than annual average data (Line 210-216).

Section 3.2

In this section, the authors mainly attributed their observed Hg seasonal and diurnal cycling to the local anthropogenic emissions and long-range transport. Recently, several studies from the China cities also showed that the regional surface Hg emissions from soils and city bare regions, and Hg chemical transformations in the air of cities, and regional Hg natural surface emissions, such as from the soils and nearby the oceans. I suggest the authors to further incorporate these potential reasons in their discussion.

Response: Thanks for your advice, we have taken those potential reasons into account in our discussion. We considered the effect of Hg surface emissions especially from the ocean in summer in the discussion of regional emissions (Line 366-371), and considered the effect of the photo-oxidation of GEM to the diurnal variation of GEM (Line 289-291). What's more, we gave a more detailed explanation of the representative variables in Section 3.3.2. The meteorological factor represented by T and RH likely encompassed the effect of natural surface emissions, and liquid phase reduction process of reactive Hg in the atmosphere.

Section 3.3.2

This section is the key discussion parts of the whole manuscript. From the GAMs modeling results, the authors stated that the meteorological factors are the most important factor to shape the GEM variations. However, the authors mainly stated these factors' contribution which derived from the modeling. From my view, the meteorological factors influencing GEM variations by several pathways. One is that the meteorological factors drive the Hg chemical transformation, such as UV, RH are highly related to the photo-reduction and GOM formations in the air, especially in the haze, these meteorological factors playing a dominant role in GEM transformation to GOM and PBM in the air. These kinds of studies have been reported in Hefei, Shanghai and Beijing. Another important role is that meteorological factors are highly related to the Hg emissions from the natural surfaces. From the current modeling results, the Hg natural emissions from the natural surfaces (e.g., soils, water, etc.) are comparable to the anthropogenic Hg emissions in China mainland. Substantial flux measurements have clearly showed that the elevated temperature and solar radiation can significantly promote the Hg re-emissions from these nature surfaces. Overall, I suggest the authors explain the cause of the contribution of meteorological factors in more detail, specifically related to the Hg emission inventory and Hg transformation mechanisms in the air, by some typical case periods of data (several tens of hour Hg, meteorological factors data) to show their interactions, not just a data presentation.

Response: These suggestions are of great help to improve the quality of the manuscript. We have made the following major revisions:

- 1, In the revised version, we used the min-maximum method to standardize the parameters before the model running. Then the meteorological parameters were screened mainly based on statistic judgement, and finally T, RH, and BLH were retained in the model. To better illustrate how the meteorological parameters affect GEM concentrations, we plotted the corresponding

curves of GEM with the meteorological parameters (**Figure 8, Line 420-430**). Generally, GEM concentrations increased with temperature, which was likely attributed to natural surface emissions. GEM concentrations increased with the raise of RH, possibly because the high RH environment was favorable for the liquid phase reduction of reactive Hg to GEM (Horowitz et al., 2017; Saiz-Lopez et al., 2018; Huang et al., 2019).

2. As you suggested, we have added a typical case in January 2013 to further discuss the influence of meteorological parameters on GEM (**Figure S8, Line 430-434**). In this case, we found that the daily GEM concentrations increased with the increase of T and RH as well as the decrease of BLH. The case also implies that increased temperature likely accelerated natural surface emissions and high RH was favorable for the liquid phase reduction of reactive Hg to GEM, and consequently contributed to GEM in the atmosphere.

References

Xu, L., Chen, J., Yang, L., Niu, Z., Tong, L., Yin, L., and Chen, Y.: Characteristics and sources of atmospheric mercury speciation in a coastal city, Xiamen, China, *Chemosphere*, 119, 530-539, <https://doi.org/10.1016/j.chemosphere.2014.07.024>, 2015.