

Interactive comment on “Observations of atmospheric mercury in China: a critical review” by X. W. Fu et al.

Response to comments of Referee #1

We would like to acknowledge the anonymous reviewer dedicating the time to read our manuscript and provide valuable suggestions. We greatly appreciate the comments and have revised the manuscript according to the reviewer’s recommendations. The corrections were marked in blue in the revised manuscript.

RC- Reviewer’s Comments; *AC* – Authors’ Comments

RC: This is an extremely long paper that provides an overview of atmospheric Hg data collected in China and the North South China Sea. This manuscript needs to be more concise. The paper needs to be reconfigured to discuss the limitations of the data. As written it is not really appropriate for ACP because it is a data summary, not a paper that moves science forward. I have some suggestions below for improvement; however, I apologize I just do not have several days to work on this paper so I will just provide some general comments.

First, the title is misleading for the paper is not a critical review, but a long summary of atmospheric data collected in China. I think the summary is useful, but a better discussion of the limitations of the data, a critical review of what is available, and a discussion of what needs to be done to move atmospheric Hg research forward in China given the current evolving thinking on atmospheric Hg measurements are needed.

AC: **We agree with the reviewer that the manuscript can provide more in-depth analysis of available data. In the revised manuscript, we (1) provided the overall spatial patterns of atmospheric Hg forms and relate the patterns to anthropogenic Hg emissions and atmospheric Hg transformations (Section 3.1.1 and 3.1.2); (2) summarized the seasonal trend of GEM and highlighted the role of monsoon dominated transport in the seasonal trend of GEM (Section 3.4); (3) for the first time, presented the long-term trend of atmospheric GEM in China and compared the observed characteristics to those found in Europe and North America (Section 3.6); (4) estimated atmospheric GEM emissions in China over the past decade using reported GEM/CO ratios and discussed the implications for improving Chinese Hg emission inventories (Section 3.7); and (5) discussed the limitations, implications of the findings to date, as well as outlined the future research needs for atmospheric Hg in China (Section 3.10).**

The added sections are new synthesis based on the data from previous studies of atmospheric Hg in China.

RC: Line21 pg 11928- the authors describe the paper as an integrated synthesis. The authors should describe the data within the limitations of their measurements. For example, discussion of how the GEM/PBM/GOM measurements were made and the limitations of these measurements need to be laid out carefully. They should look carefully at their data and see if results can be explained within knowledge of measurement limitations. When I think of ACP papers I think they should be advancing knowledge not just summarizing data. One way to deal with the length is to move a lot of information to the supplemental information, and then critically review data that is available and what is needed to improve understanding.

AC: **We have rewritten the motivations of this paper. The limitations of the measurements were described in detail in Section 3.10. The measurements methods of atmospheric Hg in China and its limitations were discussed in Section 2.1-2.4.**

RC: Throughout- 1-the paper needs English grammar editing; 2- atmospheric Hg species need to be changed to forms (Species means the chemical compounds are being measured and they are not. The authors are reporting simply on operationally defined forms); and 3- the word level should be replaced with concentration.

AC: We have gone through a thorough round of editorial revision to address the readability issue of the original manuscript. Atmospheric Hg species and levels were changed to atmospheric Hg forms and concentrations in the revised manuscript, respectively.

A few examples of issues:

RC: Line 19 pg 11926 fold instead of folds line 20 pg 11926 what is activity data line 23 and 24 pg 11926 remove the line 25 pg 11926 suggesting is speculative language especially since they do not have dry deposition fluxes and only uptake of GEM by vegetation is estimated. Thus, dry deposition is not adequately considered.

AC: ‘3 folds of’ was changed to ‘2-3 times larger than’, ‘activity data’ refers to the statistics of human activities (e.g., coal consumption, industrial productions, etc.) utilized for estimating anthropogenic Hg emissions.

We agree with the reviewer that the uptake of GEM by vegetation does not appropriately represent Hg dry deposition in general sense. Quantifying dry Hg deposition is a global challenge. There is not a standard method for measuring Hg dry deposition. In the revised manuscript, we summarized the data derived from modeling approaches as well as litterfall and throughfall measurements (Section 3.9). These methods were used in previous studies and can give an approximate estimate of atmospheric Hg dry deposition, although both are subjected to uncertainties. Estimates of dry deposition are critical for constructing regional and global atmospheric Hg budget and estimating emission outflow from China. Therefore, we decided to keep this section have provided necessary cautionary wording in the revised manuscript.

RC: Line 4 pg 11934 what is significant? No statistics have been done.

AC: We showed the statistical analysis in line 193-197 on page 7.

RC: Figure 7 These are diel not diurnal trends (diurnal means day; nocturnal means night; diel means 24 hours). Can the authors offer some explanation for the amplitudes of the diel curves? There are no error bars. What happens if this is broken out into seasonal trends? Are there a consistent number of measurements for each location? Were these all measured simultaneously? Why are there no diel patterns for some sites and not for others? Details on the locations are needed as well as the measurements are they standardized? Did these researchers follow a standard protocol?

AC: The word ‘diurnal’ was changed to ‘diel’. We have classified the diel variations of atmospheric GEM into two distinct trends and discussed the potential factors of influence in Figure 8 and section 3.5 on page 13. All these data were measured using the same method (Tekran 2527 A/B) with standardized analytical protocols. The diel trends in different seasonal were discussed in detail in earlier studies and the present study is aimed to further analyze and compare the diel trends observed at different monitoring sites, and then propose potential mechanisms responsible for the diel trends. These diel trends were derived from one and/or more than one year of continuous monitoring. The precision of hourly GEM concentration was much lower compared to the diel variations of atmospheric GEM. Also, we did not have the standard deviations for 2 of the 9 remote monitoring sites. Therefore, we did not add error bases to Figure 8 in the revised manuscript.

RC: Figure 8 there does not look like there is a diel trend and the years do not appear to be significantly different. Again there are no statistical analyses.

AC: We deleted the figure of diurnal and monthly trends in Guiyang, and added a figure regarding the annual mean GEM concentrations at Mt. Changbai from 2009 to 2015 in Figure 9. The paired T test results in Guiyang city between 2001 and 2010 were shown in 455 on page 14.

RC: Section 3.1.2 and 3.1.3 what are the uncertainties. What are the ozone and relative humidity measured at each location? How were the inlets to the instruments configured? Were the locations in clearings or in the forest?

AC: Uncertainties of the sampling methods of PBM and GOM were discussed in Section 2.2. The precision in terms of data standard deviations was shown in Table 1. Ozone was measured only at Miyun. The relationship between GOM and PBM and relative humidity were discussed in peer-reviewed literature. The sampling inlets of instruments were generally 2-3 m above ground. The information of sampling method, locations, ozone data and relative humidity can be found in the references cited in the revised manuscript.

RC: Line 21 pg 11935 significant figure is not accurate based on the detection limit.

AC: PBM in China and other regions over the world were measured using similar analytical protocols. We agree with the reviewer that measurements of BPM have large uncertainties. However, considering that the concentrations of PBM in China were generally several to dozens of times higher than those observed in Europe and North America, such significant difference should be discussion in this review paper.

RC: Line 25 pg 11937 fast deposition is speculative. What are the deposition velocities? How long would it take to deposit?

AC: These information were shown in line 271-273 on page 9.

RC: Section 3.2 describe how measurements were made and how they know this is GEM

AC: The measurements of GEM in South China Sea (SCS), Yellow Sea, and East China Sea (ECS) used the Tekran 2537 analyzer, and measurements of GEM in Bohai Sea used RA-915 +Hg analyzer. These information was provided in section 2.1. The reason for using the item of 'GEM' throughout the manuscript has been shown in section 2.1.

RC: Section 3.4. This is an interesting section. It would be good to have some trajectory related data to back up their general discussion.

AC: We have developed a trajectory based metrics for long-range transport to explain the influence of monsoon on the seasonal trends of GEM in remote and urban areas of China (Figure 7 and line 373-393 on page 12).

RC: Last sentence pg 11945- very speculative.

AC: This conclusion was derived from the discussion on diel trend of GOM at remote sites in China. It is consistent with the previous finding in North America reported in peer-reviewed literature.

RC: Beginning of section 3.6. Data from very limited sites are not adequate for establishing long term global trends, i.e. see limitations discussed of Chinese data page 11947 line 6

AC: The selected sites in Europe, North America and South Africa are remote sites far away from anthropogenic emission sources. The measured GEM concentration at the sites represent for the regional atmospheric GEM background. Therefore, the long-term studies at these sites should reflect the temporal changes in atmospheric Hg budget at global scale. We agree with the reviewer that more long-term observations should be taken into account before we make the final assessment. That is why we supplemented the updated long-term observations in China. Our results suggested that domestic Hg emissions played a more important role in the observed temporal trend of atmospheric GEM than global emission influence.

RC: Pg 11948 it seems that a detailed discussion of what was used to establish the emission estimates and the limitations of this are warranted.

AC: We have made revisions for this section as shown in line 476-487 on page 15 and in Table 2.

RC: The authors seem to have left out discussion of natural emissions and the importance of these with respect to air concentrations.

AC: The influence of natural emissions were discussed in line 218-219 on page 7, line 364-367 on page 11 and line 416-418 on page 13.

RC: In their list for moving forward I would say that they need to measure dry deposition fluxes and they need to accurately measure GOM and PBM in air.

AC: We added section 3.10 to show the limitation, implications, and future research needs, which include the need for studying Hg dry deposition in China.