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 3.1.3what 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 sapling method, locations, ozone data and relatively 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 271-273on 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 11947line6

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.**

Response to comments of Referee #2

AC: **We greatly thank the reviewer for providing the critiques. These comments are very important for improving our manuscript. We evaluated these comments carefully and have revised the manuscript based on the comments. The corrections were marked in blue in the revised manuscript.**

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

RC: This manuscript summarizes published measurements of atmospheric mercury (Hg) in China over approximately the past two decades. As stated, the objectives of this synthesis are to delineate (1) the spatial and temporal patterns of atmospheric Hg, (2) the long-range transport patterns of atmospheric Hg,

and (3) the impacts of Hg emissions on atmospheric Hg distribution and deposition in China. A critical review of Hg measurements in China is an important contribution to the scientific literature, given that Hg emissions and atmospheric concentrations in China are higher than many other parts of the world. Thus it is important to assess long-term trends in Hg emissions and atmospheric concentrations in China. It is also important to consider the implications of these current trends for long-range transport to downwind regions as well as the anticipated future trends given any efforts to regulate Hg emission at present or in the future.

That being said, the manuscript provides primarily a literature review of atmospheric Hg measurements in China, and very little new analysis of that published data is provided. For this to be a "critical review" the authors need to do a much more detailed analysis of the existing data (statistical or spatial modeling, for example). Otherwise, the paper is more of an "integrated synthesis" (as stated in the Introduction) than a "critical review". For example, the only part of the manuscript resembling a "Methods" section summarizes the existing observational approaches to quantifying ambient or deposited Hg; however, there needs to be some discussion of the new data analyses (statistical approach, model interpretation) that the authors have used to synthesize and analyze the data. Some level of new statistical or spatial analysis/modeling need to be added in a revised manuscript so that the authors are in fact adding new science to this summary of existing data. Otherwise this manuscript is probably not appropriate for ACP.

AC: **We agree with the reviewer that the review paper can provide more in-depth analysis of available data in China. 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: Additionally, several of the same authors published "A review of studies on atmospheric mercury in China" in Science of the Total Environment in 2012. Is the present manuscript simply an update of the 2012 paper? Or has some new analysis been added? This needs to be explicitly discussed.

AC: **We addressed this point in line 85-87 on page 3.**

RC: The authors do establish that atmospheric Hg in China is much higher than other parts of the world (e.g. North America and Europe), but this is not necessarily a new finding or idea. Much of the discussion of individual measurement locations should be simplified throughout the manuscript, because details of individual measurement sites are provided in the cited papers and are summarized within the tables and figures. Authors should focus on the overall spatial and temporal patterns across China, as this is the new contribution they can make to the literature.

AC: **We discussed the spatial and temporal patterns specifically in section 3.1.1, section 3.5, Figure 1 and Figure 2. The relationship between GEM temporal/spatial patterns and anthropogenic Hg emissions has been also discussed in these sections.**

RC: It initially appears that the calculation of emissions using GEM/CO ratio from 2001 to 2013 might be a novel contribution to this work; however the authors have simply summarized reported GEM/CO values from the literature and computed an average value for 2001-2013. Then seemingly they extrapolate this to the year 2009. How might this average ratio (calculated for a 12-year period, based on studies that were conducted over short durations and in different locations anytime between 2001 to 2013, not continuously

from 2001 to 2013) be affected by a possible rise in atmospheric GEM as discussed in section 3.6 (which is also highly speculative)? It seems that the GEM/CO ratio they are reporting, as well as the discrepancy between calculated and inventoried GEM emissions, are critical values but it is unclear how relevant these results are given the way in which they were calculated and the limited discussion of the analysis.

AC: **We agree with reviewer that GEM/CO ratios measured over different years may change with the emission strength of GEM and CO. We have revised this section and calculated the GEM emission for different years based on the observations (line 476-487 on page 15 and Table 2).**

RC: The authors also summarize dry and wet deposition fluxes across China in section 3.9, but this follows a much longer discussion of ambient measurements on mainland China and in the marine boundary layer. Thus, relatively little attention is given to Hg deposition and, similar to the ambient data, section 3.9 only summarizes available results from the literature. Section 3.9 should either be eliminated and this paper should focus only on analysis of ambient measurements, or more work should be done to link the ambient and wet deposition measurements.

AC: **We agree with the reviewer that the lengthy discussion regarding the wet deposition flux and Hg input from litterfall dilutes the focus. Therefore we have simplified the discussion by making Section 3.9 shorter and including a brief summary of wet deposition and litterfall in China ONLY (data shown in Table 3). We also provided explanations for the generally low wet deposition flux in China, and related the observed wet deposition to ambient Hg measurements in line 544-553 on page 17.**

Specific Comments on Text and Figures:

RC: Overall, much of the paper needs editing for minor grammatical errors.

AC: **We have gone through a thorough round of editorial revision to address the grammatical issue of the original manuscript.**

RC: In the discussion of ambient sampling methods (section 2.2), it should be acknowledged that the manual method for GOM and PBM developed in China would be subject to the same GOM interferences as the Tekran speciation system, as those interferences are specific to the KCl denuder which the manual system also uses. How do these interferences affect their interpretation of GOM measurements in this paper?

AC: **We addressed the interferences and discussed the interpretation of GOM measurements in line 143-145 on page 5.**

RC: In section 3.1.3 Line 6: St. Louis is in Missouri, not Illinois.

AC: **Corrected.**

RC: The discussion of long-term trends in section 3.6 is highly speculative but the authors do acknowledge this. However, this is an area where perhaps some statistical or modeling approaches could be used to interpret the data and/or project future trends? Then some discussion of what is still needed in order to properly evaluate the long-term trends can be provided.

AC: **We have added statistical analysis of GEM data measured at Mt. Changbai from 2009 to 2015 in Figure 9. We also discussed future research needs for studying the long-term trend of atmospheric Hg in China in line 629-641 on page 19.**

RC: Figure 3 says it shows a "correlation" between GEM and PBM, but no correlation statistics are actually provided.

AC: **The correlation statistic was added in Figure 3.**

RC: In Figure 5 it is very difficult to see the two distinct types of seasonal patterns that the authors describe. Perhaps it would help to split the data into two panels, one for each of the types. For many of the sites it is impossible to tell whether the difference between summer and winter concentrations is actually significant. Also how many years of data were used to generate this figure?

AC: **We have revised Figure 5 into two panels showing the two distinct seasonal trends. These seasonal trends were derived from multiple years of continuous observations, which were listed in Table 1. We also showed the statistical analysis of the seasonal trends at remote and urban sites of China in line 346-358 on page 11.**

RC: It is unclear what Figure 6 is meant to show. Clearly there are differences in the wind fields between summer and winter, but what is the reason for also showing geopotential height since it does not seem substantially different from one season to the next? And why show only 2011-2013?

AC: **The geopotential height is generally a function of elevation and latitude. The main reason of showing the geopotential height is to show the effect of elevation on the wind fields. As shown in Figure 6, the presence of Tibetan Plateau has a strong impact on the wind field in western and southwest China. The wind fields are generally governed by monsoons, and not expected to vary significantly in different years. The modeling period of 2011-2013 is representative of the sampling period when the observations were made in China.**

RC: Similarly to Figure 5, in Figure 7 how many years of data were used to generate this plot and what is the time frame? Does it vary from site to site? If it varies by site (perhaps as suggested by Table 1) this needs to somehow be indicated or discussed here.

AC: **We have revised Figure 8 (originally Figure 7) into two panels showing contrasting diel variation. We have also explained the reasons causing the difference in the diel pattern in line 409-411 on page 13. The sampling period of the data is indicated in the caption of Figure 8 on page 36.**