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

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.**