Response to Reviewer #1

We greatly appreciate the reviewer's constructive comments, which have helped to improve the paper substantially.

Mao and colleagues present a review of published work on spatiotemporal patterns of atmospheric mercury. The authors have compiled an impressive volume of literature. I commend the authors on presenting an unbiased summary of published work. I recommend the review for publication after revision. Too much of the present manuscript feels like a reiteration of published work. The review could be greatly improved if it were more concise and provided a greater amount of critical insight.

R: We have improved the paper substantially through: (1) removing redundancy and unnecessary details; (2) summarizing common findings from multiple studies and pointing out differences between studies in each category/scenario, and (3) more importantly, providing more critical insights in the unresolved questions and recommendations for future research needs.

General Comments
The Abstract could use a statement motivating why we care about mercury in order to help appeal to a broader readership. I also suggest tightening the conclusions and including at least one future research recommendation.

R: The abstract was revised. In addition, per the reviewer's suggestions, we have added these statements in the abstract:

"Atmospheric mercury is a global pollutant and thought to be the main source for mercury in oceanic and remote terrestrial systems, where it becomes methylated and bioavailable, and hence atmospheric mercury pollution has global consequences for both human and ecosystem health."

"In examining the remaining questions and issues, recommendations for future research needs were provided, and among them again it boiled down to the most imminent need for GOM speciation measurements and fundamental understanding of multiphase redox kinetics."

The Introduction is unfocused and needs clearly stated objectives. Some of the content in the Introduction gets repeated in later sections. Delete redundancy wherever possible.

R: We have shorten the introduction by deleting most materials in the original 3rd-6th paragraphs so as to avoid redundancy and keep it focused.

The phrase "natural emissions" is used loosely and sometimes interchangeably with "reemissions" or "legacy emissions". In light of the Minamata Convention, it is important to maintain clear language here and distinguish between natural primary sources (volcanism,

outgassing of enriched mercuricferrous belts) and anthropogenic sources being remitted by land and ocean.

R: It is an important point. Corrections were made throughout the text.

Be concise. Delete unnecessary text. The current manuscript feels unnecessarily long.

R: See our response to the general comments above.

Old data (1960-80s) is included in comparisons alongside modern data -- is this really a valid comparison? At a minimum, it seems like it would be appropriate to comment on the major differences in analytical methods and the robustness of old data. I worry about the reliability of older data (Gustin et al., 2015).

R: Point taken. The inclusion of old data was an attempt for the completeness of the review. We agree the values were not comparable to those in more recent studies. Hence, the comparison between the old and more recent data was mostly removed, and few retained was revised with cautionary notes.

Specific Comments

Line 62: Please include a citation for biodegradation. Biodegradation isn't a process commonly associated with atmospheric mercury.

R: This sentence is the continuation of the previous one and is followed by the next, referring to mercury in the atmosphere and other spheres together in the Earth system.

Lines 101-106: Mao & Talbot (2012) is cited exclusively. Are there other references that could be included too?

R: The introduction was condensed significantly to avoid redundancy. This reference was removed in the introduction together with other material.

Lines 536-549: Rivers and wastewater cannot explain North Atlantic trends in Soerensen et al. (2012) (Amos et al., 2014).

R: Amos et al. (2014) was added to counter the findings from Soerensen et al. (2012).

Line 527: The Pinatubo hypothesis is not widely embraced. I do not recommend including it in the review.

R: The manuscript has been greatly revised and edited. This part has been removed in the revised version.

Line 811: Why would ship emissions be important? My understanding is most ships burning crude oil, which is low in Hg (Pironne et al., 2010).

R: Sprovieri et al. (2010) were making general statements, not exclusively with regard to Hg, about ship emissions becoming a more important source of contaminants as emissions from other sources were being more stringently controlled, and the Mediterranean was a place where busy shipping routes ran close to population centers. The reference to Sprovieri was revised to reflect this point and the reviewer's concern.

Line 1468: "Refuting… large oceanic emissions". Please include a rationale for this conclusion. This is not an obvious conclusion from the review. If it's true, it's significant, but the conclusion needs to be buttressed with supporting evidence here in the Summary & Recommendations section.

R: This point was made by Temme et al. (2003a) based on the average NH/SH ratio of TGM hemispheric median values and the higher variability in NH TGM concentrations from their three cruises. Mason at al. (1994) hypothesized that oceanic emissions were a large source to atmospheric Hg. Temme et al. (2003a) "refuted" this point by saying that two thirds of oceans are located in the southern hemisphere and if oceanic emissions were truly a large source, the large NH/SH ratio and large variability of TGM in the NH would not have been likely. Temme et al. (2003a)'s cruises measurements covered the largest areas in both hemispheres and were conducted along the same path three times and hence cited. However, these are both hypotheses, and more studies suggested oceans as a source in various oceanic regions. "Refute" is the word Temme et al. (2003a) used in their paper. We changed "refuting" to "contradicting" now.

Lines 1484-85: "Global distributions… remain lacking…" Delete? This statement is not particularly helpful.

R: We thought that this point is in fact quite important. For a compound such as ozone, there have been numerous studies providing global distributions using satellite retrievals, in situ measurements, and model simulations, so we have a fairly good idea of the global distribution of ozone. In comparison, we do not really have such knowledge of GEM, GOM, and PBM global distributions, despite decades of monitoring and modeling studies. A lot of it remains controversial and speculative due to the lack of measurement data in the southern hemisphere, in the marine boundary layer, and in upper air, and due to a lack of the model simulations that we have confidence in.

Line 1492: "…trends derived from such data suggested composite information instead…" Perhaps there is a typo in the sentence? I'm unsure what the intended meaning is.

R: We agree that the point did not come out right. The summary section was rewritten, and the relevant point was reworded.

Figure 1: Break the y-axes, so you better see the variability in the data and the plots aren't dominated by one extra-large error bar.

R: Done.