Response to comments on "Global Observations and Modeling of Atmosphere-Surface Exchange of Elementary Mercury – A Critical Review" *by* W. Zhu et al.

We thank the reviewers for their thoughtful and constructive comments that help improve the quality of our manuscript. We have incorporated the reviewers' suggestions in the revised manuscript. Our point-to-point response to the reviewers' comments are shown below.

Anonymous Referee #1:

Overall comments:

Overall this is a very well written and fully researched review paper. In addition to simply reviewing existing studies, this paper performs original analysis of the compiled datasets in order to make large scale observations. This paper provides a comprehensive assessment of the current understanding of the atmospheric surface exchange of Hg and recommend it for publication in Atmospheric Chemistry and Physics.

The only complicating factor with the publication of this paper is that a separate group published a fairly similar paper very recently: Agnan et al., New Constraints on Terrestrial Surface-Atmosphere Fluxes of Gaseous Elemental Mercury Using a Global Database. ES&T 2016.

Given the similarities in objective and scope, I think this paper by Zhu et al, needs to: 1) acknowledge this separate paper in results and discussion, and 2) specifically identify how their paper is unique from Agnan et al., 2016, and 3) discuss similarities and differences in the two papers findings.

Response: We deeply appreciate the reviewer for the supportive comments and constructive suggestions to our manuscript. We have recognized and review the paper by Agnan et al. (ES&T 2016); and agree with the reviewer that a more robust discussion pointing the specific contribution of this paper in addition to the paper by Agnan et al. The similarity of the two papers are mainly on the overlap on existing literature on Hg flux measurement. The major differences between the two papers are: (1) approaches in the data compilation and synthesis (e.g., the statistical treatments), (2) the coverage of flux data over different landuses (soil, forest, snow, freshwater, and ocean in this paper as compared to terrestrial surfaces in Agnan et al.), (3) the inclusion of mechanistic discussion on flux quantification approaches (e.g., enclosure and micromet measurements) and air-surface exchange processes (e.g., confounding influences by environmental factors), (4) the inclusion of flux modeling approaches and scale-up of flux data for global cycle implications, and (5) the inclusion of more up-to-date field data and exclusion of laboratory data in the synthesis.

In the revised manuscript, we have provided an additional section to recognize the contribution by Agnan et al (2016) and laid out the differences of the two papers, cf. line 82-88. We have also cited Agnan et al. (2016) in other parts of our manuscript (line 284-293, line 498-501, line 603-605, line 648-649).

Specific comments:

Comment #1: Line 261: Very interesting result. Glad to see this analysis. A little more information is needed. The samples sizes are 229 and 39, but it is not clear if these numbers represent daily average values, hourly values, etc. A little more discussion about what constitutes a measurement would be helpful. Also, within the <0.3 ug/g cutoff, were there significant differences in the Hg concentrations between DFC and MM areas? If not, this would help build the case for the analysis.

Response: We thank the reviewer for pointing this out. The information regarding the measurement conditions have been added (line 271-272). Because the total Hg concentrations in soil substrates are frequently not reported in the literatures, particularly in those studies from background sites, it is not reliable to compare the substrate Hg concentrations between DFC and MM measurements due to the small available sample sizes.

Comment #2: Line 262: Why was 0.3 ug/g used as a cut-off point?

Response: The use of 0.3 $\mu g\ g^{-1}$ as the threshold of less human activity influenced background surfaces is based on investigation of background concentration from literatures and in line with the criteria used in Agnan et al. (2016). More important, Hg in a relatively low level surfaces are in general homogeneous than contaminated sites (Gustin et al., 1999), which reduced the uncertainty raised by footprint differences in comparing DFC and MM techniques.

Agnan, Y., Le Dantec, T., Moore, C. W., Edwards, G. C., and Obrist, D.: New constraints on terrestrial surface–atmosphere fluxes of gaseous elemental mercury using a global database, Environ. Sci. Technol., 50, 507-524, 2016.

Gustin, M. S., Lindberg, S., Marsik, F., Casimir, A., Ebinghaus, R., Edwards, G., Hubble-Fitzgerald, C., Kemp, R., Kock, H., Leonard, T., London, J., Majewski, M., Montecinos, C., Owens, J., Pilote, M., Poissant, L., Rasmussen, P., Schaedlich, F., Schneeberger, D., Schroeder, W., Sommar, J., Turner, R., Vette, A., Wallschlaeger, D., Xiao, Z., and Zhang, H.: Nevada STORMS project: Measurement of mercury emissions from naturally enriched surfaces, J. Geophys. Res.-Atmos., 104, 21831-21844, 1999.

Comment #3: Line 288: change matters to matter.

Response: It has been changes accordingly.

Comment #4: Line 285: Two factors that have been shown to affect soil-air Hg fluxes are grain size and soil disturbance. Only a couple of studies have shown this, but may want to consider including these two factors in the discussion if the goal is to be comprehensive as possible.

Response: We thank the reviewer for pointing out the two factors that are not as extensively studied: grain size and soil disturbance, and have provided the discussion in the revised manuscript, line 326-329.

Comment #5: Line 339. There is a paper by Mazur et al. 2014 in Science of the Total Environment that has a similar focus: the impact of forestry operations on surface-air Hg fluxes.

Response: The results of Mazur et al. (2014) has been incorporated in the discussion and the reference has been added in the citation list, cf. line 348-349.

Comment #6: Line 346: Suggest changing to "more recent" instead of just "recent". This idea has been around for more than a decade now.

Response: It has been changed.

Comment #7: Line 352: remove excess Hg0.

Response: The excess Hg^0 has been deleted.

Comment #8: Line 422: Need more information to support this statement. Earlier the text focuses on photo-pathways and this is a big jump without sufficient explanation.

Response: We agree with the reviewer on the suggestion. Previous statement was incorrect as showed in Fig.5, which has been corrected and reworded as "Both dark abiotic and biotic redox transformations are suggested to be involved (Fig. 5)", cf. line 435-436.

Comment #9: Line 472: remove "got flux calculation".

Response: It has been removed.

Comment #10: Line 476: in "the" literature

Response: It has been inserted into the text.

Comment #11: Line 558: This paragraph should also discuss the work of Kuiken et al, 2008 part 1, which shows the opposite trend....lower emission in summer due to drier conditions and lower light from more leaf cover. In the scaling paper, Hartman et al, 2009 comes to the same conclusion.

Response: We thank the reviewer for pointing this out. The phenomenon of low flux in summer as a result of low light and drier conditions has been added in the discussion, cf. line 578-580.

Comment #12: Line 601. Double check that Gustin et al, 2003 used a multivariate approach using soil Hg, flux and solar radiation. Or did that paper look at these variables separately.

Response: We thank the review for the cautionary remark. We have checked into Gustin et al. (2003) and discussed the influence of those environmental factors separately [page 345 and 347] with the citation.

Comment #13: Line 700. Remove "in"

Response: It has been removed from the text.

Comment #14: Line 713. This is a great summary of knowledge gaps, glad to see this in the paper.

Response: We thank the reviewer for the positive comment.