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 and editorial corrections in the revised manuscript. Our point-to-point response to the reviewers' comments are shown below.

Anonymous Referee #2:

Overall comments:

This paper is a thorough review of measurement and modeling studies of elemental mercury. The depth and extent of the analyses of available data does indeed make this a critical review rather than just a literature review. The discussion on the advances in the measurement techniques is beneficial. This paper provides a necessary addition to the scientific community's GEM literature and aids in furthering our understanding of the air-surface exchange of atmospheric mercury. With some minor editing on a technical scale, I recommend the publication of this paper in Atmospheric Chemistry and Physics.

I agree with Reviewer #1's comments regarding the paper by Agnan et al. (2015). The only discussion on their paper was relating to the measurement method. It would be interesting to see a discussion on the findings of the two papers and how they complement each other.

Response: We deeply appreciate the reviewer for the supportive comments and constructive suggestions. Our special thanks to the reviewer for providing detailed editorial remarks. As discussed in our response to reviewer #1, we have recognized the review 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 influence 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 108: Is this a possible typo that <1 Hz is considered a higher frequency?

Response: We thank the reviewer for pointing it out and would like to clarify it. The text has been rephrased as "Later on, monitoring ambient air Hg0 with relative higher frequency (up to 1 Hz) was achieved by using Lumex RA-915+ Zeeman atomic absorption spectrometry (AAS) analyzer operating without pre-concentration", cf. line 113-114.

Comment #2: Line 113: The use of "but" in this sentence suggests that the higher detection limit of 0.35 ng m^{-3} is a negative aspect, but could that sensitivity be considered a benefit of this sensor over previous ones?

Response: Yes, the detection limit at 0.35 mg m^{-3} is a negative aspect in terms of analytical accuracy for Hg vapor because of the low concentration gradient typically observed in most flux quantification techniques (typically <0.4 ng m^{-4}, Zhu et al., 2015). However, the high frequency of this sensor (25 Hz) is a benefit among available Hg vapor detection techniques because it offers the possibility of using eddy covariance method to measure Hg flux even it is limited over contaminated surfaces only, e.g., Pierce et al., 2015.

Pierce, A. M., Moore, C. W., Wohlfahrt, G., Hörtnagl, L., Kljun, N., and Obrist, D.: Eddy covariance flux measurements of gaseous elemental mercury using cavity ring-down spectroscopy, Environ. Sci. Technol., 49, 1559-1568, 2015.

Zhu, W., Sommar, J., Lin, C. J., and Feng, X.: Mercury vapor air–surface exchange measured by collocated micrometeorological and enclosure methods - Part I: Data comparability and method characteristics, Atmos. Chem. Phys., 15, 685-702, 2015.

Comment #3: Lines 185-187: This sentence, while accurate, discusses the lack of the ability of this sensor at background sites. This study however was over Hg-enriched soils and the sensor performed well over Hg-enriched sites. Would it be useful to note this as an advantage to this method considering the high number of sites that are Hg-enriched?

Response: We appreciate this insightful comment. It is indeed worth highlighting the possible application of CRDS-EC over Hg-enriched sites. The discussion has been revised to (cf. line 190-193):

Pierce et al. (2015) reported the first field trial of CRDS-EC flux measurement over Hg-enriched soils with a flux detection limit of 32 ng m^{-2} h^{-1}, offered the opportunity for high frequently monitoring Hg^0 flux from Hg-enriched surfaces. However, the present state of development of CRDS-EC must be further advanced for Hg^0 flux measurement at most, if not all, background sites.

Comment #4: Line 547: Perhaps consider mentioning why the fluxes would be higher in Europe than East Asia prior to 2002 and during summer and/or daytime.

Response: We agree with the reviewer on the suggestion. Detailed discussion of high fluxes over freshwater bodies in summer and daytime have been discussed in Section 3.3 and 4.3.2. We have also revised the sentence to (cf. line 561-563) "The flux over freshwater bodies in Europe is somewhat higher than those measured in East Asia (6.5 vs. 4.6 ng m^{-2} h^{-1}, p=0.40, ANOVA). These data were obtained mostly prior to 2002 (n=9) or during summer time and daytime (n=8) subject to higher blank larger extent of photo-reduction and evaporation."

Comment #5: Line 558: There are some studies that suggest the opposite (e.g. Lee et al., 2000; Fristche et al., 2008).

Response: We thank the reviewer for the suggestion, the discussion of opposite seasonal flux variation has been provided in the revised manuscript with the corresponding references, cf. line 578-582.

Comment #6: Lines 577; 593; 612; 627: The titles of the subsections in Section 4.4 include statements. Does this possibly change the flow of the paper?

Response: We agree with the reviewer that the full statements in the titles are somewhat distracting. In the revised manuscript, the subtitles have been incorporated into the text to maintain the flow of the text.