Reply to Comments from Reviewer #2

We thank the reviewers for their valuable comments which help us improve the quality of the manuscript. We have carefully revised our manuscript following the reviewers' comments. Point-by-point responses are given below. The reviewers' comments are in black and our responses are in blue.

Comment:

A method section is missing. The authors may want to provide a Methodology section to cover the following items, how the literature search/review was conducted, what is the scope of the literature search, what are the primary source of publications (e.g. peer reviewed journal articles, government reports), restrictions if any (e.g. by year of publication, or by language).

Response:

We thank the reviewer for the suggestion. However, a method section is not quite common for a review paper. Considering the manuscript is already very long, we have not added a method section. Instead, we have modified the last paragraph of the Introduction part to make it more clear what the purpose of this review work is. Please refer to Lines 75–88 in the revised manuscript:

"Significant efforts have been made in the past decade for quantifying atmospheric Hg deposition through both direct observations and model simulations, especially on dry deposition (Lyman et al., 2009; Zhang et al., 2009; Holmes et al., 2011; Lai et al., 2011; Castro et al., 2012; Gustin et al., 2012; Peterson et al., 2012; L. Zhang et al., 2012; Fang et al., 2013; Sather et al., 2013; Lynam et al., 2014; Sather et al., 2014; Huang and Gustin, 2015a; Weiss-Penzias et al., 2016a; Zhang et al., 2016b; Hall et al., 2017; Sprovieri et al., 2017). Yet large uncertainties still exist due to limitations of current methods for Hg deposition measurements and modeling (Gustin et al., 2015). The purpose of this paper is to give an overview of the uncertainties in the observation and simulation of global speciated atmospheric Hg deposition to terrestrial surfaces. In this paper, we investigated results from the observation and simulation of global Hg

deposition, reviewed methods adopted for Hg deposition measurements and modeling, estimated the uncertainties of different methods for different Hg deposition forms, and summarized the overall uncertainty level of global Hg deposition."

Comment:

The scope of the review needs more justification. The title reads, "Global deposition of speciated atmospheric mercury to terrestrial surfaces: an overview". The rational of excluding the water surfaces (Figures 1, 6, 7 do include water through) and snow/ice over land should be presented.

Response:

We have modified the title and put focus on the uncertainties in the observation and simulation of global speciated atmospheric Hg deposition to terrestrial surfaces. Surface type is not our primary concern in the revised manuscript. The "water" surfaces here refer to the terrestrial surfaces near water. We have added the explanation to both figure captions and the main text. Please refer to Lines 146–147 in the revised manuscript:

"The "water" surfaces here refer to the terrestrial surfaces near water, e.g., coastal, offshore, and lakeside sites."

Comment:

The scientific contribution could be enhanced significantly. The manuscript as written is a somewhat descriptive presentation of estimation methods (sections 2 and 3) and Hg deposition values (sections 4 and 5). Consequently, there is a lack of new insights and findings. The authors are encouraged to conduct a rigorous research leading to more depth discussion that highlights the advancement, challenges, and directions for future research. Some potential topics are listed below (also see sample papers and a sample weblink at the end) 1) Comparison of co-located measurements with different techniques 2) Comparison of Hg deposition estimates by different models 3) Model-measurement comparison 4) Observed/predicted changes in Hg deposition due to changes in quantity of Hg emissions in local, regional or globe scale 5)

Observed/predicted changes in Hg deposition due to changes in profiles (e.g. the percentage of each Hg species in total emission) of Hg emissions in local, regional or globe scale 6) Contributions to observed/simulated Hg dry deposition from different sources or regions 7) The major sources of uncertainty in Hg deposition estimates and how to reduce those uncertainties 8) What is the knowledge or data gap (relevant to Hg deposition) that hinders our understand of the global Hg cycle, or the development and evaluation of emission control measures?

Response:

We greatly appreciate the valuable comment. We agree with the reviewer that the contribution of the manuscript was not clear. We have reorganized our manuscript, made significant revision, and added more discussion on the uncertainties in the observation and simulation of global speciated atmospheric Hg deposition to terrestrial surfaces. We believe the revised manuscript is more focused and more informative. Please refer to the revised manuscript.

Comment:

The “Bidirectional air-surface exchange model for GEM” is presented. However, dry deposition of GEM is estimated in many field studies and model simulations, including most GEM dry deposition data presented in the manuscript. Thus, the authors may want to include dry deposition models of GEM.

Response:

As mentioned in responses to previous comments, we have revised the manuscript extensively to focus on the uncertainties. The bidirectional model is a more commonly used model in recent years, so we have estimated the uncertainty in the simulation of GEM deposition flux based on the bidirectional model instead of the resistance model. Previous review work (Zhang et al., 2009) has discussed the two types of models in detail.

Reference:

Zhang, L. M., Wright, L. P., and Blanchard, P.: A review of current knowledge concerning dry deposition of atmospheric mercury, Atmos. Environ., 43, 5853–5864,

10.1016/j.atmosenv.2009.08.019, 2009.

Comment:

Please provide facts to support your statements, e.g. “For PBM dry deposition, a size-segregated resistance model is more and more widely applied” (L312)

Response:

The description here was not accurate. We have modified the expression. Please refer to Lines 700–703 in the revised manuscript:

“For PBM dry deposition, resistance models regarding both fine and coarse particles are more and more widely applied based on the theory that v_d for atmospheric particles strongly depend on particle size (Dastoor and Larocque, 2004; Zhang et al., 2009; Zhang and He, 2014).”

Comment:

Most materials presented in sections 2 and 3 can be found in previous review/research papers, because those techniques have been around for a while. The authors could provide a summary table and direct the interested readers to those review/research papers, instead of a lengthy description of each method. Another option is to provide a comparative review of those methods and to include strength, weakness, recent advancements if any, and application issues.

Response:

We have modified the discussion on the methods for observation and simulation of Hg deposition to follow the estimation of uncertainties in the revised manuscript. Method details have been lessened.

Comment:

Section 4.3 (Forest deposition or Deposition over forests) could be better placed in section 5 (Global Hg deposition on different terrestrial surfaces).

Response:

We have reorganized the whole manuscript. The uncertainty analysis for forest Hg

deposition is based on methods for litterfall and throughfall deposition. Therefore, it is in parallel with wet and dry deposition. Please refer to Section 3.3 and 4.3 in the revised manuscript.

Comment:

If the authors decided to keep the equations, please 1) provide unit of each variable, 2) provide the source of each equation, 3) clarify the expansion factor in equations (8) and (9). Is it an expansion from a measurement in a small area to a forest? 4) explain how to calculate two resistances with equation (16).

Response:

We think variable unit is not quite necessary for the uncertainty analysis in this review work. We have added the sources of each equation and clarified the "expansion factor". Equation (16) means that there are two sets of v_g (gravitational settling velocity), R_a (aerodynamic resistance), and R_s (surface resistance) for fine and coarse particles, respectively.

Comment:

Please state the mechanism of Hg deposition via cloud/fog at high elevation sites (L258).

Response:

Cloud and fog can scavenge Hg in the atmosphere. At alpine or coastal sites, Hg can deposit onto the ground through cloud or fog. Cloud or fog is not able to be collected by precipitation samplers. Studies (Stankwitz et al., 2012; Weiss-Penzias et al., 2016b; Gerson et al., 2017) have shown that cloud and fog water have higher Hg concentration than rain water in the same region, and cloud and fog could have a remarkable contribution to Hg wet deposition in high-elevation forests and near-water surfaces.

References:

Stankwitz, C., Kaste, J. M., and Friedland, A. J.: Threshold increases in soil lead and mercury from tropospheric deposition across an elevational gradient, Environ. Sci. Technol., 46, 8061–8068, 10.1021/es204208w, 2012.

Weiss-Penzias P., Coale K, Heim W, Fernandez D, Oliphant A, Dodge C, Hoskins D,

Farlin J, Moranville R, Olson A. Total- and monomethyl-mercury and major ions in coastal California fog water: Results from two years of sampling on land and at sea. Elem. Sci. Anth., 4, 1–18, 10.12952/journal.elementa.000101, 2016b.

Gerson, J. R., Driscoll, C. T., Demers, J. D., Sauer, A. K., Blackwell, B. D., Montesdeoca, M. R., Shanley, J. B., and Ross, D. S.: Deposition of mercury in forests across a montane elevation gradient: Elevational and seasonal patterns in methylmercury inputs and production, J. Geophys. Res. Biogeo., 122, 1922–1939, 10.1002/2016jg003721, 2017.

Comment:

L456, the authors may want to distinguish the net emission fluxes from "natural GEM emission sources".

Response:

We have modified the expression according to the comment. Please refer to Lines 217–220 in the revised manuscript:

"The four Asian sites using micrometeorological methods all show negative values (-36.3 ± 19.6 μg m^{-2} yr^{-1}), indicating the role of East Asia as a net emission source rather than a net deposition sink (Luo et al., 2014; Luo et al., 2016; Ci et al., 2016; Yu et al., 2018)."

Comment:

Figure 6, "precipitation levels" or "annual precipitation"?

Response:

We have modified to wording accordingly.

Comment:

The papers from which data were obtained to generate each figure could be tabulated and presented as Supplement Information.

Response:

We have tabulated the raw data and created a Supporting Information file.

Comment:

There are quite a few awkward sentences and word choices, e.g. "*Ci* is the total Hg concentration in precipitation water" (L193), "Usually, GOM and PBM contribute equivalently to Hg wet deposition (Cheng et al., 2015)." (L206), add "GEM dry deposition is equivalent to GOM and PBM dry deposition, even significantly higher than in forests" (L535), "consequently exhibit significantly high litterfall Hg deposition fluxes." (L560), "Water surfaces could affect Hg wet deposition through fog scavenging." (L580), "The contribution GEM dry deposition has been underestimated previously." (L596), "Cloud, fog or even dew Hg deposition needs careful investigation" (L599), please rephrase.

Response:

We thank the reviewer for the detailed comments. We have rephrased or deleted these sentences. Please refer to the revised manuscript.

Comment:

There are some contradicting or confusing statements, e.g. "Based on available measurements of PBM size distributions and fine/coarse PBM mass ratios, Zhang et al. (2016b) assumed 30% of the total PBM mass to be coarse particles in order to estimate total PBM dry deposition flux based on the theory that PBM has the same proportion in both fine and course particles." (L318)

Response:

We have modified the statement. Please refer to Lines 706–709 in the revised manuscript:

"Based on measurements of particle size distributions and Hg mass distribution between fine and coarse particles, Zhang et al. (2016b) assumed that coarse particles account for 30 % of the total PM, and the Hg mass concentrations on fine and coarse particles are consistent."

Comment:

Avoid the use of first person, i.e. “we”.

Response:

We have avoided the use of “we”.