

Reply to Comments from Reviewer #1

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:

This paper summarizes the important processes controlling atmospheric deposition of Hg. The topic is important, and new knowledge is available in the literature, so a review paper on this topic is a good and useful product to the broad scientific research community. However, there have been recent review papers that have largely covered the same topics and ideas, which leaves some doubt about this paper as one that makes a large contribution to the literature.

Response:

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 abstract does not put forth many new ideas.

Response:

We have modified the abstract substantially based on the revised manuscript. Here is our updated abstract:

“One of the most important processes in the global mercury (Hg) biogeochemical cycling is the deposition of atmospheric Hg, including gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM), to terrestrial surfaces. Results of wet, dry, and forest Hg deposition from global

observation networks, individual monitoring studies, and observation-based simulations have been reviewed in this study. Uncertainties in the observation and simulation of global speciated atmospheric Hg deposition to terrestrial surfaces have been systemically estimated based on assessment of commonly used observation methods, campaign results for comparison of different methods, model evaluation with observation data, and sensitivity analysis for model parameterization. The uncertainties of GOM and PBM dry deposition measurements come from the interference of unwanted Hg forms or incomplete capture of targeted Hg forms, while that of GEM dry deposition observation originates from the lack of standardized experimental system and operating procedure. The large biases in the measurements of GOM and PBM concentration and the high sensitivities of key parameters in resistance models lead to high uncertainties in GOM and PBM dry deposition simulation. Non-precipitation Hg wet deposition could play a crucial role in alpine and coastal regions, and its high uncertainties in both observation and simulation affect the overall uncertainties of Hg wet deposition. The overall uncertainties in the observation and simulation of the total global Hg deposition were estimated to be $\pm(30-50)\%$ and $\pm(50-70)\%$, respectively, with the largest contributions from dry deposition. According to the results from uncertainty analysis, future research needs were recommended, among which global Hg dry deposition network, unified methods for GOM and PBM dry deposition measurements, quantitative methods for GOM speciation, campaigns for comprehensive forest Hg behavior, and more efforts on long-term Hg deposition monitoring in Asia are the top priorities.”

Comment:

There are a few missed opportunities such as when cloud/fog scavenging is mentioned the authors state: “the influence of cloud/fog scavenging is easy to neglect”. The authors should be more quantitative in their language so as to provide scientists with more concrete information on relationships and processes.

Response:

We have modified the manuscript substantially to focus on the uncertainties. We have quantified the uncertainties in both observation and simulation of different types of Hg deposition. The uncertainties in non-precipitation wet deposition have been discussed in detail. Please refer to Section 3.1.2 and 4.1.2. Discussion on the influence of cloud or fog scavenging has been added. Please refer to Lines 151–158 in the revised manuscript:

“Fog or cloud Hg deposition is not yet considered in the global Hg wet deposition observation network. However, 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. Cloud and fog scavenging of reactive Hg (GOM and PBM) could result in lower Hg concentration in precipitation.”

Comment:

Another example in the abstract that is a missed opportunity to provide some detailed information is the last line: “Future research needs have been proposed based on the current knowledge of global mercury deposition to terrestrial surfaces”. This statement is too vague and does not provide much substance. For example, in the conclusion, the 4th recommendation regarding fog, cloud, and dew is The field “requires more standardized sampling methods”. This is too vague and does not translate into a roadmap for improving the science.

Response:

We have modified the description of future research needs. Please refer to Section 6. We have also revised the abstract to make a more clear statement. Please refer to Lines 31–35 in the revised manuscript:

“According to the results from uncertainty analysis, future research needs were recommended, among which global Hg dry deposition network, unified methods for GOM and PBM dry deposition measurements, quantitative methods for GOM speciation, campaigns for comprehensive forest Hg behavior, and more efforts on long-

term Hg deposition monitoring in Asia are the top priorities.”

Comment:

My suggestion is that the authors rethink their main focus of this paper –maybe all of deposition is too broad – and provide more insights and proscriptions for future research and/or data gaps. The authors have cited a large number of references and have done considerable research in the field. An improved focus would sharpen the discussion and make the paper more interesting to read.

Response:

We thank the reviewer for the valuable comment and have taken the advice. We have sharpened the discussion in the manuscript to focus on the uncertainties in the observation and simulation of global speciated atmospheric Hg deposition to terrestrial surfaces.

Comment:

One minor comment I have is that the following statement does not make sense to me: “The slope of the relationship implies the Hg concentration in precipitation. Europe has the flattest slope among all regions, indicating its lowest Hg pollution level around the world.” Europe has the lowest Hg pollution level around the world? That does not seem correct.

Response:

We have deleted this part of discussion.