Reply to Comments from Reviewer #3

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:

In general, this paper is not easy to follow, the authors jump from one topic to another. They did not do advanced discussion. In more paragraphs, they only described methods and data, and probably two/three sentences to summarize/discuss what they learn from these methods/data. There is nothing inspiring readers. A review paper should do better than that.

Their conclusions/summaries are not new. Gustin's group has published couple review articles discussing the first three aspects in 2015, and the 4th aspect has been mentioned in multiple previous articles. I really do not find any new concepts in this article, and how can we solve the difficulties that the Hg research community is facing. For example, do the authors have any suggestion to understand behaviors of various GOM compounds in the atmosphere?

I agree this is an important research field and there are gaps which make scientists cannot fully understand global Hg cycle. A review paper related to this topic should be published to draw attention from environmental research groups. However, the way that this paper is done cannot provide useful information to scientists. I suggest the authors re-think about the article structure and put more efforts on advanced discussions.

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:

Abstract is read more like a summary than an abstract. I suggest to re-write the abstract and focus on your key aspects. Moreover, the authors must provide some potential solutions/suggests for each gap that are discussed in their conclusions.

Response:

We have rewritten the abstract 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 ±(30–50) % and ±(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:

Introduction is fine, but this is a review paper. There are more previous Hg review articles, such as Selin et al., 2007, and some key finding paper are not included in this review paper, such as Moore et al., 2014 Nature. These articles might not be directly linked to Hg deposition, but they do have indirect impacts on Hg deposition. After reading this article, I feel the authors focus on the measuring methods and numeric models, but do not discuss in advance about global deposition processes.

Response:

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. We have also added the recent modeling work for Hg wet deposition. Please refer to Section 4.1.1 in the revised manuscript.

Comment:

A summary table or multiple summary tables would help the readers to read through this section.

Response:

We have added a summary table for the uncertainties discussed in this study. Please refer to Table 1. We have also created a Supporting Information file listing all the Hg deposition studies.

Comment:

Surrogate surface: the key point of this method is the surface affinity and fluent conditions near surface, but I did not see the authors discuss these here. Huang et al.,

2011 published a paper discussing fluent conditions near KSS surface, and how this impacts mass transfer.

Response:

We have added more discussion on how the sampler designs or fluent conditions affect the uncertainty of the surrogate surface method. Please refer to Lines 382–414 in the revised manuscript:

"Different surrogate surfaces were used to measure different RM forms. Mounts with cation-exchange membranes (CEMs) are widely used for GOM dry deposition measurements (Lyman et al., 2007; Lyman et al., 2009; Castro et al., 2012; Huang et al., 2012a; Peterson et al., 2012; Sather et al., 2013). The down-facing aerodynamic mount with CEM is considered to be the most reliable deployment for GOM dry deposition measurements so far (Lyman et al., 2009; Huang et al., 2014). Knife-edge surrogate surface (KSS) samplers with quartz fiber filter (QFFs) and dry deposition plates (DDPs) were deployed for PBM dry deposition measurements (Lai et al., 2011; Fang et al., 2012b; Fang et al., 2013). However, these samplers are not well verified to reflect the deposition velocity of PBM, and hence not widely accepted. KCl-coated QFFs were used to measure the total RM (GOM+PBM) dry deposition, but failed to capture GOM efficiently (Lyman et al., 2009; Lai et al., 2011).

According to Eq. (4), the uncertainty of RM dry deposition comes from the uncertainties of RM concentration and dry deposition velocity. The uncertainty of RM concentration mainly originates from the interference of unwanted RM forms or incomplete capture of targeted RM forms. CEMs exhibited a GOM capture rate of 51–107 % in an active sampling system (Huang and Gustin, 2015b). The CEM mounts designed to measure only GOM dry deposition capture part of fine PBM (Lyman et al., 2009; Huang et al., 2014), while the KSS samplers with QFFs designed to measure only PBM dry deposition may also collect part of GOM (Rutter and Schauer, 2007; Gustin et al., 2015). Based on the RM concentration measurements and the surrogate surface method evaluations, the GOM concentration related uncertainty is estimated to be ±50 % (Lyman et al., 2009; Lyman et al., 2010; Gustin et al., 2012; Fang et al., 2013; Zhang et al., 2013; Huang et al., 2014). The design of the sampler (e.g., the sampler orientation,

the shape of the sampler, variation in turbulence, low surface resistances, passivation, etc.) leads to the dry deposition velocity related uncertainty which is about ±50 % for GOM (Lyman et al., 2009; Lai et al., 2011; Huang et al., 2012a). Calculating based on the method described by Eq. (2), the overall uncertainty of GOM dry deposition observation is ±70 %. There is not enough information to quantify the overall uncertainty of PBM dry deposition observation in a similar way. Based on the distribution of daily samples in the study of Fang et al. (2012b), the overall uncertainty of PBM dry deposition measurements is assumed to be roughly ±100 % or within a factor of 2."

Comment:

Enclosure methods: Choi and Holsen 2008/2009 articles are also important, and the authors did not discuss about the bio-process/photo-process related to Hg reduction in DFC.

Response:

We have added the discussion of the influence of DFC material based on the study of Choi and Holsen (2009). Please refer to Lines 474–476 in the revised manuscript:

"Choi and Holsen (2009) reported that the polycarbonate DFC blocks most of the UV-B light from reaching the soil where Hg^{2+} can be reduced to Hg^{0}, and hence the GEM emission flux might be underestimated by at most 20 %."

Comment:

Micrometeorological methods: This method has been used to understand GOM flux as well, but no discussion here.

Response:

We have added discussion on micrometeorological methods for GOM dry deposition measurement. Please refer to Lines 372–375 in the revised manuscript:

"The micrometeorological methods and the enclosure methods were also adopted in some studies (Poissant et al., 2004; Zhang et al., 2005; Skov et al., 2006), but not widely used due to the high uncertainties in the measurements of GOM and PBM

concentrations using the Tekran system."

Comment:

In forests: Choi and Holsen 2009, and there are more articles from Driscoll's group discussing Hg cycle in forests.

Response:

We have cited the study of Choi and Holsen (2009). We have also cited articles from Driscoll's group:

Blackwell, B. D., and Driscoll, C. T.: Using foliar and forest floor mercury concentrations to assess spatial patterns of mercury deposition, Environ. Pollut., 202, 126–134, 10.1016/j.envpol.2015.02.036, 2015a.

Blackwell, B. D., and Driscoll, C. T.: Deposition of mercury in forests along a montane elevation gradient, Environ. Sci. Technol., 49, 5363–5370, 10.1021/es505928w, 2015b.

Bushey, J. T., Nallana, A. G., Montesdeoca, M. R., and Driscoll, C. T.: Mercury dynamics of a northern hardwood canopy, Atmos. Environ., 42, 6905–6914, 10.1016/j.atmosenv.2008.05.043, 2008.

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.

Luo, Y., Duan, L., Driscoll, C. T., Xu, G. Y., Shao, M. S., Taylor, M., Wang, S. X., and Hao, J. M.: Foliage/atmosphere exchange of mercury in a subtropical coniferous forest in south China, J. Geophys. Res. Biogeo., 121, 2006–2016, 10.1002/2016jg003388, 2016.

Comment:

GOM resistance: page 10 line 299-310, Gustin et al., 2015 has summarized this, this is not a new idea. I just feel, the authors are writing a review article, but they are repeating the concepts from the summaries in other's review articles without adding their new

thoughts.

Response:

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:

Page 13 line 401-402, is ambient concentrations not important?

Response:

We have deleted this sentence.

Comment:

Page 14, line 412-414, Europe has…., any ambient data to support this argument?

Response:

We have deleted this argument.

Comment:

Line 427, deposition fluxes concentrations, what does “fluxes concentrations” mean?

Response:

We have modified this statement. Please refer to Lines 182–183 in the revised manuscript:

“Most studies on GOM dry deposition were conducted in North America and Europe”

Comment:

Line 435-439, the authors should explain why they are showing significantly different? Different surface affinity?

Response:

We have discussed the surrogate surface method in detail. Please refer to Lines 382–414 in the revised manuscript:

“Different surrogate surfaces were used to measure different RM forms. Mounts with

cation-exchange membranes (CEMs) are widely used for GOM dry deposition measurements (Lyman et al., 2007; Lyman et al., 2009; Castro et al., 2012; Huang et al., 2012a; Peterson et al., 2012; Sather et al., 2013). The down-facing aerodynamic mount with CEM is considered to be the most reliable deployment for GOM dry deposition measurements so far (Lyman et al., 2009; Huang et al., 2014). Knife-edge surrogate surface (KSS) samplers with quartz fiber filter (QFFs) and dry deposition plates (DDPs) were deployed for PBM dry deposition measurements (Lai et al., 2011; Fang et al., 2012b; Fang et al., 2013). However, these samplers are not well verified to reflect the deposition velocity of PBM, and hence not widely accepted. KCl-coated QFFs were used to measure the total RM (GOM+PBM) dry deposition, but failed to capture GOM efficiently (Lyman et al., 2009; Lai et al., 2011).

According to Eq. (4), the uncertainty of RM dry deposition comes from the uncertainties of RM concentration and dry deposition velocity. The uncertainty of RM concentration mainly originates from the interference of unwanted RM forms or incomplete capture of targeted RM forms. CEMs exhibited a GOM capture rate of 51–107 % in an active sampling system (Huang and Gustin, 2015b). The CEM mounts designed to measure only GOM dry deposition capture part of fine PBM (Lyman et al., 2009; Huang et al., 2014), while the KSS samplers with QFFs designed to measure only PBM dry deposition may also collect part of GOM (Rutter and Schauer, 2007; Gustin et al., 2015). Based on the RM concentration measurements and the surrogate surface method evaluations, the GOM concentration related uncertainty is estimated to be ±50 % (Lyman et al., 2009; Lyman et al., 2010; Gustin et al., 2012; Fang et al., 2013; Zhang et al., 2013; Huang et al., 2014). The design of the sampler (e.g., the sampler orientation, the shape of the sampler, variation in turbulence, low surface resistances, passivation, etc.) leads to the dry deposition velocity related uncertainty which is about ±50 % for GOM (Lyman et al., 2009; Lai et al., 2011; Huang et al., 2012a). Calculating based on the method described by Eq. (2), the overall uncertainty of GOM dry deposition observation is ±70 %. There is not enough information to quantify the overall uncertainty of PBM dry deposition observation in a similar way. Based on the distribution of daily samples in the study of Fang et al. (2012b), the overall uncertainty

of PBM dry deposition measurements is assumed to be roughly ±100 % or within a factor of 2."

Comment:

Page 17, line 537-540, different surface (e.g. forest vs grassland), there are many differences between these two surface types, such as leaf area index, but the authors just simply summarized all these difference depositions based on chemistry and not talking about the characteristic of surfaces.

Response:

High GOM concentration at high elevation leads to high GOM deposition. Leaf area index (LOI) also has impact on GOM dry deposition, but not as much. We have added the uncertainty analysis in the simulation of GOM dry deposition with resistance model. Please refer to Section 4.2.2 in the revised manuscript.