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Review of “Particulate-Phase Mercury Emissions during Biomass Burning and Impact on Resulting Deposition: a Modelling Assesment” by Francisco De Simone et al.

General Comments:

De Simone et al. present a detailed assessment of the impact of mercury emissions from biomass burning on resulting atmospheric mercury deposition. The assessment is completed through utilization of an updated emissions database and an updated global mercury chemical transport model. Within this framework, the authors investigate a variety of model parameterizations and the role of other uncertainties on resulting magnitudes and spatial distributions of mercury deposition. Overall, this work represents a sizable effort and further informs the scientific community regarding speciation of mercury emissions, chemical oxidation mechanisms, and spatial and temporal variations, all within the specific context of biomass burning and with relevance to the implementation of mercury policy.

This manuscript represents a substantial contribution to the field, and is very much with the scope of ACP. However, there are several items that could be addressed by the authors and incorporated into revisions that would likely strengthen the manuscript overall.

We thanks the referee for his positive general comments and for his feedback that helped us to improve the general quality of the manuscript.

Specific Comments:

1. Much of the manuscript focuses on the potential impact of Hg P emissions. However, the issue of particle size is not discussed in the paper. Certainly, there have been assumptions made within the model regarding particle size, with direct implications to the potential transport distance prior to Hg P removal from the atmosphere (via either dry or wet processes). The manuscript would benefit from added discussion specific to particle size.

As discussed in the paper, although there is of experimental evidence that some significant fraction of Hg emitted from BB is bound to particulate, until now in models, Hg emissions from BB were considered only as Hg(0). This is the first modeling study that considers a fraction of Hg from BB to be bound to particulate. Within this scope, the main objective of the paper is to investigate how such speciation impacts the fate of Hg. We consider this study a substantial advance in Hg modeling scientific literature.

Undoubtedly, the particle size distribution will have an impact on the final fate of Hg(p) emitted. However there are large uncertainties regarding the size distribution of particles emitted, and how it evolves during the different phases of BB (see Janhäll and Pöschl, 2010 and the reference therein) . Moreover in this first study we prefer to focus mostly on the mechanism related to the emission speciation and on the uncertainty related to some of the processes Hg(p) undergoes in the atmosphere, such as temperature dependent gas-particle partitioning.

In the revised paper we included the following text:

No further \ce{Hg^{p}} particle dimension distributions other than the standard log-normal particle size distribution, as described in detail in \citep{Jung2009}, were considered in this study due to large uncertainties regarding the dynamic size range of particle emitted during BB, see \citet{Janhall2010} and the references therein.

2. Table 5 presents summary statistics regarding comparison of model output with available observations from measurement networks. However, there is little text in the body of the manuscript in support of the inclusion of Table 5. The manuscript would benefit from added discussion to characterize and specify how well the model performed in comparison to observations.

In the revised text we have included a new subsection within section 3 dedicated to the comparison with Hg measurements from the GMOS network for 2013, to validate the model, and to assess any feedbacks/constraints related to the different assumptions considered about the Hg^p emissions from BB. More particularly, when considering the Hg emissions from all other sources, the very small perturbation produced by moving a fraction of Hg BB emissions from Hg^0 to Hg^p in almost all sensitivity runs causes very little perturbation to the TGM and wet deposition results. Conversely the Hg^p in air concentration samples collected in a number of sites from GMOS networks for the year 2013 enabled us to assess the impact of Hg^p emissions from BB and to distinguish between the different assumptions. In particularly at two remote sites the model runs including a fraction of Hg(p) from fires resulted in a better agreement with measurements.

We included the new Section 3.4 "Constraints from Global Measurements networks" see page 7 of the revised paper.

3. No explanation or justification is provided on the selection of 2013 as the model time period. This rationale should be provided in the revisions, along with some indication of the representativeness of 2013 compared to other recent years.

The authors have already investigated many uncertainties related to Hg emissions from BB in De Simone et al., 2015, including the year-to-year Hg BB emission variability for a decade. As explained above, this study focusses on the speciation of Hg emissions from BB, and on the effects on the resulting deposition, and it is investigated for the first time in a CTM. Results for other years could be somewhat different . However we decided to choose 2013 because it was one of the years best covered by measurements within the GMOS project. This allows us to have feedbacks from the comparison with measurements collected at a global scale.

We modified the text to include the rationale for the choice:

This study cover a single year, the 2013, which has been chosen due the large availability of measurements from GMOS network \citep{Sprovieri2016_conc, Sprovieri2016_wet,Damore2015}.

These results apply for the investigated year (2013) and could be to some extent different considering other years, due to the complex interaction of the numerous actors determining the final fate of \ce{Hg}. However few alternatives of analysis period exist due the limited time coverage of global measurement network(s).

4. Figures 1, 2, 5, & 9 seem to be very instructive. However, they are not easily legible. The size/resolution of these figures should be improved for the benefit of the reader.

We thanks the referee for this useful feedback. We will enlarge the figure at the maximum allowed resolution.

Technical/Editorial Comments:

The units reported in Table 3 need additional clarification (Mg and %).

Page 1 - line 13, “71% to 62%”. . .of total deposition? Seems this sentence is missing

some needed context.

Page 2 - line 7, should be “fraction of Hg emitted” (add “of”).

Page 2 – line 24, period needed after “(Randerson et al., 2012)”.

Page 3 – line 8, “equal to the 15%” (remove “the”).

Page 4 – line 2, “Hg emissions is of great importance” (add “of”).

Page 5 – line 18, “first model level level leads to” (remove “level”).

Page 5 – line 19, “approx” should be “approximately”.

Page 7 – line 1, instead of “have no influence”, perhaps “have little influence”?

Page 8 – line 14, “between the the measurement” (remove “the”).

In the revised paper we have fixed the editorial issues identified by the referee.

Janhäll, S., Andreae, M. O., and Pöschl, U.: Biomass burning aerosol emissions from vegetation fires: particle number and mass emission factors and size distributions, Atmos. Chem. Phys., 10, 1427-1439, doi:10.5194/acp-10-1427-2010, 2010.