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Interactive comment

Interactive comment on "Particulate-Phase Mercury Emissions during Biomass Burning and Impact on Resulting Deposition: a Modelling Assessment" by Francesco De Simone et al.

Anonymous Referee #1

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Review of "Particulate-Phase Mercury Emissions during Biomass Burning and Impact on Resulting Deposition: a Modelling Assessment" by Francisco de Simone.

De Simone and co-authors have explored the sensitivity of an atmospheric mercury model (ECHMERIT) to assumptions about mercury emissions from biomass burning. The main focus of their sensitivity tests is the fraction of mercury that is emitted as Hg(p) vs. Hg(0), although they also test model sensitivity to emission time resolution and oxidants for Hg(0). They use several different plausible Hg(p) fractions (0 to 30%) and various way to apportion that fraction (constant or proportional to biomass burning CO, PM, or OC). The partitioning of emissions is an important issue, as the authors explain, because Hg(0) has a long atmospheric residence time and circulates globally

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while Hg(p) has a short residence time and deposits near the emission source. These are reasonable sensitivity tests and I expect that other mercury scientists and modelers will be interested in the results.

The main weakness of the paper is that it provides no comparison to observations, except for an unexplained and unused table in the Appendix. The paper therefore provides no insight into which, if any, of the many model configurations provide reasonable comparisons with observations. There are abundant surface and aircraft measurements of Hg(0), Hg(p), CO, OC, and PM that could be used for this purpose (GMOS, AMNET, CARIBIC, ARCTAS, INTEX-B). If the ECHMERIT model is run in a climate mode, so that it does not match the daily weather conditions at measurement sites, the simulated distributions and correlations between multiple species can still be compared with observations. Without comparison between model and observations, I do not think that this paper in its current form is suitable for publication in ACP.

Another significant problem with the current version of the manuscript is that the methods do not contain enough detail to understand how the emissions were constructed. How are biomass burning emissions of Hg (=Hg(p)+Hg(0)) calculated from the biomass burning CO or DM provided by GFED? Please provide the relevant emission factors or emission ratios. In simulations where Hg(p) fraction depends on OC, PM, or FMC the manuscript needs to clearly explain how the Hg(p) fraction is calculated from OC, PM, or FMC. Do the emission factors (e.g. CO/DM, OC/DM) vary geographically with biome type? A simulation with 100% Hg(p) from biomass burning is discussed in Sect. 3.3 but not described in the methods. Regardless of how Hg is calculated in the emission inventory, please report the Hg/CO ratio because this would enable comparison to many observations that are reported this way.

Other issues Is there chemical reduction in the model? If so, is Hg(p) affected by it?

The total Hg emissions from biomass burning in this work are 400 Mg/yr. A previous analysis by the same authors reported much higher mean emissions of 675 Mg/yr

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(Pirrone et al., 2010). What is the reason for such a large change?

None of the figures show the spatial map of Hg(p)/Hg ratio (or Hg(p)/Hg(0) ratio), which is the central focus of the paper. In addition, all 4 panels of Figure 2 are visually indistinguishable (and indistinguishable from Fig 1, except for magnitude). I think this space would be better used to show the Hg(p)/Hg emission ratios under the various schemes based on CO, PM, and OC.

Some additional observational studies of Hg in biomass burning plumes should be discussed: Ebinghaus et al., 2007; Holmes et al., 2010.

Page 1 Line 3 (P1L3): Add that the Hg which is not Hg(p) is assumed to be Hg(0).

P1L13: 71% to 62% of what?

P1L15: Statement about mercury in water-stressed and warming forests is speculation that is not supported in the paper.

P1L19: Statement exaggerates the magnitude of biomass burning emissions relative to other anthropogenic emissions; it is certainly less than 1/2 of anthropogenic Hg emissions. First, it is widely acknowledged that a very large portion of biomass burning is anthropogenic, even though emission inventories are not labelled this way. Second, the Muntean et al., 2014 paper does not include mercury emissions from small-scale gold mining, so anthropogenic emissions are much larger than they estimated.

P3L2: particle emissions are presumably also calculated.

P4L2: "of" great importance

P4L9: Define FMC

P4L23: Is the total Hg emission the same in all simulations? How is it calculated from the GFED DM or CO?

P4L23. "Considering" should begin a new sentence.

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P5L14: How are the data in the figures normalized?

P5L16: It seems very unlikely that the fairly smooth zonal-mean distribution would be altered by finer spatial resolution.

P6L23: What does "passive tracer" mean? In atmospheric modeling, "passive" usually means that a tracer does not alter the model's transport or physics. (i.e. it is passively transported.) I would therefore expect that all of the Hg species in all of the simulations are passive in this sense.

P6L29: Not quite correct. Oxidant choice still has a big effect on the deposition pattern.

P7L1: Statement says that vertical profile of emissions doesn't matter, but I expect that the vertical profile would be quite important for scenarios with high Hg(p) emission fraction. Like other aerosols and reactive gases, Hg(p) emitted into the free troposphere should disperse much farther than Hg(p) emitted into the boundary layer.

P7L6: 66% of what?

Fig 7c: Panel title says "Hg(p) fraction =30%" but one of the plotted quantities is "100% Hg(p)". Only one can be correct.

Table 2: How are the correlations calculated? Are they the spatial correlation of the annual mean? Is temporal variability considered in the correlations? What does "Ensemble" mean here?

Table 3: Title should say "from biomass burning"

Table 4: Title says "Mercury deposition (Mg)" but only some rows have units of Mg.

Table 5: Terms "BASE Full" and "Br Full" are not defined.

References Holmes, C. D., Jacob, D. J., Corbitt, E. S., Mao, J., Yang, X., Talbot, R. and Slemr, F.: Global atmospheric model for mercury including oxidation by bromine atoms, Atmos. Chem. Phys., 10(24), 12037–12057, doi:10.5194/acp-10-12037-2010,

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2010.

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Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R. B., Friedli, H. R., Leaner, J., Mason, R., Mukherjee, A. B., Stracher, G. B., Streets, D. G. and Telmer, K.: Global mercury emissions to the atmosphere from anthropogenic and natural sources, Atmos. Chem. Phys., 10(13), 5951–5964, doi:10.5194/acp-10-5951-2010, 2010.

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