

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

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Review of "Particulate-Phase Mercury Emissions during Biomass Burning and Impact on Resulting Deposition: a Modelling Assessment", by De Simone et al., Atmos. Chem. Phys. Discuss.

GENERAL COMMENTS

De Simone et al. present a model-based study of mercury emissions and deposition arising from global biomass burning (BB), examining a range of different model inputs and assumptions, with particular emphasis on the fraction of mercury emitted from BB as particulate mercury [HgP]. Overall, this seems to be an excellent investiga-

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tion, although as noted below, there are some areas that might need some additional explanation and/or justification.

SPECIFIC COMMENTS

1. The model year 2013 was selected. How does 2013 compare with other years in terms of BB emissions? Should note that conclusions from this work apply to 2013 and will likely be at least somewhat different for different years.

2. (P2.L27). Would be helpful if you could say something about the differences in the inventories. E.g., a few sentences at least regarding the essential differences in how they were constructed, and of course, what the different emissions were in each inventory.

3. Section 2.2 Experimental Setup. Would be helpful if you included here (or elsewhere) additional details about the model. Here are some details, for example, that might be helpful:

* Is HgP created from Hg(0) oxidation in the model, and if so, what fraction of the oxidation products are assumed to be HgP with different reactions, etc.?

* Once HgP is emitted into the model (and/or created within the model), can it be transformed to any other form of mercury, i.e., can HgP be converted to Hg0 or Hg2 in the model?

* Is there any conversion or partitioning of Hg2 to HgP in the model? If so, how is this estimated, and is it reversible?

* What particle size(s) are assumed for HgP? What information exists on the particle size distribution of HgP in the BB plumes? This would seem to be a very important factor, considering particulate deposition is critically influenced by particle size. This could be noted as a relatively uncertain aspect of the simulation that is not being addressed in the present study.

* How is particulate dry deposition handled in the model? Is gravitational settling velocity factored in? If so, what are the size(s), shape factor(s), and density(s) of HgP-carrying particles?

* How is particulate wet deposition handled in the model? In my modeling work, I have found the parameterisations used in HgP wet deposition to have a very big impact on the fate/transport of HgP.

* Has the model been evaluated by comparison against HgP measurements? If so, what were the results?

4. The model is being run with a relatively coarse grid (e.g., on the order of 2.8×2.8 degrees at the equator), and so, as with any model of this type, sub-grid phenomena could be adding uncertainty to the results. Especially, for example, for emissions from BB, the height of emissions could significantly impact the near-field deposition. In real-world BB situations, the emissions will not be uniformly distributed throughout the PBL, and deposition from the real vertical distribution could be much different than that with the assumed uniform-PBL assumption. In some cases, the near-field deposition could be much greater, to the extent that the emissions are emitted nearer to the ground. Along these same lines, the authors do carry out a simulation with emissions confined to the first layer of the PBL. While the height of this layer does not appear to be specified in the paper, I'm not sure it should be considered such an unrealistic simulation, as is done in the analysis. The fact that it seems to give relatively different results could be seen as evidence that emission height really does make a difference. While I am not that familiar with the literature, I believe there have been numerous studies published regarding the height of BB emissions under different conditions. As a related point, the manuscript notes that "In particular high HgP fractions were observed during smouldering phases, whereas very low or undetectable HgP levels were found during flaming combustion." [P4.L18-19]. This could mean that the highest HgP emissions might occur with relatively low injection heights, i.e., if the injection heights under smouldering conditions are lower than the heights under more intense combustion conditions.

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5. Figures 6 and 7 are a really interesting way to present the results! However, it took a little time to get my head around what they were saying at first. Perhaps a little more explanation could be added in the caption for these figures?

Technical corrections and/or suggestions

(...Note that in the following, if a wording change or other correction is being suggested, I have simply included the final wording being suggested, rather than any sort of "track changes" notation. Apologies if this leads to any lack of clarity.)

P1.L22. "Its relative importance may increase in the coming years, e.g., if the Minimata Convention and/or other efforts lead to reductions in anthropogenic emissions."

P2.L16-17. "...resulting from BB, when variations in HgP fractions and production processes are considered."

P2.L17-19. "The most recent version of the GFED BB emission inventory (van der Werf et al., 2010; Randerson et al., 2012; Mu et al., 2011), has been included in the global online Hg chemical transport model ECHMERIT, to simulate Hg deposition from BB for the year 2013 and to quantify the influences of variations in model inputs, assumptions and parameterisations."

P2.L23. "... version of the inventory..."

P2.L24. Need period at end of sentence.

P2.L27. Wouldn't these be considered "sensitivity" runs, rather than "control" runs?

P2.L27. "... see Andela et al. (2013) (and references therein) for a description..."

P3.L4. "Unless explicitly stated,..."

P3.L9. "This value is within the range of observations (Obrist et al., 2007; Finley et al., 2009). However, since there are uncertainties in Hg speciation from BB (Zhang et al., 2013), further simulations were carried out with varying fractions of HgP (0%, 4% and

30%)."

P3.L15-17. "The principal vertical profile used (PBL-Profile) maps Hg emissions uniformly within the Planetary Boundary Layer (PBL), whereas in the second, the vertical profile of the standard version of the ECHAM-HAM model was used (HAM-Profile)(Zhang et al., 2012)."

P3.L16. Could the "HAM" acronym be defined the first time it's used?

P3.L21-23. "These simulations primarily employ a O₃/OH Hg₀(g) oxidation mechanism. However, since the precise atmospheric Hg oxidation mechanism remains unclear (Hynes et al., 2009; Subir et al., 2011, 2012; Gustin and Jaffe, 2010; Gustin et al., 2015), a number of runs were performed using a Br-based oxidation mechanism."

P3.L28-29. "Finally two simulations were conducted including Hg emissions from all sources and including re-emissions, to evaluate model performance against measurements (see Appendix A)."

P3.L28-29. What additional emissions were used for these "all-source" simulations?

P3.L32. "The majority of Hg releases from BB is believed to occur as Hg₀(g)."

P4.L7. "properties" is misspelled.

P4.L13-16. What equation(s) were used, with what parameters? That is, you say that the Hg₀ to HgP ratio is determined by FMC, but what is the mathematical relationship used?

P4.L27. I cannot really see very many "notable differences" in Figures 1 and 2. Part of the issue is that the figures are very small and the color ramp does not have a lot of contrast. Could the figures be bigger?

P5.L3. At a number of points in the document, it is stated that only the 85:15 emissions speciation results are shown "for clarity". Its not clear to me why showing the results for other speciation profiles would make things less clear. There would be more figures,

but would clarity really suffer?

P5.L14. How were the latitudinal deposition profiles normalized?

P5.L18. What is the height of the first model level?

P5.L20-22. A few comments about the following sentence: "This last vertical distribution scenarios are unrealistic, however the differences obtained here contrast with the findings of De Simone et al. (2015) and are due to the fraction of HgP included in this study."

(a) Not exactly sure what you are trying to say here in terms of comparison to findings of De Simone et al. (2015).

(b) As noted above in the Specific Comments, I'm not sure I agree that the vertical distribution being referenced is unrealistic.

(c) This sentence needs to be reworded somewhat for grammar and clarity.

P5.L28. Do you mean the "deposition peak"?

P6.L1. Maybe would be clearer if the section was called something like this: "Impact of atmospheric oxidation pathway and speciation profiles on geographic distribution of deposition".

P6.L10-12. This sentence is a little confusing, particularly with the use of "all" towards the end. This "all" confused me before I realized you didn't really mean "all".

"To better understand the combined effect of Hg speciation and oxidation pathway on the deposition distribution, agreement maps were created, to highlight the model cells where different simulations all predict significant deposition..."

Maybe better to say something like this:

"To better understand the combined effect of Hg speciation and oxidation pathway on the deposition distribution, agreement maps were created, to highlight the similarities

and differences in the distribution of high-deposition model cells in different simulations..."

P6.L12. What statistical distribution is the "standard deviation" calculated for, e.g., is it the combined data set of cell-by-cell deposition for all cells in all relevant simulations?

P6.L14. "Using the O₃/OH mechanism, the number of model cells in which the model predicts high deposition..."

P6.L21. maybe "contrasts" (or simply "presents") rather than "confronts"

P6.L22. Not sure what you mean by "passive tracer" in this context. It still deposits, right? In other simulations, how are HgP emissions not like a "passive tracer" in the same context? I guess you are implying here that there is no chemical reactions in which Hg⁰ is oxidized to HgP, and/or that there are no processes converting HgP to another form of Hg. And so, there should be no impact of the oxidation mechanism chosen. But, as noted above in the specific comments above, you could add some additional detail to the text regarding these and other issues to make things clearer.

P6.L26. Seems like maybe this section could be divided into two. One called "Uncertainty" and one called "Biomass Burning versus Anthropogenic Impact"

P7.L1. Could refer the reader to the figure or table that shows the point you are making. Also, instead of "actually have no influence", could say something like "have little influence". And as noted above, you haven't convinced me that the emissions into the first model level – or at least emissions into something less than the full PBL – are really "unrealistic".

P7.L7. I don't see the Antarctic in the tabular results, but you give results here?

P7.L12. "... as in De Simone et al. (2015)." (and same correction a few lines later)

P7.L13-16. What is an "inspected ensemble"? How was the eventual ensemble created – medians of values for each cell, or mean values for each cell, or some other

method?

P7.L24. "just about everywhere" (seems like there are a few locations less than 25%?)

Table 1. Model Evaluation (not Model Validation)

Table 2. Would be helpful to explain the "R" and "P-KS" parameters a little either in the Table or in the text. At least to me, it seems a little too cryptic.

Table 3 and Table 4. Maybe could make these into some sort of graphic, either instead of or in addition to?

Table 5. What measurement sites? How many sites? What networks? What averaging time for "r" and for "NMRSE%"? Need some more detail here. What about comparison against HgP measurements? This would seem to be important for this paper!

Figure 3, and in fact, most figures: Why so small? For Figure 3, could make it much wider and I think would be much clearer. Difficult to see data when lines overlap so much. Maybe consider some sort of differential dotted/dashed line(s) so that they might be able to be distinguished even when congruent?

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