

Thank you for your thoughtful comments. Including your suggested revisions has improved the quality of the manuscript. Our responses are indicated below in blue text.

This is a great paper that represents great progress in the field of atmospheric chemistry. It definitely deserves publication. I have a few specific comments:

1. On page 2 lines 14-15, you talk about how oxidation by O₃ and OH is unlikely. It is important, I think, that you bring out and discuss (or debunk, if they merit it) other oxidation/reduction mechanisms that have been proposed, including heterogeneous mechanisms. No one disputes that halogen-involving reactions are important. But there are some strong voices that claim other mechanisms may also be important, and I think it would be useful to give them consideration.

We add a sentence (Page 2, lines 19-20):

“OH and ozone could still potentially be important oxidants on aerosols (Ariya et al., 2015).”

We chose to leave the more detailed discussion of other proposed mechanisms in Section 2 and Tables 1 and 2 instead of in the introduction.

2. On page 2 lines 30-32, you suggest that in-plume reduction isn't important, and give the Deeds and Landis papers as references. Both papers show evidence for in-plume reduction, and the Landis paper talks about how the process is likely widespread. Maybe I am misinterpreting your text or theirs, but I'm not sure the statement you make here is supportable by the evidence you have given.

We revised this sentence for clarity and added text (page 2 lines 33 to 37, page 3 lines 1) as follows:

“Fast in-plume reduction of Hg^{II} emitted by coal-fired power plants was first reported by Edgerton et al. (2006) and Lohman et al. (2006) but more recent field observations suggest that on average only 5% (range 0-55%) of emitted Hg^{II} is reduced in the plume (Deeds et al., 2013; Landis et al., 2014). Recent aircraft observations (Weigelt et al., 2016) and emission inventories (Zhang et al., 2016) suggest that previous reports of in-plume reduction of Hg^{II} may reflect in part an overestimate of Hg^{II} emissions.”

3. I am curious about why in Figure 1 the model shows quite high annual mean Hg^{II} at the surface near the south pole and overall shows much higher Hg^{II} in the southern troposphere than in the northern. I think this is a surprising result, and I don't think it was discussed in the manuscript. I would be glad to see some explicit discussion of the reason for the north-south difference in Hg^{II} concentrations. Unfortunately, we have very few (if any) reliable Hg^{II} measurements with which to validate this result.

We add the following discussion to Section 4.1 (page 8 lines 18-20).

“Hg^{II} reduction in the model is faster in the northern hemisphere than in the southern hemisphere because of higher OA concentrations. Hence Hg^{II} concentrations are higher in the southern than in the northern hemisphere (Figure 1).”

4. Page 7 lines 8-10: again, some discussion of how inclusion of heterogeneous oxidation chemistry might play a role could be useful here.

We add the following sentences (now page 7, lines 34-37):

“We did not consider heterogeneous Hg^0 oxidation on particle surfaces (e.g., Vidic et al., 1998; Flora et al., 1998; Lee et al., 2004) due to inadequate information to formulate atmospheric rates. Further study of heterogeneous Hg^0 oxidation is needed (Ariya et al., 2015).”

5. Page 10 lines 1-5: It is not at all clear to me that these modifications to GEOS-Chem “solve” the problem of underprediction of wet deposition in the Gulf of Mexico. Based on Figure 6, the model is still strongly underestimating Gulf of Mexico wet deposition. I don’t think the model even gets the basic spatial trends right, based on my visual inspection of Figure 6. It may be fair to say that it is doing better than before, but to say it has it all nailed down seems to be an overstatement.

We rewrite this paragraph (now page page 10, lines 34-36 to page 11, lines 1-2) to address your and other reviewers’ comments:

“Previous GEOS-Chem simulations with Br-initiated oxidation of Hg^0 failed to capture this maximum because Hg^{II} production favored higher latitudes (Holmes et al., 2010; Amos et al., 2012). The inclusion of NO_2 and HO_2 as second-stage oxidants in our simulation shifts Hg^{II} production to lower latitudes and matches the general location of the Gulf of Mexico maximum although the magnitude is still underestimated. Increasing horizontal resolution in the model could improve the definition of the maximum (Zhang et al., 2012).”

6. Page 11 lines 30-31: I don’t agree that the “wet deposition fluxes in the model are consistent with observations.” This assertion needs to be toned down.

We revised this and the following sentences as follows (page 12, lines 32-36):

“Hg wet deposition fluxes in the model are consistent with spatial patterns observed in North America and Europe. Inclusion of NO_2 and HO_2 as second-stage HgBr oxidants in the model shifts Hg^{II} production to lower latitudes compared to previous versions of GEOS-Chem and enables the model to capture the location of the observed maximum in wet deposition along the Gulf Coast of the US. However, the magnitude of this Gulf Coast maximum is still underestimated.”

We also modified the Abstract and Section 5 text, which are consistent with the above.