

We would like to thank the referee for their thoughtful and useful comments. We have provided our responses to the referee's comments below (in blue).

This study describes the development of a higher resolution nested version of the well known GEOS-Chem model, and describes the results obtained simulating the wet deposition of Hg over continental North America, and compares these results with the extensive observations made in the Mercury Deposition Network. The article also discusses the speciation of emissions from major anthropogenic sources, and assesses the quality of the simulation results, representing in plume reduction by using different speciation profiles for certain types of anthropogenic emission source. Overall the paper is clearly written and well presented. The subject is relevant, in 2013 UNEP hopes to complete negotiations for a legally binding instrument on mercury before the twenty-seventh regular session of the Governing Council / Global Ministerial Environment Forum in 2013, see, <http://www.unep.org/hazardoussubstances/Mercury/Negotiations/tabid/3320/Default.aspx>, which is something the authors may wish to add to their Introduction.

We have added on sentence on the UNEP negotiations at the end of the first paragraph of the introduction.

The approach used by the authors is sound; using the same model (physics parametrisations, and chemistry scheme) to perform both the global and regional simulations makes more sense than taking boundary and initial conditions from a global model, as input for a regional model which calculates atmospheric chemistry and physics differently. The results certainly seem to give a greater insight into the processes influencing the spatial and temporal distribution of the wet deposition of Hg over North America. There is no doubt that the article deserves publication, although there are a few general and a couple of specific points which the authors should address by the authors before final publication in ACP.

Generally speaking throughout the article there seems to me to be a tendency to overestimate the amount of confidence that can be placed in the results obtained from model simulations, and to present model results as if they were fact, some of the instances where a caveat might be useful are noted in the following comments.

We have modified various parts of the text to make it clear that our statements are based on model results. We have taken care not to overstate our confidence in model results.

In the abstract the authors use the term high-resolution. In terms of regional modelling, would a half by two-thirds degree resolution be classified as high, although certainly it is higher than the global model obviously. Higher resolution, maybe?

We have removed references to high resolution. Throughout the text, we have applied reviewer#3's suggestion of nested model and global model.

Also in the abstract the last three sentences read as facts rather than findings from model simulations. The last sentence regarding dry deposition should certainly be qualified as there are very few dry deposition measurements with which to compare the simulation results, and the methods used for the measurements are still under development.

The last sentences of the abstract were modified accordingly.

In the introduction the authors could add the following references: regarding emissions; one or both of AMAP/UNEP (2008); Pirrone et al. (2009), and concerning the influence of boundary

conditions Pongprueksa et al. (2008), which showed very clearly the correlation between boundary conditions and deposition fluxes in regional models.

We have added references to Pirrone et al. (2009) and Pongprueksa et al. (2008).

In the description of the model (p2607 onwards) the authors give an annual total of 220 Mg yr⁻¹ for the contribution of Hg emissions from biomass burning, this is noticeably less than the 300 Mg yr⁻¹ in Holmes et al. (2010), which the authors cite, and significantly less than the 675 ± 240 Mg yr⁻¹ in Friedli et al. (2009) which is not cited. Is this because the authors prefer to use an enrichment factor $\Delta[\text{Hg}]/\Delta[\text{CO}]$ at least in part derived from measurements over North America?

We used the same $\Delta[\text{Hg}]/\Delta[\text{CO}] = 100$ nmol/mol as Holmes et al. (2010), based on measurements obtained during the ARCTAS campaign over California and Nevada, as well as measurements obtained over the Pacific Northwest and over Canada. Our value of 220 Mg yr⁻¹ is lower than the 300 Mg yr⁻¹ of Holmes et al. (2010) because of the different model years in these two studies (2008-2009 in this study vs 2006-2008 in Holmes' study) and the CO emission from Global Fire Emission Database (GFED) varies from year to year. This is indeed lower than the Friedli et al. (2009) estimate. However, this source is small compared to anthropogenic emissions and other major natural sources such as ocean and soil re-emissions (Holmes et al., 2010).

On page 2609, the parametrisation process to describe polar AMDEs is described, however there are no references, and the figure of 5 ppt is given as the concentration of BrO in the boundary layer when a series of conditions is met. What period do the authors consider as springtime, how much sea-ice per model cell is necessary, is there a cut off value for the incoming solar radiation at which there is no BrO, how are stable conditions defined, and why below 268 K? The reader is left with rather a lot of questions and the authors should expand a little, or provide a reference for the source of the parametrisation.

The reference (Holmes et al., 2010) with more detail about the parameterization is now added.

In the description of the IPR simulation, p2611, the authors describe the studies that show that the oxidised Hg content of coal fired power plant plumes is significantly less downwind of the plant than it is when measured in close proximity to the stack, which suggests that there is an in plume reduction of HgII. If I recall correctly it was proposed that SO₂ is involved in the reduction mechanism. The authors adjust the proportions of Hg species in the emission inventory for CFPPs. The authors then apply similar changes to the emissions from incinerators (is there evidence of high concentrations of SO₂ from incinerator stacks?), and use 96% Hg(0), 0% HgII, and 4% HgP; this speciation profile is cited as from Streets et al. (2009). Streets et al. (2009) actually cites Streets et al. (2005), which for Hg speciation from waste and residue burning in the table 7 of their article cites three articles by Friedli et al. namely Friedli et al. (2001, 2003a,b), which refer to biomass burning and agricultural waste burning. Therefore I am not convinced that the application of the in plume reduction parametrisation used for CFPPs is entirely valid. Given the underestimation of Hg wet deposition in Florida at certain times of the year, it may well be a good idea to rerun the model removing this assumption.

In the revised manuscript, we now use the same in plume reduction assumption in incinerators as in power plants power plants: 75% conversion of Hg₀ to HgII. We note that this is speculative as to our knowledge the downwind Hg speciation of incinerator plumes

has not been sampled. Our assumption is based on the fact waste incinerator plumes can contain high levels of SO₂ (Psomopoulos et al., 2009; Stevenson 2002). The resulting IPR speciation in waste incinerators is 80.5% Hg(0) and 19.5% Hg(II). Assuming that IPR only occurs in CFPP and not in waste incinerator plumes leads to a partial correction of the STD model overestimate. This has now been noted in the text.

References:

Psomopoulos, C. S., Bourka, A. and Themelis, N. J.: Waste-to-energy: A review of the status and benefits in USA, *Waste Management*, 29, 1718-1724, 2009.

Stevenson, W., 2002. Emissions from Large MWC Units at MACT Compliance. Memorandum to Docket A-90-45, US EPA, Research Triangle Park, NC.

The total emissions from natural and anthropogenic sources are described on page 2612 and illustrated in Figure 1. I am surprised that there are no (or extremely low) emissions from the sea all along the West Coast of the United States, and also that the emissions from parts of the North Atlantic appear to be as high or higher than they are from the most part of the continental US (middle row, left, figure 1).

The GEOS-Chem Hg simulation contains a bi-directional exchange of Hg⁰ between air and sea (Strode et al., 2007; Soerensen et al., 2010). The ocean emission source flux is determined by the gradient of Hg⁰ concentrations across the interface. According to the model, there is a net uptake of Hg⁰ along the west coast of the United States because the high productivity leads to rapid sinking of Hg on marine particles and thus net transfer of Hg⁰ from the atmosphere to the ocean. The high emissions over the Atlantic Ocean (30-40N latitude) are caused by relatively higher intermediate oceanic Hg concentrations as well as the strong entrainment of sea water to ocean mixed layer along Gulf Stream (Soerensen et al., 2010).

It also seems from the two figures in the middle row that the area of intense emissions in the vicinity of the Great Lakes in the global model is not present in the regional model. Could the authors comment on this please?

The global and nested models use the same emission inventories with different resolutions. The intense emissions in the global model near the Great Lakes is due to sum of several large point sources. These point sources are resolved as discrete "points" in the nested model (strictly speaking, they are still boxes in the nested-model grid, but the size is only 1/60 of the coarse inventory) because of the higher resolution.

Also on page 2612, the sensitivity study with primary anthropogenic emissions turned off, this refers only to Hg emissions I assume?

Yes, this is correct. We have clarified this point in the text.

On page 2616, last line. Is 'very well' perhaps a slight overstatement?

This statement has been modified in the revised manuscript.

On page 2617, the last sentences of section 3.2. I wonder if these underestimations would improve if the authors remove their assumption about the speciation profile from waste incineration and rerun the model. Or perhaps they tried and it led to poor results in other areas.

Would it be possible to include a little more discussion here, perhaps mentioning what happens in the waste incineration in-plume reduction assumption is removed?

We have tried removing the modification to the source profile of waste incineration. We found that this does not help the wet deposition over the SE very much, but causes large model bias over the NE. This is because Hg emissions from waste incinerators occur mainly over the ORV (51%), followed by the SE (11.8%), MW (6.2%), NE (5.8%) and other regions (the remaining 25.2%). This has been noted in the revised text.

Page 2617, Section 3.3. In the discussion of the simulated mean surface concentrations of atmospheric Hg there are a number of comments in the text which seem unusual. Temme et al. (2003) and Lindberg et al. (2007) both give the hemispheric background of Hg⁰ as 1.5 - 1.7 ngm⁻³, (northern), and 1.1 - 1.3 ngm⁻³ (southern). Therefore values of 1.7 - 1.8 ngm⁻³ for TGM in the Ohio River Valley cannot be considered 'high', nor can 1.5 - 1.6 ngm⁻³ in Nevada and Utah be considered 'elevated' and 1.3 - 1.5 ngm⁻³ over the Great Plains, southern Canada and northern Mexico is not 'typical' background. The concentrations in the MBL seem anomalous as well, mean values of the measurements in the Atlantic (northern hemisphere) are higher than 1.2 - 1.4 ngm⁻³ (see Sprovieri et al. (2009)), and the authors ascribe the low MBL values to low emissions (fig 1, middle panels) which as mentioned before hardly seems to be the case looking closely at the figure. These concentrations seem low and require explanation.

We have revised the manuscript following the reviewer's suggestions. The low concentrations in the Atlantic MBL are due to the short lifetime of Hg (rapid oxidation of Hg(0) to Hg(II) followed by scavenging/deposition onto sea salt aerosols).

On page 2618, the authors state that at higher altitudes, HgII concentrations are high due to faster oxidation and slower removal processes. This is another instance where modelling studies suggest that this is indeed the case, however there are not enough experimental results of certain enough quality to state this definitively. The authors should qualify this statement.

This statement has been changed in the revised manuscript.

On pages 2619-20 the authors discuss the seasonal cycle of TGM and refer to the summertime increase in oxidation and subsequent deposition, citing a number of previous studies. However the studies by Bergan and Rodhe (2001) and by Selin et al. (2007) used a different chemical mechanism based on oxidation by OH (and O₃ in the case of Bergan and Rodhe (2001)), therefore the authors should explain why they think this comparison is valid, or point out the fact that whatever the atmospheric oxidation pathway for Hg⁰ is, observations indicate that it is more rapid in summer and any attempt at modelling the atmospheric Hg cycle needs to be able to reproduce this phenomena.

The revised manuscript refers to these previous modeling studies and states Hg(0) oxidation in all these studies maximized during summer because this corresponds to the peak in oxidant (whether it is Br, OH, or O₃).

p2622 section 3.6. Do the authors mean all anthropogenic Hg emissions?

Yes, the "anthropogenic emissions" in the first sentence of section 3.6 is revised as "anthropogenic Hg emissions".

On p2624 the authors mention that the model resolution cannot capture the local scale enhancements in Hg deposition which occur close to point sources. Is it possible that the IPR

simulations therefore underestimate the contribution of North American sources to north American deposition? And therefore that the 12% estimated in the last line of the Conclusions may not be as robust as it seems?

As the MDN and AMNet stations used in our study are not directly downwind of large point sources, they should represent regional levels of Hg(II) concentrations and wet deposition which can be captured by our global and nested models. We agree that at local scales the model predictions likely underestimate the domestic anthropogenic emission contributions. This has been noted in the text.

References

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