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 manuscript describes a new nested modeling approach to the simulation of atmospheric mercury where a single model is applied first in a low-resolution global mode to provide boundary values for a secondary limited-area high-resolution nested simulation. Having the same chemistry formulations for mercury in both simulations should minimize some of the modeling artifacts encountered in previous works.

However, differences in the resolution of the vertical axis could still lead to artifacts especially near the lateral boundaries of the limited-area model. It is not clear from the text whether the 47-layer structure of the global model was also used in the nested model.

The vertical grid is the same in the global and nested models. This has been clarified in the revised manuscript.

I would also caution that "high-resolution" is a relative term. There certainly have been simulations of atmospheric mercury performed at finer horizontal resolution than the modeling applied here. The authors might consider identifying their model domains as "parent" and "nested".

We have eliminated references to "high-resolution" and use "nested model" instead or refer to higher horizontal resolution in the nested versus global model.

The authors are definitely correct when they state "Considerable uncertainty remains on the Hg0 oxidation mechanisms and their kinetics." In this modeling work, bromine atoms are the sole oxidant for Hg0. This brings me to question whether it is appropriate to completely discount all other oxidation reactions and whether the emissions, transport, chemistry and deposition processes for bromine are accurately modeled. I see limited supporting evidence to answer these questions.

We agree with the reviewer large uncertainties remain on the questions of whether bromine atoms are the sole oxidants for Hg and on how well we understand the budget of bromine in the troposphere. It is possible that Hg(0) oxidation by OH and O3 do play a role, however the work of Calvert and Lindberg (2005) and Hynes et al. (2009) suggests that these reactions are too slow to dominate in the atmosphere. We have added a statement describing the consistency of the assumed BrO columns with satellite observations.

Despite many uncertainties, this new modeling effort has managed to show fairly good agreement with measurements of wet deposition and air concentration of mercury. The improved accuracy achieved with the "in-plume reduction" assumption is especially noteworthy and begs for the identification of the chemical kinetics involved. Unfortunately, our limited understanding of the fundamental processes of atmospheric mercury necessitates the use of such speculative assumptions in simulation modeling. This work makes use of some important assumptions which improve the modeling results and is worthy of publication. However, I do have some questions that require explanation and some suggestions for improvement of the manuscript as detailed below.

Page 2609, last paragraph in section 2.1: I'm a bit confused by "the HgII tracer is assumed to be partitioned between gaseous and particulate phases". HgII in the particulate phase would be HgP would it not? Later in section 3.3 you bring PBM (particulate-bound mercury) into the discussion.

It needs to be made clear throughout the text what mercury species the model actually resolves and how these relate to the observed species. I personally deplore the term “reactive gaseous mercury” or “RGM” as it is an operational term coined back in the days when mist-chambers were used to detect an unexplained fraction of soluble and presumably reactive gaseous mercury. It is unfortunate that HgII is still a necessary generalization in atmospheric mercury modeling. I was surprised at the success of the HgII gas/particle partitioning in Amos et al. (2012) without knowledge of the mercury compounds present. But we should not fault modelers for trying to move forward in the face of these uncertainties as long as they are adequately explained.

In the revised manuscript, we have updated our model simulations to follow the approach described by Amos et al. (2012), thereby clarifying assumptions about different Hg species. We now transport two species, elemental mercury, Hg(0), and divalent mercury, Hg(II). Hg(II) is assumed to be in equilibrium between gas and particulate phase at all times based on local temperature and PM2.5 concentrations, following Amos et al. (2012). Our model results are only affected in a minor way and our conclusions remain unchanged. In terms of comparison to observations, modeled Hg(0) concentrations are compared to GEM, modeled gas phase Hg(II) is compared to RGM, and modeled particulate phase Hg(II) is compared to observed PBM.

I am glad to see that Hg in this model is not being scavenged by the Bergeron process of vapor condensation to cloud ice. But what becomes of the now concentrated mercury left in the remaining cloud water? What if that cloud water evaporates entirely? Does GEOS-Chem maintain and transport cloud water separately from cloud ice?

We trace the Hg concentration in cloud water, ice and air by mass conservation. The Bergeron process removes cloud water but does not influence the concentration of Hg(II) in cloud water, which is calculated based on the assumption of instantaneous equilibrium of Hg(II) between gaseous and liquid phase. The rainout and washout processes are treated as scavenging processes, i.e. the model subtracts the mass of Hg being scavenged from a grid box and adds this mass onto the grid box below (or to the ground if it’s the bottom level). If the cloud water evaporates, Hg(II) is released back to the gas phase.

Page 2611, lines 24-26: What is the rational for applying the in-plume reduction of mercury for incinerators? The phenomenon in regard to CFPPs is poorly explained on the basis of chemistry. Expanding its application to incinerators seems unfounded unless the modeling showed it was necessary to achieve the best agreement with observations.

Please see the response to anonymous reviewer #2.

Page 2612, lines 6-11: Since CFPPs are such an important source of atmospheric mercury it would seem advantageous to apply some sort of temporal resolution during the simulation period. Previous modeling efforts have been able to temporally allocate pollutant emissions from power generation sources by time of day and by season based on well documented power demands. The constant-rate assumption applied here could lead to unrealistic vertical mixing and horizontal transport. Was this a necessary simplification? If so, this should be explained.

We do not include any temporal variation in anthropogenic Hg emissions. While this information is not available in the global inventory, it is available in the NEI inventory (on daily, weekly, and monthly scales). This is a shortcoming in our approach that is now noted in the text.

Page 2613, lines 24-26: Is the higher wet deposition flux over land really attributable to higher updraft velocity? Is it not actually due to the precipitation arising from upward vertical motion on either side of the land/water discontinuity, whether over land in the daytime or over water at night? The presumption of higher wet deposition over land might be based on a lack of measurement over water.

The contrast in wet deposition flux between land and ocean in the Gulf Coast and Florida coast regions are due to a combination of differences in precipitation and emissions that are resolved in the nested model but not in the global model. This statement only applies to the impact of resolution in the model. Our original statement was unclear, and was modified in the revised manuscript.

Page 2614, lines 13-15: The correction for snow bias is not well explained. Do you correct all weekly wet deposition amounts during the entire year by the annual (rain+snow)/snow ratio? This would unduly increase the MDN-observed values in the warm seasons and not fully account for the low collection efficiency of the MDN samplers in the cold season.

We correct the weekly wet deposition amounts based on the fraction of snow over total precipitation over the corresponding month, i.e. no correction is applied if there is no snow. This is now clarified in the text.

Figure 4 as discussed in section 3.2: Why did you not show the full 2-year time line? It would be interesting to see how the simulations performed for each month of the 24 month period in addition to each month in 2008 and 2009 averaged together. Viewing Figure 4 with the mistaken impression that it shows model performance over a single annual cycle could give a false impression of model accuracy based on averaging. The same holds true for the many line graphs in Figure 8 where it would be informative to see month-by-month comparisons of model to observation rather than multi-year averages of observations compared to 2008-2009 averages of model results.

We have updated Figure 4 to show both 2008 and 2009. Due to lack of sufficient observations at CAMNet and AMNet sites we were not able to do the same for Figure 8 (please see our response below).

Page 2617, lines 22-24: The IPR results for TGM in the marine boundary layer show rather low concentrations compared to most of the observations I recall seeing. See Table 11.1 in Sprovieri et al (2009) which shows considerably higher concentrations. I’m not convinced the cycling of mercury between air and water in the MBL is simulated well.

The GEOS-Chem model seems to underestimate observed TGM concentrations over both North Atlantic and Pacific oceanic basins, especially during winter and spring as discussed in previous studies (Holmes et al., 2010; Soerensen et al., 2010). This is now noted in the revised manuscript.

Page 2618, lines 7-10: Vertical mixing seems to be a reasonable explanation for the higher RGM and PBM in the nested model. However, some documentation of the stronger ventilation of the PBL in the nested model would strengthen the argument.

Wang et al. (2004) have evaluated the different extent of vertical ventilation of models with resolutions from 4°×5°, 2°×2.5° to 1°×1° with great detail. This reference is added in the revised manuscript.

Figure 8 as discussed in section 3.4: The size of the graphs needs to be increased so that the axis labels are legible. The STD simulation results seem to be missing from the CAMNet graphs. As mentioned before, I think it would be preferable to see the full two-year timeline of monthly modeled and observed values for each site where possible. I understand the CAMNet sites did not operate during the 2008-2009 period of the model simulation. I have doubts about comparing model simulations of one time period to observations taken during an entirely different period.

We have increased the size of the labels for this Figure. Results from the STD and IPR simulations are nearly identical at the CAMNet sites and fall on top of each other on the Figure. We have tried to compare the model values with AMNet observations on a monthly basis over the two model year 2008 – 2009, but we found there are only 3 sites having the full 24-months data. The selected CAMNet sites have observations only before 2007. So instead of showing individual years, we calculate the multiple-year mean of monthly Hg concentrations for AMNet/CAMNet observations.

Figure 9 as discussed in section 3.5: The lower ARCTAS TGM concentrations above 7000 meters with their presumed exclusion of some fraction of HgII suggest that HgII concentrations above the tropopause may be significant. The IPR modeling results show very little vertical structure to both the HgII and Hg0 concentrations, which suggests an incomplete treatment of atmospheric mercury processes near and above the tropopause.

We note in the revised manuscript the discrepancy between model and observations above 7 km altitude.

Page 2622, lines 10-13: How does having more of the anthropogenic Hg emissions in elemental form in the IPR simulation cause more efficient wet deposition of background and natural Hg emissions? Is there a certain "holding capacity" of the atmosphere for elemental Hg such that adding more from anthropogenic sources caused more efficient oxidation and deposition? This needs further explanation.

The IPR simulation does not affect natural emissions but increases the Hg(0)/Hg(II) ratio for anthropogenic emissions. For anthropogenic emissions outside of N. America, this leads to reduced local deposition of Hg as Hg(II) and increased export in the form of Hg(0). This added anthropogenic Hg(0) is mixed on hemispheric scales and incorporated into the background pool. Once oxidized to Hg(II), it can thus lead to an increase in the wet deposition flux of background Hg over North America. This has been clarified in the revised manuscript.