Responses to comments of Reviewer 1

We wish to express our great appreciation to Reviewer 1 for the very careful review, instructive comments, and recognition of the value of our work. We have revised the manuscript following these comments. Your comments and suggestions significantly improve the quality of this manuscript. In the response below, we address each of these comments. The Reviewer's comments are italicized and our responses immediately follow.

Major comments

*Overall, the paper lacks a discussion of *why* the model performs well in some cases and poorly in others. The reader wonders what these modeling successes and failures tell us about mercury processes or fate?*

Revised: Following the reviewer's comments, we added discussion on possible reasons in corresponding to the good and bad performance of the model in the summary and conclusion part.

Emissions should be better documented, given that this is a new model and source estimation is a key issue in mercury research, particularly for natural surfaces. I recommend including figures showing the global distribution of emissions from volcanoes, soils and vegetation, and oceans, each separately …

Revised: I totally agree with reviewer on that emissions is a key issue in mercury research, especially for modeling. Following reviewer's comments, we provide figures and short introduction to mercury emissions of CAM-Chem model as supplement materials to this paper.

As this is a new model which includes online calculations of oxidants and reductants of Hg. The key species affecting Hg would be O3, OH, sulfite, HO2 and halogens. These reactant concentrations need to be documented, but currently only Br concentrations are, by referring to work by Parrella et al. If information for other reactants is already available in the literature, please cite it and give some summary information, such as surface O3 concentration biases compared to a measurement network, a measure of global OH, like methane lifetime, and sulfur deposition biases compared to a measurement network. If these comparisons are not already published for a closely related version of CAM-Chem, then the paper needs to include this information and probably some figures. I realize this may be significant work, but the quality of the Hg simulation hinges on the quality of the oxidant and reductant concentrations.

Revised: Following the reviewer's comments here, we have found best possible collections of references to evaluations of other chemical compounds from CAM-Chem modeling community. It covers the evaluations/validations of CAM-Chem model in simulating ozone, OH, sulfate etc. I hope this revision meet this comment.

Lei et al. summarize the current understanding of mercury kinetics on pages 9851-9852 (hereafter, I use only the last 2 digits of the page numbers), focusing on the conflicting reports of

*Hg(0) reactivity with OH and O3. While this is a useful and important discussion, I disagree with some aspects of their description. Most importantly, Calvert and Lindberg (2005) and Hynes et al. (2008) argue that *neither* OH nor O3 are likely important in the atmosphere, based on the reaction enthalpies and instability of intermediate compounds. While subsequent work by Rutter et al. (2012) may suggest some role for O3, albeit via some unknown reaction mechanism, I know of no subsequent studies that have rehabilitated an important role for OH and Lei et al. provide none. Thus, I view OH as an implausible oxidant for Hg(0), regardless of whether O3 is. The discussion should reflect this.*
Revised: Following reviewer's comments, we have revised the text to prevent overstatement and present the uncertainty in the role of OH. Thanks for this point.

While the observations by Rutter et al. (2012) may indeed support some role for O3 in oxidizing Hg(0), I don't think they should be described as "more accurate" than earlier work, unless Lei et al. provide reasons why they think the earlier experiments were deficient. Furthermore, I don't think Rutter's laboratory studies "directly refute" the concerns of Calvert and Lindberg (2005) that environmental reaction rates may differ substantially from laboratory rates due to the high concentrations of radicals and surface area in all laboratory experiments.
Revised: Following reviewer's comments here, we revise the text. Rutter's experiment is done in a more practical environment indicating aerosols may play a role of catalyst in O3-Hg oxidation. Calvert and Lindberg (2005)'s point is more from the theoretical aspect disregarding the influence from environment.

The comparison of vertical profiles with ACE-Asia data is problematic. First, the observed values shown in Fig. 4 are very different from those in Friedli et al. (2004; Fig 4), although they should be the same. For example, Friedli et al. reported 1.3 ng/m3 at 7.5 km versus 0.6 ng/m3 in this work. Second, the ACE-Asia profile appears anomalous when compared to the extensive profiles available from NASA flights (Talbot et al., 2007; 2008), CARIBIC (Slemr et al., 2009), and Banic et al. (2003). I suggest comparing the model to these other NASA, CARIBIC and Banic profiles.
Revised: First, the figure 4 contains some error in pasting to word from excel. Second, as several other reviewers pointing out, the vertical comparison is a problem. Similar to this comment, some other reviewers (Dr. Feng and Reviewer 2) also point out that ACE-Asia data may not good to use in comparison. Therefore, we redo the vertical comparison by using aircraft observations from NOAA air resource lab and NASA flights, which make consistent measurements in land and prevent the possible disturbance.

The manuscript focuses extensively on comparing CAM-Chem/Hg against an early version of the GEOS-Chem model (Selin et al. 2007). It is not clear to me why comparisons are not made to several other global and hemispheric Hg models (GRAHM, ECHMERIT, CTM-Hg, MSCE-Hg). …
Revised: Following reviewer's comments here, we add the discussion in reference to results from other model. We are not intending to disregards other models. Just in developing this model, we are comparing with from a 2010 version of Geos-Chem model.

Minor revision:
p51/l21 - Avoid "/" as its meaning is unclear. "and" would probably suffice here. p52/l6 - The reverse would make more sense (i.e. mercury concentrations respond to ozone) due to the far greater ozone concentrations.
p54/l10 - Please be more specific about what CAM-Chem model version this work is based on, e.g. version number. The paragraph cites Lamarque et al. (2011) who described CAM4-Chem, but the first sentence says this study is based on CCSM3, which I believe used CAM3.
p56/l10 - Sentence unclear.
Revised: Thanks for careful review. Above minor issues have been revised.

p56/l19 - "uptake by the marine boundary layer" is unclear. Please specify what deposition processes are treated in the MBL.
p57/l4 - How was 20% reemission chosen? If it was tuned, please specify what observation it was designed to reproduce.
p57/l5 - "rapid reemission of mercury... does not need to be compensated for by dry deposition." Meaning unclear
Revised: We use air-sea exchange to calculate the uptake. The reference to re-emission treatment have been added. The sentence has been fixed, which was inappropriately modified from a comparison with CMAQ in previous draft.

p58/l2 - Something is unclear. As written, it sounds like F1 is the annual-mean emission flux from soils in GEOS-Chem (global total or for each grid box?). Since GEOS-Chem already includes temperature and solar radiation in its calculation of F1, including them also in the equation for F2 will shift the spatial distribution of emissions.
p58/l20 - Is Cw a global mean or specified separately for each ocean grid box?
Revised: F1 is the grid emission dataset. That is the static base emission. F2 is dynamic and should shift from F1 depending on practical temperature, and solar radiation. Cw is specified separately for each ocean grid box based on [Soerensen et al., 2010].

Sect 2.5 This section mainly compares to past modeling results. Please also discuss budgets that are derived mainly from observations. e.g. Pirrone et al. (2010 ACP), Mason (2008 UNEP Hg report)
Revised: They are very important sources, especially the UNEP report which has been carefully read and cited in our assessment report to EPA. We add discussion on comparison with other results.

p59/l17 - What is the "rapid reemission" of deposited mercury over the ocean and how is it different from "emission from the mixed layer"? Sect 2.3 said that "rapid reemission" occurs only over land and snow, in lieu of a detailed model for emissions from vegetation and snow. Since emissions from the mixed layer are already included (sect. 2.4), an additional rapid reemission appears to double count ocean emissions.
Revised: The sentence is unclear. What we mean here is that the emissions are from both old mercury (long term ocean storage) and new mercury (newly deposited).

p60 - I agree with the comment by F. Slemr that the Pacyna et al. (2005) inventory likely overestimates Hg emissions from South Africa by a large amount. For further information, see Masekoameng et al. (2010), Leaner et al. (2009) and references in Slemr's comment.
Revised: It is really important comments. We have revised text to include this point. We were suspecting the emission in South Africa would be a problem, but did not find confident reference to point out. Thanks.

p60 - A major feature of Fig 2 is the high TGM concentrations predicted by the model over upwelling regions along the west coasts of S. America and Africa. These high concentrations are not seen in the ocean or atmospheric concentrations simulated by Soerensen et al. (2010), which is the basis for the ocean emissions in this work. Please comment on the discrepancy and why emissions over upwelling regions are much larger in this work.
Revised: You made very careful review. We noticed this issue before. The ocean emission scheme use the emissions from Soerensen et al., as basis. The calculated (dynamic) practical emissions are highly depending on the meteorology factors (surface wind, temperature, etc.). We noticed that the scheme is sensitive to temperature. The emissions of Hg in upwelling regions usually have more contribution from deep ocean storage. In addition, some regions (e.g. eastern pacific) often has positive temperature anomaly, which can increase the Hg emissions. The sensitivity can be considered as the uncertainty of ocean emission scheme. I have added text to point out.

p61/l4 - The paper by Lamborg et al. (2002) does not discuss the inter hemispheric concentration ratio, as far as I can tell. A different paper by Lamborg and others does contain this information.
p61/l5 - The text says the inter hemispheric ratio is 1.68, but that seems incorrect based on Fig 3. I estimate the TGM concentrations in the SH to be 1.5 ng/m3 and 2.0 in the NH. That gives a ratio of 1.3, which
Revised: I corrected the reference list. For the ratio, you are right. It is 1.37 based on the latest calculation. Previous value may come from a testing version or use an alternative emission sources.

p61/l13 - Please explain how these comparison sites were chosen. Why are other measurement sites (e.g. other CamNet sites) not included? The text says the sites are non-urban, but several are urban, including Seoul, Tokyo, Beijing, Chongqing and Detroit.
Revised: We try to select several sites on each region, so that the comparison can evenly cover the global area. We did not get much confirmed data for comparison during the simulation period. By your suggestions, we added some sites from CamNet observations. These sites you picked up are really from urban. We modify the text to continue using these data, since we could not find better data to get a global coverage.

Sect. 3.2 This would be an excellent place to remind the reader that the kinetic coefficients for Hg(0) reactions with OH, O3 and Br are uncertain. If the coefficients are much slower than assumed in this modeling work, then Br chemistry may have a larger impact on TGM and Hg cycling.
Revised: You are right. We include your comments.

p66/l4 - This paragraph discusses the relationship between the TGM distribution and wet deposition. This is not helpful since TGM is mostly Hg(0) and Hg(0) is hardly susceptible to wet scavenging. It would be more appropriate to show and discuss the relationship between RGM and PHg concentrations and wet deposition.
Revised: It is revised.
p66/l9 - There are more than 4 sites with continuous records of 1999-2001 wet deposition. Why select these 4?
p66 - It would be helpful to discuss why the model correlations are much better in some regions than others. What can we learn from these correlations?
Revised: We select 4 sites based on availability of NADP data and spatial distribution for a better spatial coverage. We modified to mention this in the text.
My understanding to the result is that model tends to better capture the variability on sites in regions with less anthropogenic impacts. Model emission is also the main problem for bias.

p67 - How long are the sensitivity simulations?
Revised: The same period 1998-2001 as control runs.
p67/l8 - I am unpersuaded by this paragraph. As I wrote in the pre-ACPD review, Fig.9 incorrectly shows a surface pressure of 1000 hPa over Western N. America. The figure's vertical coordinate is probably incorrect over all regions with elevated surfaces. I believe this plotting artifact causes the apparent dip in TGM concentrations in the red box. The rising concentrations to the east of the red box are probably caused by US emissions. The panels in the right column of Fig. 10 do not show strong deposition of Asian Hg in the region of the red box. Since Fig 9 and this paragraph are not critical to the paper, I recommend cutting both.
Revised: I agree with your comments on figure 9. Since figure 9 is used just to show the existence and pattern of long range transport. Therefore, I use another figure to replace figure 9 to better explain this issue. The new figure is made from sensitivity study of total mercury on Asian mercury transport, which shows the transpacific transport is relatively stronger in middle troposphere than that in low layers. This part of study is required to be added in this paper initially by both my advisor and PI in considering a better interpretation of U.S. mercury. It is better for me to keep it. Thanks!

p68/l13 - This statement seems to contradict itself. Just say that deposition to the Western US from US sources is bigger, but that Asian sources contribute significantly.
*p69/l20 - This sites are definitely *not* distributed evenly. Change this.*
Revised: Thanks for the help.

Table 1 Why do some oxidation reactions produce PHg and RGM, while others produce only RGM? The text says that all oxidation reactions produce both.
Revised: This is a typo error. I have corrected. All the HgO in CAM-chem model are considered the half /half partition. All HgX (X: Cl; Br) are considered totally RGM.