

We thank the reviewer for constructive comments and suggestions. In the following we would like to reply the comments point by point.

1 General Comments

The manuscript describes the addition of a Hg module to a global chemical transport model, with a focus on the impact of emissions from China. The model performs similarly to most of the other models already described in the literature.

The model lacks a bi-directional exchange flux at the land-atmosphere and ocean-atmosphere interfaces, the model does not contain the option to use a Bromine based oxidation mechanism for Hg oxidation, even in the Arctic, and the comparison between model and observations has been performed using mismatched years. The authors themselves point out that this inconsistency is a weakness in their study. The authors have also used an emission database that is known to be flawed, but have not attempted to rectify this. The dry deposition to wet deposition ratio is out of line with other studies apart from one GEOS-Chem study quoted by the authors, Selin et al. (2007), which was revised a year later, Selin et al. (2008), more recent GEOS-Chem simulations also suggest more equal dry and wet deposition fluxes (Amos et al., 2012; Zhang et al., 2012). There does not seem to be any major improvement over previous Hg models in this manuscript, and in fact a number of important processes are less well described than in other models, I do not think there is much reason to publish this article in GMD.

Response: Thanks very much for the comments. From our perspective, the present work is unique or important in the following aspects. Firstly, online nested Hg simulation with flexible horizontal resolution was developed and evaluated in this study. Compared to traditional multi-scale modeling approach (using a global model to provide initial and boundary conditions to a regional model) (Seigneur et al., 2001), online nested method use the same physical and chemical parameterizations in the global and nested domains which could avoid uncertainties induced by different boundary conditions. Compared to offline nested method used in the GEOS-Chem model (Zhang et al., 2012), online nested method can provide boundary conditions

with higher time resolution from the global domain to the nested domain. Typically, the time resolution of boundary condition in offline and online nested model is 3 hour (or 1 hour) and 10 minute (or 5 minute), respectively. Therefore, online nested simulation would potentially improve model performance in regional scale. Secondly, as stated in the introduction, little model validation has been conducted over East Asia (especially China) in previous global modeling studies due to lack of observational data. We have made great efforts to collect various Hg observations from literatures and conducted comprehensive model evaluation over East Asia in this study. Finally, the global impacts of the primary anthropogenic emissions from the world's largest single emitter, China, have been assessed for the first time in this study. We hope the reviewer and the editor to reconsider this paper to get published by GMD.

The responses to the general comments with model methodology used in our model as raised by the reviewer are as follows.

1/ bi-directional exchange flux at the land-atmosphere and ocean-atmosphere interfaces

In the present model, the treatments of Hg reemissions from land and ocean mainly follow the method used by Jung et al. (2009). Besides, global and regional total emission amounts are prescribed according to estimates in previous studies. This can make sure the global and regional total reemissions from land and ocean are generally reasonable. As show in Figure S3 in the supplement, the spatial pattern of the reemissions from land and ocean seem to be similar with previous modeling studies. Certainly, we also agree with the reviewer that lack of a bi-directional exchange flux at the land-atmosphere and ocean-atmosphere interfaces is a weak aspect in the present work. In the future, parameterizations of air-sea and air-land Hg dynamic exchange should be included in the model to better resolve Hg reemissions.

2/ Bromine based oxidation mechanism for Hg oxidation

Large uncertainties in atmospheric chemistry of Hg is one of the fundamental limitations of current models. The primary gaseous oxidants of Hg(0) in current models include O₃, OH, H₂O₂ and reactive halogen species (e.g. Br, Cl, I, Br₂, Cl₂, BrO, ClO, IO, etc.). Lei et al. (2013) demonstrated that adding Br chemistry has little

impact on overall global TGM patterns based on sensitivity experiments using the CAM-Chem Hg model. Wang et al. (2014) also pointed out that Br is less important than O₃ and OH as oxidants for Hg(0) in Hg simulation over China with high oxidation capacity. Besides, several latest Hg modeling studies (Simone et al., 2014; Gencarelli et al., 2014) still used O₃-OH oxidation mechanism alone in their models. It seems that at the current level of understanding the O₃-OH oxidation mechanism is still sufficient for Hg simulation. However, the importance of Br atoms in gas phase reaction of Hg has been identified by several studies (Holmes et al., 2006, 2010), and these reactions are believed to be essential in Polar region and marine boundary layer. We accept the advice of the reviewer and add Br chemistry for gas oxidation of Hg, to provide the option of using Br oxidation mechanism and address the impact of Br chemistry in our model. Detailed results are given in the response to the specific comment (2.2.1).

3/ Mismatch of the time periods between simulation results and observations

Detailed explanation concerning the influence of the mismatch of the time periods between model results and observations is given in the response to the specific comment (2.3.1). Overall, we think the influence of the mismatch of the time periods may be relatively large for dry and wet deposition comparisons in East Asia but relatively small for other comparisons. However, no dataset of Hg observations in East Asia is available at present. So we have no better choice.

4/ Flawed emission database

We used the AMAP 2000 inventory (Pacyna et al., 2006) because it was used by the HTAP multi-model experiment for Hg (Pirrone and Keating, 2010) and also widely used by many other published modeling studies (Selin et al., 2008; Jung et al., 2009; Lei et al., 2013). We are very sorry that we do not know the emissions in South Africa are wrong in the AMAP 2000 inventory when we started the present work in 2010. We have replaced the Hg emissions in South Africa using the AMAP 2010 inventory and rerun the model. This update results in significantly decreasing of local Hg emissions and concentrations in South Africa. More detail analysis is given in the following response (Section 2.2.3).

5/ dry deposition to wet deposition ratio

Dry and wet deposition account for 78 and 22 % of total deposition in the simulation results, respectively. This ratio is different with the results presented by Selin et al. (2008) and Amos et al. (2012). But it is comparable with the results of Selin et al., (2007). Actually, a more latest modeling studies (Lei et al., 2013) also gave a similar ratio. Dry and wet deposition contribute 70% and 30% to total deposition in their results. We think these differences might be caused by different Hg chemistry and deposition parameterizations and also meteorology inputs used in different models.

2 More Specific comments

2.1 Introduction

The Minamata convention has its own website <http://www.mercuryconvention.org/>. Rather than the HTAP report, (Pirrone and Keating, 2010), the most recent Technical Background report to the Global Mercury Assessment might be more appropriate, AMAP/UNEP (2013). The GEOS-Chem reference is out of date there are a number of more up to date publications, with various improvements on Selin et al. (2007). The same is true of CMAQ-Hg, and ECHMERIT (De Simone et al., 2014). The global model used by Environment Canada (GRAHM, see Dastoor and Durnford (2013) and references) is not included in the list, neither is WRF/Chem-Hg (Gencarelli et al., 2014). Zhang et al. (2012) is probably the most recent article looking at local/long-distance sources of Hg to the US, perhaps it should be cited earlier.

Response: Thanks for the suggestions. The references in the introduction will be updated according to the advices of the reviewer.

2.2 Model Description

2.2.1 Mercury Chemistry

All the Hg(II) produced by the reactions between Hg and O₃ and OH is assumed to be in the gas phase, this is not in line with most other models and will have a major

impact on deposition flux fields in many regions. The authors should justify this, or ideally rerun the model splitting the oxidation products between the gas and aerosol phases to have an idea of how important this is. Indeed (Amos et al., 2012) partition the products between gas and particulate phase as a function of temperature and PM2.5. The lack of a Br oxidation mechanism is a serious shortcoming, as it is known that Br oxidises Hg, and therefore is significant not only in the Arctic but also in the MBL, and the difference in the concentration fields and deposition flux fields should using this mechanism should have been evaluated.

Response: In the present model, the oxidation products of Hg(0) by O₃ and OH in gas phase are all treated as gaseous Hg(II) for simplicity. As stated by the reviewer, this scheme is out of date. Especially, this scheme may be unsuitable in places with severe PM pollution (e.g. China, India). We will update the gas particle partitioning scheme based on the work of Amos et al. (2012) and Wang et al. (2014). The impact of gas-particle partitioning of RGM on Hg concentrations and deposition will be addressed in the revised manuscript.

We have added Br chemistry for gas oxidation of Hg and test the impact on Hg concentrations. As shown by Table R1, we add five Br chemical reactions in the gas phase (Seigneur and Lohman, 2008) in addition to the O₃-OH oxidation mechanism to test how the Br oxidation reactions affect the Hg distributions. Similar to the treatment of Holmes et al. (2006, 2010), the five reactions are treated as a single reaction, with an effective Hg(0) first-order rate constant that is a function of the individual reaction rates and the concentrations of Br, BrO and OH based on the assumption that Br, BrO and OH concentrations don't change by their reactions with Hg. This is also the same with the implementation described in CAMx (2014). The effective first-order rate constant is calculated as follows:

$$k_{eff} = \frac{k_1[Br](k_3[Br]+k_4[OH])}{k_2+k_3[Br]+k_4[OH]} + k_5[BrO] \quad s^{-1}$$

Table R1. Bromine reactions added in the model (T is the temperature in degrees Kelvin, and P is the pressure in atmospheres).

NO.	Reaction	Rates
BR1	$\text{Hg}(0)(\text{g})+\text{Br}(\text{g})\rightarrow\text{HgBr}(\text{g})$	$k1 = 3.6 \times 10^{-13} P \left(\frac{T}{298}\right)^{-1.86} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$
BR2	$\text{HgBr}(\text{g})\rightarrow\text{Hg}(0)(\text{g})$	$k2 = 3.9 \times 10^9 \exp\left(\frac{-8537}{T}\right) \text{ s}^{-1}$
BR3	$\text{HgBr}(\text{g})+\text{Br}(\text{g})\rightarrow\text{HgBr}_2(\text{g})$	$k3 = 2.5 \times 10^{-10} \left(\frac{T}{298}\right)^{-0.57} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$
BR4	$\text{HgBr}(\text{g})+\text{OH}(\text{g})\rightarrow\text{HgBrOH}(\text{g})$	$k4 = 2.5 \times 10^{-10} \left(\frac{T}{298}\right)^{-0.57} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$
BR5	$\text{Hg}(0)(\text{g})+\text{BrO}(\text{g})\rightarrow\text{Hg}(\text{II})(\text{g})$	$k5 = 1.0 \times 10^{-15} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$

In the GNAQPMS-Hg model, Br and BrO are not explicitly simulated. Therefore, we specify typical vertical profiles of Br and BrO concentrations over land and ocean, with higher values over ocean (2.9×10^{-8} and 2.9×10^{-7} ppm for Br and BrO) than over land (5.0×10^{-9} and 5.0×10^{-8} ppm for Br and BrO). During the night, the concentrations of Br and BrO are assumed to be zero, considering that the photolysis of Br_2 is the primary source for these radicals.

Figure R1 shows the difference of surface TGM concentrations resulting from introducing Br reactions. Decrease in TGM concentrations is found in the whole globe. This is because additional Br chemistry transforms more $\text{Hg}(0)$ into $\text{Hg}(\text{II})$, which subsequently enhances the deposition of $\text{Hg}(\text{II})$, leading to the reduction of TGM concentrations. Larger TGM reduction is found in the Northern Hemisphere than in the Southern Hemisphere. In general, the change in TGM concentration is less than 0.2 ng m^{-3} in most areas which indicates that introducing Br chemistry seems to have little impact on overall TGM magnitudes and patterns. These results are similar to Lei et al. (2013) which test the impact of Br chemistry using the CAM-Chem-Hg model. Although adding the Br chemistry does not significantly change the TGM pattern, but it may affect the gaseous Hg partitioning between $\text{Hg}(0)$ and $\text{Hg}(\text{II})$, and hence may affect the global Hg deposition patterns. More in-depth tests and analysis are needed to address these impacts in the future.

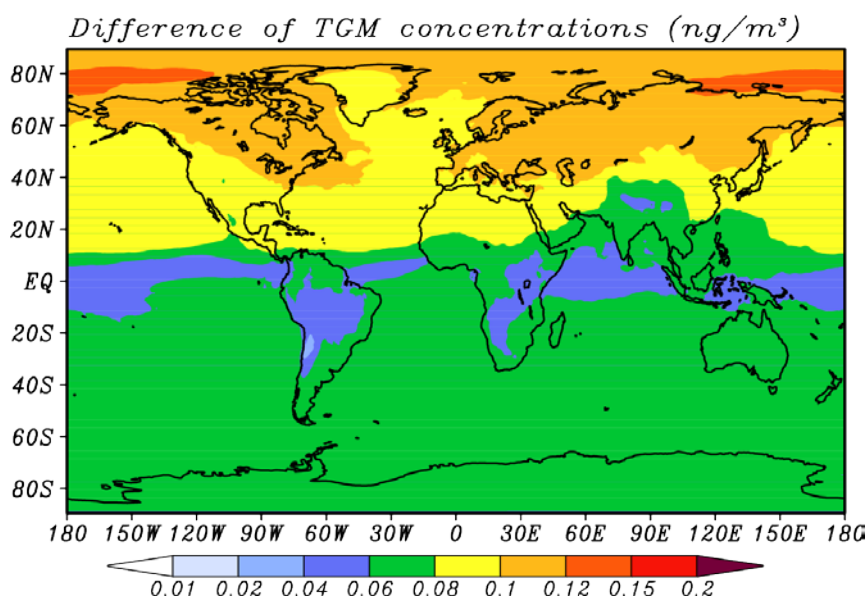


Figure R1 Change in surface TGM concentrations (ng m⁻³) by introducing bromine chemistry (positive value means the TGM concentration decreases after added bromine chemistry)

2.2.2 Mercury Deposition

Pirrone and Keating (2010) is not an appropriate reference for this comment, the authors should cite the individual publications describing the results from the different Hg models.

Response: “Both dry and wet removal pathways are equally significant to the total deposition of mercury.” is one of the main findings stated in Section 4.3.2 (Page 108) in the HTAP report (Pirrone and Keating, 2010). So we think Pirrone and Keating (2010) should be an appropriate reference here.

2.2.3 Mercury Emissions

The authors have ignored the latest anthropogenic emission inventories, AMAP/UNEP (2013); Rafaj et al. (2013); Muntean et al. (2014). The authors increase the Asian emissions in the inventory following Selin et al. (2008), however they do not reduce the South African emissions which are known to be wrong (AMAP/UNEP, 2008), nor do they include artisanal mining, which Selin et al. (2008) did, giving totals for different regions. The authors later state that high TGM concentrations are found downwind of mining areas in South Africa without pointing

out that is where the Hg emission inventory was very wrong. The 2005 emission inventory revised the Hg emissions from gold production down by two orders of magnitude (150 Mg). As the 2000 and 2005 inventories are on the same grid, perhaps it would have been possible to substitute the 2000 data with the 2005 data relatively easily? The use of biogenic CO emissions from oceans and land should be justified. I would have thought that this led to an overestimate of emissions particularly in the Southern Ocean where wind speeds and productivity are high. The expression “Additionally, ocean emissions are adjusted” does not provide enough detailed information to assess the author’s methodology.

Response: We have always pay close attention to the development of anthropogenic Hg emission inventory and we aware that global emission inventories with base years of 2005 and 2010 are available from AMAP (AMAP/UNEP, 2013) and EDGAR (Muntean et al., 2014).

We started the present work in 2010. We used the AMAP 2000 inventory (Pacyna et al., 2006) because it was used by the HTAP multi-model experiment for Hg (Pirrone and Keating, 2010) and also widely used by many other published modeling studies (Selin et al., 2008; Jung et al., 2009; Lei et al., 2013). We are very sorry that we do not know the emissions in South Africa are wrong in the AMAP 2000 inventory. As shown by Figure R2, we have replaced the anthropogenic Hg emissions in South Africa by using the AMAP 2010 inventory. The emission amounts in South Africa decrease by about a factor of 4 (from 259 Mg to 64 Mg). After updating the emissions, the surface Hg concentrations in South Africa decrease by up to 1 ng m^{-3} , but have little changes elsewhere (the difference of concentrations are smaller than 0.01 ng m^{-3} in most areas) as shown by Figure R3. The simulated TGM concentrations at Cape Point decrease from 1.77 ng m^{-3} to 1.23 ng m^{-3} , more close to the observed value.

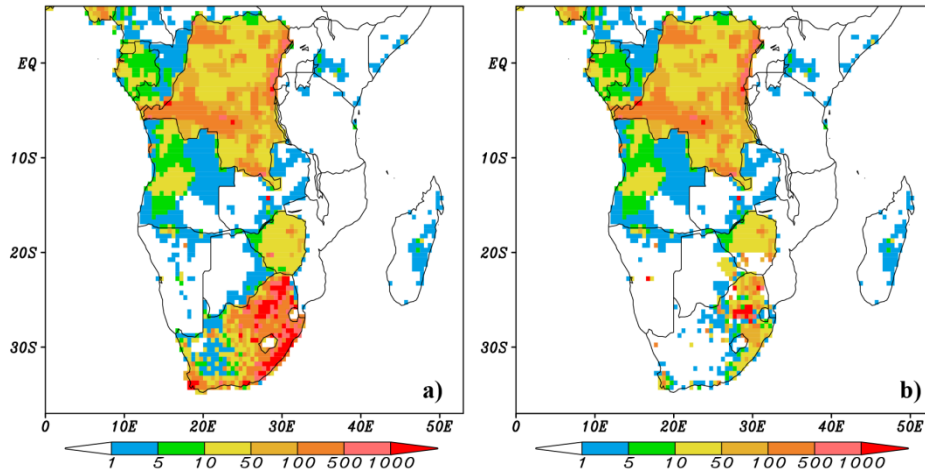


Figure R2 Annual anthropogenic Hg emissions (kg/grid) in South Africa, a) the AMAP 2000 inventory and b) the AMAP 2010 inventory in South Africa (16-34⁰E, 36-20⁰S) + the AMAP 2000 inventory elsewhere

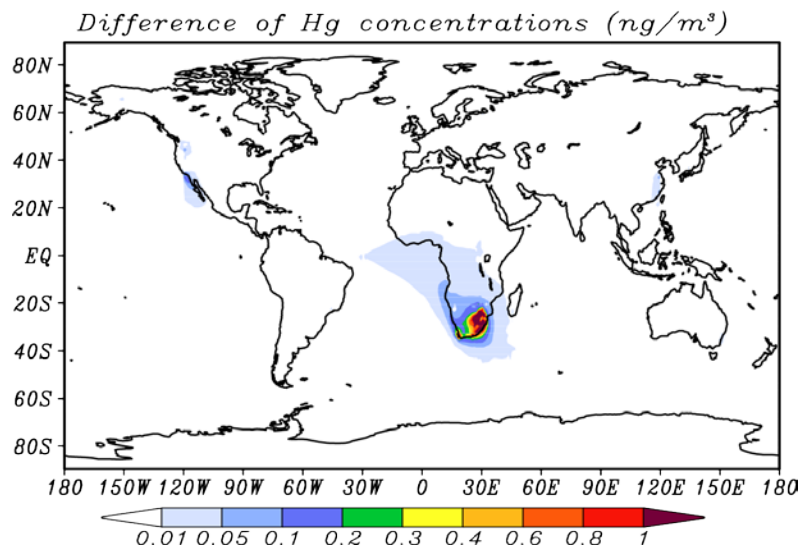


Figure R3 Difference of global surface Hg concentrations after updating the anthropogenic emissions in South Africa by using the AMAP 2010 inventory

As stated in the manuscript, Hg reemissions from land and ocean are not only mapped according to biogenic CO emissions, but also constrained by global and regional total emission amounts prescribed according to estimates in previous studies. This can make sure the global and regional total amounts and patterns of land and ocean reemissions are generally reasonable. As show in Figure S3 in the supplement, the spatial pattern of the reemissions from land and ocean seem to be similar with previous modeling studies (Selin et al., 2008). More detailed information about how to adjust regional ocean emissions to reflect several spatial distribution characteristics

found by previous studies will be given in the revised manuscript.

2.2.4 Model setup

Twenty vertical layers does not seem very many. Why use NO_x emissions from 1983 to 1990? The latest version of MOZART is version 4, why use v2.4?

Response: Yes. To save computation time, we used a medium vertical resolution with 20 layers from the surface to 20 km in the present model. We think this resolution is enough for Hg simulation and it is higher than many other Hg models (Jung et al., 2009).

Online parameterization of lightning NO_x emissions were not included in the present model. Instead, we used the lightning NO_x emissions averaged from 1983 to 1990 from Price et al. (1997).

Yes, the latest version of MOZART is version 4. We just used MOZART output of O₃, NO_x, CO as initial and top boundary conditions in our simulation. This dataset has been also used in many of our other studies (Li et al., 2009; 2011) and shown good model performances. Besides, as shown in Figure S5, the O₃ seasonal cycle in 900, 500, 250 hPa were well reproduced. Therefore, we think the dataset from MOZART-v2.4 is sufficient.

2.3 Model evaluation

2.3.1 Observational data

There is a coordinated global Hg monitoring network <http://gmos.eu/>, see also http://www.geo-tasks.org/geoss_portfolio/health_gmos.php. The temporal mismatch between the observations and the modelling period all but renders any comparison between simulations and measurements invalid. I fail to see how the authors imagine they can publish this.

Response: We known that there is a coordinated global Hg monitoring network (Global Mercury Observation System, GMOS) established in the end of 2010. However, observations from GMOS are still not available publicly at this moment. We hope the observations from GMOS will open to the public in the near future.

We recognize that the mismatch of the time periods when comparing the model with the observations is a weakness of the present work. However, as stated in the manuscript, only Europe and North America have routine monitoring networks for atmospheric Hg concentrations and deposition. Actually, observations of wet deposition and precipitation in Europe and North America are from EMEP and MDN respectively, and the time periods are exactly the same with simulation results. In contrast, no public Hg observation datasets are available in East Asia. So we have no choice but to use observations (collected from literatures) with mismatch time periods in East Asia.

All observations of Hg concentrations at land sites used in this study are averaged over time periods larger than 1 year. Analyses of long-term measurements show that trends in mean TGM during the last decade are small (of order $1\%a^{-1}$) or negligible at most background sites in the Northern Hemisphere (Temme et al., 2007; Wangberg et al., 2007). Therefore, the influences of the mismatch of time periods between model results and Hg concentration observations would not be large. Similar observational datasets (as shown in Table S1-S3 in the supplements) are also used by previous modeling studies (Selin et al., 2007, 2008; Holmes et al., 2010).

Observations from ship cruises are just used for initial comparison of simulated results over ocean following previous studies (Selin et al., 2007, 2008; Holmes et al., 2010). These observations are not used for quantitative model evaluation and not including in the calculation of statistical parameters of model performance.

Annual dry and wet deposition measurements in East Asia (Table S4 in the supplement) are all obtained from literatures. Considering that dry and wet deposition fluxes are affected by environmental factors (e.g. precipitation) and they might differ from one year to another, so the influence of the mismatch time periods would be relatively larger. Again, no observations of Hg deposition are available at present. So we have no better choice.

Overall, we think the influence of the mismatch of the time periods between model results and observations is relatively large for dry and wet deposition comparisons in East Asia but relatively small for other comparisons. Therefore, we don't think the

model comparison results presented in this study are invalid.

2.3.2 Global mercury budget

The total atmospheric burden of Hg is very, very high, compare Mason et al. (2012), so high in fact that it is almost certainly wrong. Quoting a recent study using GEOS-Chem, Horowitz et al. (2014), "Our simulated present-day atmospheric reservoir of 5800Mg is slightly higher than the observational range (4600 - 5600 Mg), but this could be accommodated by uncertainty in Hg re-emission from soils."

The ratio between dry and wet deposition seems to me to be improbable. Are the authors sure they don't have a problem in their wet deposition scheme, are convective and synoptic precipitation included? Have they checked the WRF output with precipitation observations? Of course it could be the dry deposition that it over-estimated but this dry/wet ratio is quite different from most modelling studies and needs to be investigated. The article quoted as corroboration of the dry/wet ratio, Selin et al. (2007), was improved upon Selin et al. (2008) and the dry to wet deposition ratio revised.

Response: The total atmospheric burden of Hg is higher compared to the studies of Mason et al. (2012) and Horowitz et al. (2014). However, there are also other studies giving comparable total atmospheric burden of Hg with our results, e.g. 10400 Mg in Shia et al. (1999), 6900 Mg in Seigneur et al. (2001), 7690 Mg in Seigneur et al. (2004) and 6072 Mg in Lei et al. (2013). High total Hg atmospheric burden in our model might be due to the following reasons: 1) Uptake of RGM by sea-salt aerosol is a dominant sink of RGM in marine boundary layer (Selin et al., 2007; Holmes et al., 2010). This mechanism is not included in the present model. 2) Oxidation of Hg by Br atoms is not included in the present model. 3) Oxidation products of Hg in the gas phase are all assumed to be gaseous Hg(II) in the present model. Take gas-particle partitioning of RGM into account in our model would increase the concentrations of particle Hg and result in larger deposition. By introducing the above processes, the sink of Hg will increase and the atmospheric burden of Hg will decrease.

Dry and wet deposition account for 78 and 22 % of total deposition in the

simulation results, respectively. This ratio is different with the results presented by Selin et al. (2008) and Amos et al. (2012). But it is comparable with the results of Selin et al., (2007). Actually, a more latest modeling studies (Lei et al., 2013) also gave a similar ratio. Dry and wet deposition contribute 70% and 30% to total deposition in their results. We think these differences might be caused by different Hg chemistry and deposition parameterizations and also meteorology inputs used in different models.

Figure R4 compares the WRF precipitation output with those from GPCP and NCEP reanalysis datasets. We can see that WRF can well reproduce the global distribution of precipitation. However, it seems to overestimate precipitation in regions near the equator but underestimate in the Northwest Pacific, the North Atlantic and ocean south of 30S. Considering large uncertainties in precipitation simulation in current models, these biases would be well acceptable. Finally, convective and synoptic precipitations are all included in our model. However, we do not distinguish the convective and synoptic precipitation in the wet deposition process, instead we just used total precipitation (convective+synoptic). The approach to treat Hg wet deposition is similar to the implementation described in ECHMERIT (Jung et al., 2009) and CAMx model.

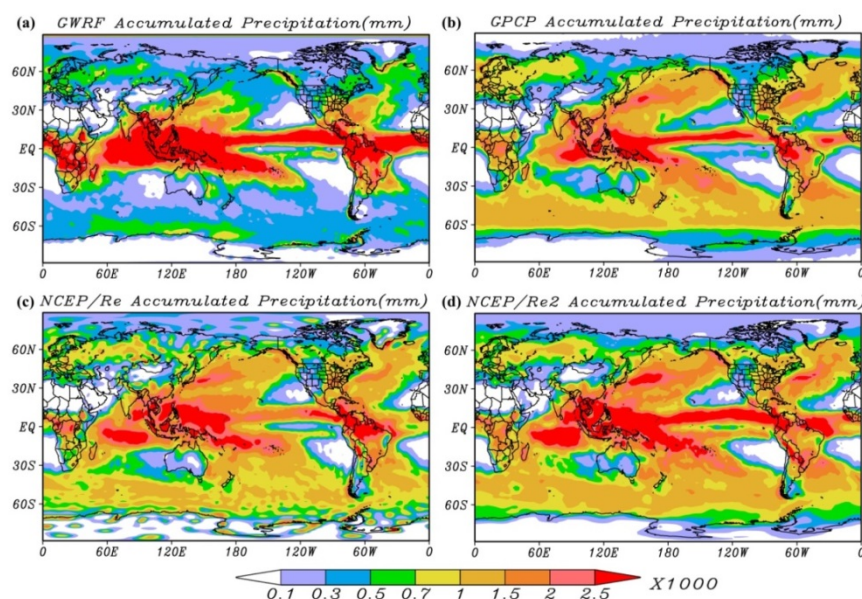


Figure R4 Global spatial distribution of accumulated precipitation simulated by WRF (a), and obtained from the GPCP (Global Precipitation Climatology Project) (b),

2.3.3 Total gaseous mercury

This comment, “High surface TGM concentrations are found in or downwind of areas with intensive mercury-relative mining (e.g. Western USA, Southern Africa),” is very odd. Looking at the emissions inventory for 2000, higher emissions in the Western US, compared to the Eastern US for example, are not apparent, and it is not clear what “intensive mercury-relative mining” might be. The emissions from South Africa are an error in the inventory, which the authors should be aware of. Why there should be such high TGM concentrations in Alaska is not clear either, the emissions database would not seem to indicate that there would be. And it seems unlikely that forest fires would cause such high concentrations over a whole year. Perhaps the authors should check their emissions interpolation routines. On the underestimate of the inter-hemispheric gradient, the articles cited are not the most recent and certainly in the authors model linking Hg emissions to CO emissions from the ocean will contribute to this problem. The authors finish this section mentioning the problem with the emission inventory, so why did they use it (or not change the South African emissions as they did with the Asian emissions), and why make the earlier comment about high concentrations downwind of mining errors if they know this is caused by erroneously high emissions?

Response: Yes, in the anthropogenic emission inventory (Figure S1 (a)), higher emissions in the Western US compared to the Eastern US are not apparent. However, as shown in Figure S2 (b), high geogenic emissions are found in Western US and Alaska. As stated in the manuscript (Line 26 Page 6957), the geogenic emissions represent mobilization of Hg by degassing from geological reservoirs and they are distributed according to the locations of Hg mines as an indicator of Hg deposits. The spatial distribution of geogenic emissions is consistent with those used in the GEOS-Chem model (Selin et al., 2007, 2008). Therefore, high TGM concentrations found in the Western US and Alaska are consistent with Hg emissions and are reasonable.

On the underestimate of the inter-hemispheric gradient, we agree that linking Hg emissions to CO emissions from the ocean might possibly contribute to this bias, and we will check further. Besides, we think there are two other major reasons causing this discrepancy. Firstly, the inability of present model to reproduce the air–sea exchange of Hg reasonably as stated in the manuscript. More specifically, this is due to upwelling mercury from the sub-surface ocean, possibly reflecting the legacy of past anthropogenic emissions (Holmes et al., 2010). This process will be implemented in a future model version. Secondly, simulated TGM concentrations in East Asia and Europe were underestimated, and the NMBs were -32% and -8%, respectively. All the above factors will be considered in the revised manuscript.

Again, we are very sorry that we do not know the emissions in South Africa are wrong in the AMAP 2000 inventory when we started the present work in 2010. We have replaced the Hg emissions in South Africa using the AMAP 2010 inventory and rerun the model. This update results in significantly decreasing of local Hg emissions and concentrations in South Africa.

2.3.4 Oxidized mercury

The bias reported in Table 3 for North America and Europe requires at least a comment.

Response: Thanks for the reminder. The explanation has been given in Line 5 Page 6963 in the manuscript. As follows “This discrepancy may partially be attributed to excessive oxidation of Hg(0) by relatively high concentrations of OH and O₃ (especially over the ocean) and uncertainties concerning Hg chemical speciation in emission inventories.”.

2.3.5 Wet deposition

It would have been useful to see a global map of wet deposition to compare the distribution to previous model results. There appears to be a discontinuity in figures 7c and d (roughly 20N, 120E).

Response: The global map of simulated Hg wet deposition is given in Figure R5. It

can be seen that the spatial pattern is related to precipitation, Hg emissions and oxidized Hg concentrations. Large wet deposition is found in East Asia, southeast America, and regions near the equator. This global pattern is generally reasonable compared to previous modeling studies (Holmes et al., 2010; Chen et al., 2014).

Yes, there is a discontinuity in figures 7c and d. This is caused by the two-way nesting simulation of WRF. Actually, we set three nesting domains (global, East Asia and the east part of China) in the WRF simulation, but we just used the uppermost two domains in the Hg simulation to save computation time. The discontinuity lies just right to the boundary of the innermost domain (e.g. 21N, 123E). We have checked this discontinuity in precipitation fields by compared two WRF simulations with two-way nesting option on and off, and found no big difference between these two simulations. Therefore, this discontinuity would not have large impact on our Hg modeling results.

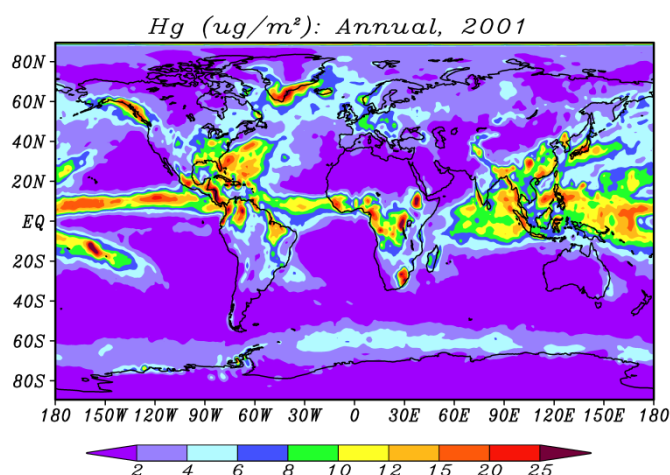


Figure R5 Simulated global annual Hg wet deposition in 2001

2.3.6 Dry deposition

The authors refer to observations, whereas dry deposition is not measured (unfortunately) but is inferred or calculated from bulk, throughfall and wet deposition measurements.

Response: We agree with the reviewer. The following explanation as “It should be noted that data used to evaluate model simulation of dry deposition is not directly measured, but is inferred or estimated based on measurements of total Hg in through fall and rainwater, wet deposition and atmospheric concentrations.” will be added to

Section 3.6 to remind the readers.

2.3.7 Model performance and comparison

The authors state that the model performs better for Europe and North America than for Asia. This is patently not true in the case of oxidized Hg. Also in this section the problem of comparing simulations from one year with observations from another comes up again. This makes very little sense. If the measurements, particularly in China were made relatively recently, and data from North America and Europe are available up until the present, what sense is there simulating 2001? Especially when it is known that the anthropogenic emissions database for that year is flawed. The 2005 emissions inventory has been available since 2008 and the 2010 inventory has been available since last year. It strikes me as a bizarre choice and scientifically speaking, inept and inappropriate.

Response: As illustrated in Table 3, the values of three statistical parameters (R, NMB, RMSE) for TGM and wet deposition are all better in North America and Europe than in East Asia. For oxidized Hg, the situation is more complicated. Two of the statistical parameters (R and RMSE) are much better in North America and Europe than in East Asia. However, lower NMB is found in East Asia. This is because the model overestimate oxidized Hg in some sites but underestimate in the other sites and therefore result in low model bias. The larger RMSE found in East Asia would support this explanation. Accordingly, we state that the model performs generally better for Europe and North America than for Asia.

Detailed explanation concerning the influence of the mismatch of the time periods between model results and observations has been given in the above response (2.3.1). We think the influence of the mismatch of the time periods may be relatively large for dry and wet deposition comparisons in East Asia but relatively small for other comparisons. However, no dataset of Hg deposition observations in East Asia is available at present. So we have no better choice. All observations of Hg deposition in East Asia (Table S4 in the supplement) are obtained from literatures. Every site has its own time period. It ranges from 1999 to 2009. If we change the base year of the

model simulation, it is still hard to match the time periods of all these observations.

Again, we are very sorry that we do not know the emissions in South Africa are wrong in the AMAP 2000 inventory when we started the present work in 2010. We have replaced the Hg emissions in South Africa using the AMAP 2010 inventory and rerun the model. This update results in significantly decreasing of local Hg emissions and concentrations in South Africa.

2.4 Conclusions

Unfortunately the model is unconvincing, the results are as well, therefore it is difficult to be sure that the conclusions drawn here are valid.

Response: In the above response, we have provided detailed information and explanations to answer the doubt raised by the reviewer concerning our model methodology and results. Besides, following the advices of the reviewer, we will introduce latest advances in Hg chemistry (e.g. Br chemistry, gas-particle partitioning of RGM) into our model to assess their impacts on Hg simulated results presented in this study. Overall, we think the present modeling results are generally reasonable and comparable to previous Hg modeling studies based on our comprehensive model evaluation, although several uncertainties are still needed further investigation. The conclusions drawn here are strictly based on the modeling results presented in this study and we insist that they are generally valid.

References:

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