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

General Comments:

This manuscript developed a Hg module in a nested atmospheric model, by considering the emissions, chemistry and deposition. The authors have evaluated the modeling of total gaseous mercury (TGM), oxidized mercury, wet and dry deposition of Hg. At last, the nested model was used to study the outflow of Hg from China (mainland?). This work may be potentially important, however, I have several serious concerns with the novelty and methodology of this study. There are many mercury models and it's not clear whether the model developed in the paper is more advanced than other models. For example, Br chemistry has been considered in other models (Amos et al., 2012), but not in the present model. Treatment of the re-emissions from land and ocean is a very weak aspect. In addition, there is a lack of detailed methodology in the model, in particular for some key chemical and deposition processes, making it hard to judge if the model is advanced or not. At last, as a major weakness, the diurnal variations and vertical trends are not evaluated, leaving it questionable whether the model captures the key chemical processes of Hg. In general, the present paper doesn't provide enough novelty to get published by GMD.

Response: Thanks the reviewer to state that our work may be potentially important. We agree that lack of enough model description and some potential key processes of Hg chemistry made the reviewer hard to judge if the model is advanced or not. We will do our best to plug into these processes and modules to improve the model. This study is tried to develop an online global nested Hg transport model with flexible horizontal resolution, but focus on the region of China and made better model performance. We afford a new online nested method to improve model performance in regional scale. 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 with the same physical and chemical

parameterizations among global and nested domains 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. Besides, 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 (especially published in Chinese journals) 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 investigated.

We made the major revisions for this paper to get published by GMD following comments of the reviewers and the editor. The responses to the serious concerns with model methodology used in our model as raised by the reviewer are as follows.

1/ Br chemistry

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 O_3, OH, H_2O_2 and reactive halogen species (e.g. Br, Cl, I, Br_2, Cl_2, 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 O_3 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 O_3-OH oxidation mechanism alone in their models. It seems that at the current level of understanding the O_3-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.

As shown by Table R1, we add five Br chemical reactions in the gas phase (Seigneur and Lohman, 2008) in addition to the O_3-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:

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

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	Hg(0)(g)+Br(g)→HgBr(g)	$k1 = 3.6 \times 10^{-13} P \left(\frac{T}{298}\right)^{-1.86}$ cm³ molec⁻¹ s⁻¹
BR2	HgBr(g)→Hg(0)(g)	$k2 = 3.9 \times 10^{9} exp\left(\frac{-8537}{T}\right)$ s⁻¹
BR3	HgBr(g)+Br(g)→HgBr₂(g)	$k3 = 2.5 \times 10^{-10} \left(\frac{T}{298}\right)^{-0.57}$ cm³ molec⁻¹ s⁻¹
BR4	HgBr(g)+OH(g)→HgBrOH(g)	$k4 = 2.5 \times 10^{-10} \left(\frac{T}{298}\right)^{-0.57}$ cm³ molec⁻¹ s⁻¹
BR5	Hg(0)(g)+BrO(g)→Hg(II)(g)	$k5 = 1.0 \times 10^{-15}$ cm³ molec⁻¹ 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 Hg(0) into Hg(II), which subsequently enhances the deposition of Hg(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 Hg(0) and Hg(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.
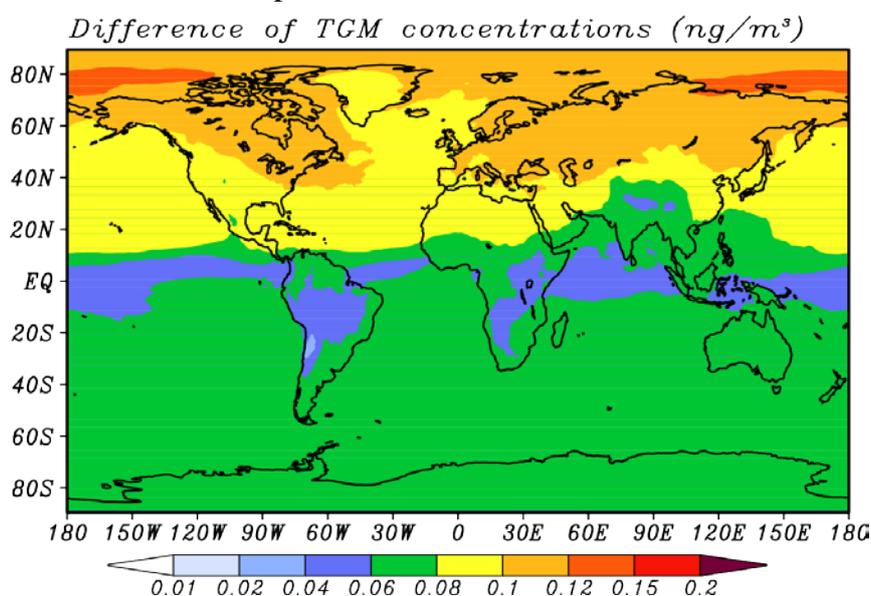

Figure R1 Change in surface TGM concentrations (ng m^{-3}) by introducing bromine chemistry

(positive value means the TGM concentration decreases after added bromine chemistry)

2/ Hg reemission

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 treatment of the reemissions from land and ocean 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.

3/ Lack of detailed model methodology

More detailed description concerning Hg chemistry and deposition parameterizations used in our model will be given in the revised manuscript. Besides, latest advance in Hg chemistry modeling (e.g. gas-particle partitioning of RGM, Br chemistry) will be included and tested in our model in the revised manuscript.

4/ Evaluation of diurnal and vertical variation

Considering no public hourly observational data is available, we didn't conduct evaluation of Hg diurnal variation in the manuscript. We will show the simulated diurnal variation of TGM with comparison to previous studies to confirm the model capability in the revised manuscript. For Hg vertical variation, we will try to collect relevant observations from aircraft field campaigns (e.g. INTEX-B, ARCTAS, and CARIBIC) to verify the model and give some comparison results in the revised manuscript.

Specific comments:

Introduction:

The major Hg chemistry and mechanisms are not well described (e.g. gas particle partitioning). The authors need to explain what they have improved in the modeling of Hg in the present work. Otherwise, there is no novelty.

Response: The major Hg chemistry and mechanisms are described in Section 2.2-2.4. Figure 1 and Table 1 also give the detailed Hg chemical reactions in gas and aqueous phase included in our model. The description of gas particle partitioning of RGM will be also added to Section 2.2 in the revised manuscript. In the first version of our model, we mainly follow previous studies to treat Hg chemistry and mechanisms. Our focus in this study is the online nested simulation and comprehensive model

evaluation in China and also the global impact of Chinese anthropogenic Hg emissions. To our point of view, the improvement in the modeling of Hg in the present work is the online nested capacity of our model. Online nested simulation has two advantages at least. Firstly, it can increase model resolution in the targeted region which could potentially improve the model performance. Secondly, the global and nested simulations use the same physical and chemical parameterizations which could avoid uncertainties induced by different boundary conditions. Therefore, we think the present work is a useful attempt and important.

2.2 Mercury chemistry

1/ the effects of temperature and relative humidity on Hg chemistry are not well explained.

Response: The reaction rate constants provided in Table 1 are for temperatures in the range of 20 to 25^0C. No temperature dependence information is available. The effect of relative humidity on Hg chemistry is not taken into account in our model. This treatment is in line with most Hg models.

2/ the treatment of the gas-particle partitioning of Hg (II) is not clear.

Response: In the present model, the oxidation products of Hg(0) in gas phase are all treated as gaseous Hg(II). As stated by another 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.

Mercury deposition:

1/ the method description is not clear. Detailed equations and parameterizations
for dry and wet deposition are needed, otherwise it is hard to judge if the model
is rigorous or not.

Response: The dry and wet deposition schemes used in our model are similar to the implementation described in the ECHMERIT (Jung et al., 2009) and CAMx (CAMx, 2014) model.

Dry deposition:

In the model, dry deposition is treated as a first-order removal mechanism. The deposition flux of a pollutant to the surface is the product of a characteristic deposition velocity and its concentration in the surface layer. Deposition velocities are derived from models that account for the reactivity, solubility, and diffusivity of gases, the sizes of particles, local meteorological conditions, and season-dependent surface characteristics. Dry deposition parameterizations of gases and aerosols are based on the work of Wesely (1989) and Slinn and Slinn (1980), respectively.

For gases, deposition velocity V_d is calculated from three primary resistances r (s m^{-1})in series as described below.

$$V_d = \frac{1}{r_a + r_b + r_s}$$

The aerodynamic resistance r_a represents bulk transport through the lowest model layer by turbulent diffusion. The quasi-laminar sub-layer resistance r_b represents molecular diffusion through the thin layer of air directly in contact with the particular surface to which material is being deposited. The surface resistance r_c depends upon the physical and chemical properties of the surface.

For particles, surface deposition occurs via diffusion, impaction, and gravitational settling. Particle size is the dominant variable controlling these processes. Particle deposition velocity for a given aerosol size is calculated using the following resistance equation.

$$V_d = V_{sed} + \frac{1}{r_a + r_b + r_a r_b v_{sed}}$$

V_{sed} is the gravitational settling (or sedimentation) velocity which is dependent on aerosol size and density.

The detail formulations of how to calculate r_a, r_b, r_s and V_{sed} for gases and

aerosols can be found in Wesely (1989) and Slinn and Slinn (1980) or the user's guide of the CAMx model (CAMx, 2014).

In the GNAQPMS-Hg model, dry deposition of Hg(0), Hg(II) and Hg(P) are all accounted for by adaption the parameterizations described above. Several physical properties (e.g. Henry's law constant, molecular weight, surface reactivity) of the Hg species are specified in order to calculate their deposition velocities. The Henry's Law constant for Hg(0) is set to be 0.11 M atm-1 (Lin and Pehkonen, 1999) with a temperature factor of -4970 K (Clever et al., 1985), and the surface reactivity is set to zero. Hg(II) represents $HgCl_2$ and $Hg(OH)_2$. Its Henry's Law constant is assumed to be the same as HNO_3 because they have similar solubility (Bullock and Brehme, 2002). Like HNO_3, Hg(II) has a strong tendency to stick to surfaces and its dry deposition occurs readily, so the surface resistance for Hg(II) in the dry deposition scheme is set to zero. The Hg(P) dry deposition velocity is set equal to that for sulfate, similar to that applied in the CMAQ-Hg and STEM-Hg model (Bullock and Brehme, 2002; Pan et al., 2008).

Wet deposition:

In the model, wet deposition of the chemical species are calculated using an approach with medium complexity. In-cloud and below-cloud scavenging are included. The basic formulation implemented in the model is a scavenging approach in which the local rate of concentration change $\frac{\partial c}{\partial t}$ within or below a precipitating cloud depends on a scavenging coefficient Λ:

$$\frac{\partial c}{\partial t} = -\Lambda c$$

The scavenging coefficient is estimated differently for gases and particles, based on relationships described by Seinfeld and Pandis (1998). For gases, two components are calculated: 1) direct diffusive uptake of ambient gases into falling precipitation; and 2) accretion of cloud droplets that contain dissolved gases. For particles, there are also two components: 1) impaction of ambient particles into falling precipitation with an efficiency that is dependent upon particle size; and(2) accretion of cloud droplets that contain particle mass. Overall, the scavenging coefficient depends on an assumed

scavenging efficiency, the total rainfall intensity (large-scale and convective precipitation), cloud water content and species solubility according to Henry's law, a mean cloud or rain droplet radius and rain droplet falling velocity. The large-scale and convective precipitation are not distinguished in this method. For species with low solubility (with a Henry's law constant of less than 100 M atm^{-1}), no wet deposition is calculated. More detail description of how to calculate the scavenging coefficients for gases and particles can be found in Seinfeld and Pandis (1998) or the user's guide of the CAMx model (CAMx, 2014). The physical properties (e.g. Henry's Law constant, surface reactivity, molecular diffusivity) of Hg species used in the wet deposition module are the same as those in the dry deposition module.

2/ it seems all precipitations are treated in the same manner, without distinguishing the large-scale and convective precipitation.

Response: Yes. In this model version, we do not distinguish the large-scale and convective precipitation in the wet deposition process. We consider the in-cloud and below-cloud scavenging of Hg and use different treatment for gaseous and particulate pollutants. The approach is similar to the implementation described in ECHMERIT (Jung et al., 2009) and CAMx model.

3/ for wet deposition, the release of Hg (P) when water freezes to ice is not considered.

Response: Yes. We will try to consider this process by following the work of Holmes et al.(2010) and Amos et al. (2012) in the future work.

Mercury emissions:

1/ how is the emissions from biomass burning, geogenic emissions, land and ocean specified for Hg(0), Hg(II), and Hg(P)?

Response: Following previous modeling study (Jung et al., 2009; Holmes et al., 2010), all Hg emissions from natural sources (e.g. biomass burning, geogenic emissions, land and ocean) are treated as Hg(0) in our model. This will be added to

the revised manuscript.

2/ neglecting the seasonality of Hg emissions from anthropogenic sources is a weakness of the present work.

Response: Seasonality of anthropogenic Hg emissions is important in model simulation. Unfortunately, global anthropogenic Hg emissions from AMAP and EDGAR which have been widely used in Hg modeling all have no seasonal variation. Therefore, it is very hard to obtain information about the seasonality of Hg anthropogenic emissions. We will try our best to get the seasonal profiles of anthropogenic emissions in China from Wang et al. (2014) by personal communication and consider seasonal variation of emissions in our new simulation.

3/a major weakness in this section is the treatment of Hg reemissions from land and ocean. The total emissions from land and ocean are not justified by any observations, and the method used in spatial allocation is not convincing. I don't see any relationship between the biogenic CO emission and the Hg reemission.

Response: 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. Reemissions are somehow relevant to biological activity (e.g. vegetation) so we used biogenic CO emissions as temporal and spatial surrogates. Certainly, reemissions are also relevant to deposition pattern, soil and water Hg content and environmental elements (e.g. solar radiation, wind speed). So the method of spatial allocation used in this study might induced some uncertainties, but we think the general spatial patterns are reasonable as show in Figure S3 in the supplement. In future work, parameterizations of air-sea and air-land Hg dynamic exchange should be included in the model to better resolve Hg reemissions.

Model setup

1/ some information are missed in this section (e.g. what is the time step in the model calculation? what is the vertical coordinate used in the model?)

Response: The time step in the model calculation (including emission, advection, diffusion, chemistry and deposition) is 600s. The meteorology input frequency is 6h in the global domain but 3h in the nested domain. These instructions will be added to the revised manuscript.

The description of the vertical coordinate used in the model has been given in Section 2.5 in Page 6958. That is "Vertically, the model uses 20 terrain-following layers from the surface to 20 km a.s.l., with a decreasing resolution with height. Roughly, the lowest 14-18 layers are in the troposphere and the remaining layers are in the stratosphere.".

2/ a coarse-resolution inventory (0.5 degree for AMAP, and 0.5 degree when using GEIA inventory for an interpolation) does not match the resolution in the model (0.3 degree).

Response: Thanks for the comment. We used a mass-conservative interpolation method to remap the emission inventory to match the model grid in the global and nested domains. Firstly, we divide the emissions in a 0.5^0x0.5^0 grid into 2500 small grid with resolution of 0.01^0x0.01^0. Then every 0.01^0x0.01^0 grids are match to the 1^0x1^0 and 0.33^0x0.33^0 grids based on their central latitudes and longitudes. This method make sure the total emissions in the global and nested domains are the same.

Model evaluation

Line 17, Page 6960: the time periods of the measurements do not match with those of the simulations. Dismatch of the time periods when comparing the model with the observations is a major weakness. In particular, there is a large bias when comparing modelled annual mean Hg concentrations with daily measurements by cruise. Unfortunately, the authors only attribute model-observation discrepancies to this dismatch, without making any efforts to assess this influence.

Response: We agree that dismatch of the time periods when comparing the

model with the observations might be 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 dismatch 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 dismatch 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 dismatch 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 dismatch 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. By saying "It should be noted that the time periods of the measurements do not all match with those of the

simulation, and this difference may partially explain any model–observation discrepancies.", we just want to remind the readers that this is one of the factors causing the model-observation discrepancies. Detailed assessments of the influence are difficult and outside the scope of this study.

Total gaseous mercury (TGM)

1/ Fig. 3: scatter plots by region are needed to evaluate the model performance when comparing model with observations.

Response: Scatter plots by region between simulated and observed surface TGM have been given in Figure 10 (a) in the manuscript.

2/ a major weakness in this section is that the reasons for the discrepancies are not well explained. There are uncertainties in emissions, chemistry, and deposition. Without discussion on these sources of errors using enough sensitivity tests, it is hard to judge if the treatments of these processes in the model are rigorous or not.

Response: Following the advices of the reviewer, we will provide more detailed explanations for the TGM model-observation discrepancies found in our results.

3/ the modeled TGM over the Pacific is 1.4-1.6 ng/m3, compared to the observed 2.6-3.0 ng/ m3. However, this large discrepancies have not been explained.

Response: As stated in Line 4, Page 6962, this discrepancies can be attribute to the inability of current models to reproduce the air–sea exchange of Hg reasonably. 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.

4/ Fig. 5: in East Asia, as a most important source region, the model doesn't capture the low concentrations in summer and overestimates the TGM concentrations in autumn, and these discrepancies are not explained. As a result, it seems that the model doesn't capture the key processes governing the chemistry and deposition of

Hg.

Response: We will try to give more detailed comparisons of TGM monthly variation site by site and the discrepancies will be explained in the revised manuscript.

Oxidized mercury

Line 5, Page 6963: the authors don't provide convincing explanation for the overestimation of the oxidized mercury concentrations. As a result, it seems that the model fails to simulate the key processes governing the chemistry and deposition of Hg.

Response: As stated in the manuscript, we attributed the overestimation of the oxidized mercury concentrations to excessive oxidation of Hg(0) by relatively high concentrations of simulated OH and O_3 and uncertainties concerning Hg chemical speciation in emission inventories. We think these are the leading factors. Of course, uncertainties of Hg chemistry (gas-particle partitioning of RGM, in-plume reduction of RGM) and deposition processes in the present model should also contribute to this discrepancy. These factors will be further considered in the revised manuscript.

Dry deposition

Line 13, Page 8: the authors attribute the model overestimation to Hg(II) and Hg(P) emissions. However, a discrepancy of 98 v.s. 648 pg m-3 is out of the uncertainty range of emissions. It seems that the model fails to simulate the key processes governing the chemistry and deposition of Hg.

Response: We agree that many factors including model emission, chemistry, deposition, horizontal resolution should contribute to this overestimation. We believe that uncertainties of emission magnitude and speciation would be a major factor. As can be seen from Figure S1 and Figure 9, Hg emission and dry deposition share a similar spatial distribution with high values in the central and east coast of Japan. Especially, the largest emission and deposition are found in the Tokyo area. Therefore, we can see that dry deposition have high relation to emission. The emission magnitude is possibly overestimated in the Tokyo area due to the fact that population

density is used as surrogate to map the emission. Besides, emission speciation for Hg(0), Hg(II) and Hg(P) should also be important because dry deposition in the Tokyo area is mainly contributed by Hg(II) deposition.

3.7.1 East Asia vs. North America and Europe

1/ Line 5, Page 6966: I suggest that the authors give some estimates of the Hg emissions from 2000-2010 to support their first explanation.

Response: Thanks for the suggestion. Wu et al. (2006) estimated that anthropogenic emissions in China increased at average annual rate of 2.9% during the period 1995-2003. Liang et al. (2013) pointed out that Hg emissions in China had increased by 164% during 1992-2007. These studies can support our first explanation. We will add these references to the revised manuscript.

2/ Line 6, Page 6966: I suggest that the authors compare the model-observations discrepancy over East Asia between the global and nested model to support their second explanation.

Response: The comparison of model performance over East Asia between the global and nested simulations has been done and shown in Table 3 and Figure 11. The results are consistent with the second explanation.

3/ Line 6, Page 6966: there is no evidences showing that the emission uncertainty in East Asia is larger than that in North America and Europe.

Response: Uncertainties of emission inventory are mainly origin from the activity data and emission factor. These fundamental data in developing countries (e.g. China) are not as adequate and accurate as those in North America and Europe. Study by Muntean et al. (2014) has shown that uncertainties of Hg emissions in "Non Annex I" (developing) countries are larger than those in OECD90 and EIT countries. Besides, Large underestimations in Hg anthropogenic emissions over East Asia have been demonstrated in several previous studies (Jaffe et al., 2005; Pan et al., 2007). Based on above reasons, we believe that the emission uncertainty in East Asia is larger than

that in North America and Europe.

4/ In general, it should be careful when comparing the different model performance among different regions. The miss of some chemical processes in the model should also explain the poor model performance over East Asia. Insufficient explanation of the discrepancy causes the model to be very uncertain.

Response: We agree with the reviewer. The missing of some chemical and physical processes such as gas-particle partitioning of RGM, in-plume reduction of RGM, dynamic reemission in the present model should also explain the poor model performance over East Asia. We will try to give more detailed and in-depth explanations of the model discrepancies over East Asia.

3.7.2 Global vs. nested simulations

The authors state that the emission, chemistry and deposition are self-consistent between the global and nested simulation. However, from Figure 7 and Figure 9, the regional TOTAL wet and dry deposition seem to be very different between the two simulations. I am not sure if this is only due to the smooth effect of mapping. I suggest that: 1/ the authors remove the smooth effect in these maps by showing the original model resolution;

Response: By stated self-consistent we just want to express that the total emissions, the physical and chemical parameterizations (including advection, diffusion, dry and wet deposition, chemistry) used in the global and nested simulation are the same. Actually, as stated in the manuscript (Section 3.7.2), Hg dry deposition amounts decrease notably in the coastal regions while wet deposition amounts increase in mountain regions of southeast China in the nested simulation. This resulting in model performance improvement in the nested simulation and will also resulting in different total dry and wet deposition between the global and nested simulation. The study of Zhang et al. (2012) using GEOS-Chem model also show similar results. We don't think the regional total deposition will keep the same in the global and nested simulations. Following the reviewer's advice, we will used gridfill shaded figures

which can remove the smooth effect to replace the original Figure 7 and Figure 9.

2/ there should be maps showing the absolute and relative differences between the two simulations;

Response: We agree with the reviewer. We will add some figure in the revised manuscript to show the absolute and relative differences between the two simulations.

3/ there should be a detailed comparison of Hg budgets between the two simulations. Then, the author should provide enough evidences to substantiate that the two simulations are really self-consistent.

Response: We will calculate and compare the Hg budgets in the two simulations. However, we don't think the two budgets will be the same. As can be imagined, more detailed structure of the emissions, landuse and precipitation are revealed in the nested domain and this will influence the Hg dry and wet deposition.

Impacts of Chinese primary anthropogenic sources

1/ the authors state that 30% of surface Hg concentrations was contributed by China's primary anthropogenic sources. Then, what sources and which regions contributes to the remaining 70%? If the contribution of Hg reemissions is large, only accounting for the impact of China's primary anthropogenic sources would have very limited significance.

Response: In this study, we just focus on the impacts of Chinese primary anthropogenic sources due to the fact that emission control measures can only be implemented on primary anthropogenic sources in most of the time. Except Chinese primary anthropogenic sources, other local sources including natural emissions (from soil, vegetation and water), biomass burning emissions and reemissions of previous deposited Hg, and long-range transport should contribute to the remaining 70% of surface Hg concentrations in China. Quantitative assessment of the contribution of different sources and regions to surface Hg concentrations in China is out of the scope of this study. Actually, in-depth analysis of the source apportionment of Hg

concentrations and deposition has been conducted by Wang et al. (2014) by using the GEOS-Chem model.

2/ change "Hg concentrations" to "surface Hg concentrations" at any place if necessary.

Response: This will be done in the revised manuscript.

3/ the trans-Pacific transport of Hg is not validated by any observations (e.g. the time series at Okinawa).

Response: Actually, model evaluation of Hg concentrations and deposition in East Asia (including China, Japan, Korea) and North America have been shown in the manuscript. This to some extent validated the trans-Pacific transport of Hg. Following the reviewer's suggestion, we will try to collect Hg observations in Okinawa and give some initial comparison.

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