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Interactive comment on "Intercontinental transport and deposition patterns of atmospheric mercury from anthropogenic emissions" *by* L. Chen et al.

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Dear reviewer,

Thank you very much for your valuable suggestions and comments. Responses to the comments are presented below (Note: questions from the reviewer are in bold and responses from the authors are behind **REPLY**):

(1) The manuscript describes the simulation of atmospheric Hg emission, transport, atmospheric chemistry and deposition using a well-known model over a period of years using tagged emissions to investigate source receptor relationships between various

regions of the globe. The results obtained are much as would be expected given the lifetimes of the atmospheric Hg species in the atmosphere, atmospheric circulation patterns and global precipitation patterns. The use of the average of a multi-year simulation, further enhances the predictability of the results obtained, as it serves to smooth out the year on year differences. **No changes are made to the original model, which is already well documented in the literature.**

REPLY: This manuscript is an applied study. We didn't change the original model, as did in many previous studies. A well-known model was used to establish systematic and comprehensive source-receptor relationships of atmospheric mercury from primary anthropogenic emissions on intercontinental scales for the first time. The results showed quantitative and comprehensive source–receptor relationships among major continental regions worldwide and provided a timely picture of current mercury fluxes before implementation of the UNEP Treaty on mercury. Details of the novelties of the study could be seen in the following replies to the questions.

(2) **No new comparisons between observations and modelled values of Hg species concentration fields are made, nor are new comparisons between observed deposition and modelled deposition made.**

REPLY: Thanks for the comments. As suggested by the reviewer, comparisons between observations and modeled values of TGM concentrations will be added in the revised manuscript. Results of the comparisons are presented in Fig.1 at the end of this Response Letter. The observations are from CAMNet, AMNet, EMEP and East Asia literature data. We used the observational data during 2005-2011 and averaged the data through the period for each site. The correlation coefficient (R=0.79) in Fig. 1a indicates that the model agrees with the observations very well.

In the original manuscript, comparisons between the observed and modeled values were not conducted, because: 1) The objective of this manuscript is to use the model to evaluate intercontinental transport and deposition patterns of atmospheric mercury

from primary anthropogenic emissions, and not to improve the model; 2) extensive work has been done for the model verification (Holmes et al., 2010; Soerensen et al., 2010; Amos et al., 2012); 3) There are also practices in literature that the well-constrained model is applied for specific research purpose without comparisons between observations and modeled values, such as Corbitt et al. (2011), Amos et al. (2013).

(3) **The authors have chosen to look at continent to continent source receptor relationships when it is ocean receptors, via Hg methylation and bioaccumulation in predatory fish and their consumption, which, with the exception of extreme cases of occupational exposure, represent the most important risk to human wellbeing.**

REPLY: Hg deposition to oceans and subsequently bio-accumulation in marine fish is one of the most important exposure risks to human. Oceans are the most important receptors and their sources are mainly from land (Corbitt et al., 2011). Therefore, as source regions, continents are important. Meanwhile, Hg deposition to terrestrial ecosystems, such as lakes and watersheds, and bio-accumulation in some vegetation is also important which drew less attention than oceans. Some studies have been conducted on the continent to continent source-receptor relationships. Jaffe and Strode (2008) demonstrated that Asian anthropogenic sources contributed to 7–20% (average 16%) of mercury deposition in North America. Zhang et al. (2012) indicated that 10–22% and 13–20% of mercury wet and dry deposition, respectively, in the US originated from North American anthropogenic sources. Corbitt et al. (2011) studied the contributions of Hg deposition on some continental receptors (e.g. North America, South America, Europe) from other continental sources, and they also showed the contributions from natural sources. Lin et al. (2010) claimed that global anthropogenic sources accounted for 75% of mercury deposition in East Asia, with 25% from natural sources. These studies have focused on selected continental regions (e.g. East Asia, North America) and were not systematic and comprehensive worldwide source-receptor re-

lationships studies. This study attempted to establish systematic and comprehensive source-receptor relationships on intercontinental scales for the first time.

(4) I believe there are of aspects concerning this article which render it currently unsuitable for publication. In the following, roughly in the order in which they appear in the article, are the major, and some minor points which render the manuscript unsuitable for publication. The Abstract starts by mentioning global policies that regulate anthropogenic Hg emissions. **The draft text of the Minamata Convention was agreed in January 2013, it was adopted at the Conference of Plenipotentiaries in Kumamoto on the 10th October 2013, and in two days over ninety countries signed it. I think this should have been mentioned,**

REPLY: Thanks for the comments. This manuscript was submitted on 26th July 2013, and the event occurred on 10th October 2013. This will be updated in the revised manuscript.

(5) **in the Introduction there is a mention of a UNEP global treaty on Hg, with one not very pertinent reference and no link to the UNEP site. Neither the UNEP Global Mercury Assessment nor its Technical Background Report are mentioned.**

REPLY: Thanks for the comments. An official reference, link to the UNEP site and the UNEP report will be added in the revised manuscript.

(6) The Abstract continues, saying that global policies require quantitative and comprehensive source-receptor relationships between continental regions. Most human exposure to Hg (except in cases of extremely polluting industries or artisanal gold mining) is through fish consumption. **Therefore the important source-receptor relationships are between continental emission sources and oceanic receptors, especially those which are major fisheries. Contamination of lakes and rivers can be important, but really only a local/ regional scale, not global.**

REPLY: Thanks for the comments. As seen in our previous answer, we do agree with the reviewer that source-receptor relationships between continental emission sources and oceanic receptors are very important. However, source-receptor relationships between continental regions are also important (Jaffe and Strode, 2008; Lin et al., 2010; Corbitt et al., 2011; Zhang et al., 2012, see question (3) for detail). This study is the first one to establish systematic and comprehensive source-receptor relationships on intercontinental scales. The novelties of this study lie in: First, the study covers all the continents and the world is divided into eleven regions. Some regions have never been considered before (eg, Russia, Africa, Southeast Asia). Second, both air concentrations of the three mercury species and mercury deposition are considered in this study. Third, the source contributions are divided into primary and secondary contributions, and readers can get valuable information from secondary contributions. Fourth, fluxes of some important processes, which have not been considered in previous studies (Holmes et al., 2010; Soerensen et al., 2010; Amos et al., 2012), such as partitioning between Hg(II) and Hg(P), adsorption by sea-salt aerosol, the deposition and burden of Hg(P), were given in our budget. Above all, as sources and receptors of mercury, continents are important, and detailed studies are needed.

(7) **I would also suggest that global policies require global measurements, and ideally global monitoring, something which has been lacking in the case of Hg.**

REPLY: Thanks for the comments. We do agree with the reviewer. Global measurements and monitoring are not only important to global policies, but also important to model simulations. More efforts should be made to add the databank of global monitoring of mercury.

(8) The Abstract then discusses the different Hg species which are emitted, and their transport/deposition characteristics. **The emission inventory used is based on 2005 as a reference year. While this is appropriate for at least part of the of the modelling simulation period, it certainly is not towards the end. Part of UNEP's Global Mercury Assessment was the preparation of the AMAP/UNEP 2010 global inven-**

tory. This inventory suggests that by 2010 the major anthropogenic source of Hg to the atmosphere was artisanal and small-scale gold mining, and not coal combustion. This inventory therefore has a distinctly different global emission distribution with respect to that of 2005.

REPLY: Comparisons between observations and modeled values under 2005 inventory and 2005-2010 inventory were conducted. Based on linear interpolation, the two inventories were used to interpolate into yearly inventory during 2005-2011 for 2005-2010 inventory. Results are presented in Fig.1a and Fig.1c at the end of the Response Letter. The results show that surface air Hg concentration distributions of 2005 inventory (R=0.79) and 2005-2010 inventory (R=0.80) are similar. The consistent comparisons show that 2005 inventory is also feasible and valid for the simulation during 2005-2011.

In addition, 2005 inventory is used widely by many previous studies under meteorological fields of recent years (2004-2010) (Corbitt et al., 2011; Amos et al., 2012; Soerensen et al., 2012; Zhang et al., 2013). So it could represent the simulation period in this study. Meanwhile, it is convenient for this study to compare the surface air Hg concentrations and global Hg budget with other studies by using the same inventory.

For the emission inventory in 2005 and 2010: 1) The total emissions are similar (1920.6 tons in 2005 and 1875.5 tons in 2010); 2) The emissions in each region are also similar (East Asia: $\sim$1210 tons in 2005 and $\sim$1000 tons in 2010; North America: $\sim$150 tons in 2005 and $\sim$110 tons in 2010). The main difference between them is emission amount in each species. 2010 inventory has higher emissions in Hg(0) and lower emissions in Hg(II) than 2005 inventory. However, this is not essential for this study. Because the speciation adjustment based on in-plume reduction was involved in this study (see the response to question (15)). After adjustment, Hg(0) emissions were higher and Hg(II) emissions were lower than before. Emission amount in each continent is important for this study, and in this respect, there is no significant difference between the two inventories. In addition, the change of the main emission sector (power plants to ASGM) is

also not important for this study, because we only focused on emission amount in each continent.

(9) **(Which was also AMAP/UNEP, not GEIA as the authors state later in their manuscript, the ECCAD-GEIA portal provides a link to the AMAP site)**

REPLY: Thanks for the comments. This will be corrected in the revised manuscript. Many previous works on AMAP global emissions inventories employed a population distribution dataset available from GEIA, and ECCAD-GEIA portal (http://eccad.sedoo.fr/eccad_extract_interface/JSF/page_login.jsf) provides a link to the AMAP site. Thus many previous studies used this appellation (Selin et al., 2008; Holmes et al., 2010; Soerensen et al., 2010; Amos et al., 2012; Zhang et al., 2012).

(10) In the Introduction, a number of the points above could be reiterated. **The authors mention some previous studies on source-receptor relationships (line 11 page 25188) but do not cite any references.**

REPLY: Thanks for the comments. "Previous studies" here refers to the studies stated in the former sentences, including Jaffe and Strode (2008), Zhang et al. (2012), Lin et al. (2010). "Previous studies" will be replaced by "these studies above". We will also check the whole manuscript carefully.

(11) **The Task Force on Hemispheric Transport of Air Pollution (TF HTAP) (http://htap.org/) published its first comprehensive report in 2010 and its work is continuing. The report contains some source receptor studies and the description of some modelling experiments designed to evaluate the impact of regional emission reductions.**

REPLY: Thanks for the reminding. We will cite it in the revised manuscript.

(12) Section 2.1: **The model description describes the recent changes made to the GEOS-Chem model which have been made to try to improve the representation of Hg processes in the model, and to improve the model vs observation**

comparisons. None of it is the authors own work.

REPLY: Please see the response to question (1).

(13) Section 2.2: **As mentioned above the emissions database used by the authors is from AMAP/UNEP not GEIA.**

REPLY: Thanks for the comments. We will replace it by AMAP/UNEP.

(14) **It was not valid for the later part of the period simulated by the model in this work.**

REPLY: Please see the response to question (8).

(15) The discussion on in-plume reduction regards the speciation used in the emissions database for power plant plumes. Hg speciation in these plumes depends very much on the flue gas cleaning technology employed, which in turn varies throughout the world. The studies cited refer specifically to the US, and the changes in Hg speciation adopted may not be valid worldwide. Quoting the manuscript text "we adjustment the mercury speciation : : :..", and this is precisely what they are, given the uncertainties in Hg models, they are 'tuned' to fit available data and thus hopefully to provide further insights into the global and/or regional Hg cycle. **In the case of Amos et al., 2012 and Zhang et al., 2012, the changes made to the model were justified by comparison to observations. However these were all in North America and it does not necessarily follow that the improvements in the simulations are valid globally. Specifically Zhang et al., 2012 adjusted the emission speciation in the National Emission Inventory (NEI) which is specific to the US, there was no mention of applying the same emissions speciation ratio to non-US emissions. In order to justify extending this change to the emission ratios the authors should compare their simulation results to measurements. However there are no comparisons with measured data made in this article even though data for the period are available, including for China, see Lin et al., 2010.**

REPLY: Comparisons between observations and modeled values under in-plume reduction and no in-plume reduction were conducted. Results are presented in Fig.1a and Fig.1b at the end of the Response Letter. Fig. 1a shows the comparison between observations and modeled values under in-plume reduction and Fig. 1b shows that under no in-plume reduction. The two panels indicate that the model performs better with in-plume reduction (R=0.79) than without it (R=0.78), especially for East Asian sites with high TGM concentrations. Results show that the changes in Hg speciation adopted are valid worldwide.

Hg speciation in the atmosphere after emitting from power plant plumes in this study and the cited studies does not depend very much on the flue gas cleaning technology employed. Dirigible measurements by A ter Schure et al. (2011) suggested that high Hg(0) and low Hg(II) in downwind of plumes didn't reflect errors in the emission inventories. It reflected rapid in-plume reduction operating by a mechanism in the background atmosphere which had not been found. Thus in-plume reduction is a chemical phenomenon which occurs in the atmosphere near the power plants. It is unrelated to the initial emission speciation in the power plants. The chemical phenomenon does not vary throughout the world. Kos et al. (2011) implemented a 90:8:2 speciation in the GRAHM global model and Amos et al. (2012) followed the speciation from Zhang et al. (2012) in the GEOS-Chem global model to do global simulations. No matter which emission inventory is involved (US emissions or non-US emissions, NEI or UNEP/AMAP), the well-confirmed atmospheric chemical rule is applied to obtain reliable results. This methodology is applied widely in GEOS-Chem global simulations.

(16) Section 2.3: The tagging of the Hg emissions is problematic because the authors use the same emissions database for the whole simulation period. **The most recent anthropogenic emissions database from AMAP/UNEP shows such significant changes in the global distribution of the emissions that it renders practically invalid the last 3 or 4 years of the simulation period.**

REPLY: Please see the response to question (8).

(17) **The recent papers by Amos et al. (GBC CHECK) Noelle, clearly point out the importance of legacy Hg in the global biogeochemical cycle of Hg.**

REPLY: As emission source, legacy Hg reemitted by ocean and land was simulated in this study (the category: Nature). We calculated its source contributions to each continent. This study aimed to report source-receptor relationships among continents from primary anthropogenic emissions. Legacy Hg was not the emphasis of this study although we considered its contributions. Control measures could be implemented on primary anthropogenic emissions but difficult on legacy Hg. That is why we focused on the source-receptor relationships for primary anthropogenic emissions, since they were important for present-day Hg cycle and policies.

(18) **Results section In the introduction to the results: the AMAP/UNEP emissions database include three height levels the highest of which is over 150m, was this taken into account.?**

REPLY: Yes, we considered the emission at the three levels and used the combined data in the simulation. Because the height of the boundary layer in GEOS-Chem exceeds 150m in most regions.

(19) **The period 2005 – 2011 is not representative of a decade as three different global emission databases are available for 200, 2005, and 2010.**

REPLY: Here, the "over the last decade" is a wrong expression, and will be replaced by "during this period".

(20) Section 3.1: **the emission inventory is from AMAP/UNEP.**

REPLY: Thanks for the comments. We will replace it by AMAP/UNEP.

(21) **The authors state that the result obtained for the Hg(0) surface concentrations are consistent with Holmes et al, 2010, as it is the same model with a few updates, that is to be expected. It would be more interesting to know how the model compares with observations.**

REPLY: Thanks for the comments. In the revised manuscript, comparisons between observations and modeled values of TGM concentrations will be added. Results could be seen in Fig. 1a which shows that the model performs well (R=0.79). The long-term simulation results were feasible and applicable in establishing source-receptor relationships. Compared to Holmes et al. (2010), we used the same model with some updates. The simulation period and emission inventory in this study were different from them.

(22) **The interhemispheric gradient is described as stronger, stronger than what?**

REPLY: Here "stronger" should be "strong". Revision will be made.

(23) Line 5 p 25194. **'Because of accumulation : : :.. ', whether Hg(II) and HgP accumulate in the in the upper troposphere and stratosphere because they are more stable at low temperatures, does not necessarily explain their low concentrations in the boundary layer where they are removed by deposition processes more rapidly than Hg(0), which would explain the predominance of Hg(0), as would the fact that most emissions are Hg(0).**

REPLY: Thanks for the comments, We will revise this sentence with "Compared to accumulation of Hg(II) and Hg(P) in the upper troposphere and stratosphere (Selin et al., 2007; Lyman and Jaffe, 2011), their concentrations at the surface are low because of rapid deposition." Lyman and Jaffe (2011) demonstrated oxidized mercury (Hg(II) and Hg(P)) was positively correlated with stratospheric tracers (ozone and potential vorticity (PV)) and negatively correlated with CO, indicating that Hg(II) and Hg(P) increased with the increase of stratospheric influence.

(24) **The rapid oxidation of Hg(0) by Br in polar regions depends on the season.**

REPLY: We will revise the sentence with "High surface Hg(II) concentrations occur in polar regions, likely because of high Br atoms concentrations and Hg(0) oxidation rates during the AMDEs in spring in polar regions when using the mercury+Br model".

(25) **Comparing regional model output which has a predominantly continental domain, with global model output is not valid.**

REPLY: Thanks for the comments. The sentence "This is in agreement with values of 30–50% found in some regional model studies (Lin et al., 2007, 2010)" will be deleted in the revised manuscript.

(26) Section 3.2: **The global budget is similar to that from previous studies using the same model. As would be expected, I imagine.**

REPLY: Although fluxes of some processes are similar to those from previous studies, fluxes of some other important processes, such as partitioning between Hg(II) and Hg(P), adsorption by sea-salt aerosol, the deposition and burden of Hg(P) which have not been considered in previous studies (Holmes et al., 2010; Soerensen et al., 2010; Amos et al., 2012) were given in our budget. Meanwhile, due to some model updates, fluxes of some important processes are different from previous studies in our budget. For example, the photoreduction flux of Hg(II) to Hg(0) is 1900 Mg a^{-1}, nearly half that from Holmes et al. (2010); Hg(0) dry deposition to ocean (50 Mg a^{-1}) is significantly different from earlier global model predictions (Selin et al., 2008; Holmes et al., 2010; Corbitt et al., 2011). We will replace the sentence "Our global budget is similar to those presented in Holmes et al. (2010), Soerensen et al. (2010) and Amos et al. (2012)" with "Fluxes of some processes in our global budget are similar to those presented in Holmes et al. (2010), Soerensen et al. (2010) and Amos et al. (2012)".

(27) **p25195 the authors state 'we improve the proportion of Hg(0) based : : :: : :', when it was actually Amos et al., and Zhang et al., who improved the model.**

REPLY: Sorry for the confusing. We will change the "improve" with "increase".

(28) **And as mentioned before the extrapolation of the Hg emission ratios from Zhang et al. to the whole of the global emissions database, has not been justified.**

REPLY: Please see the response to question (15).

(29) **Last line; sequestered? Buried with sediments in the deep ocean?**

REPLY: Many previous studies used this word to describe that Hg was buried with sediments in the deep ocean (Holmes et al., 2010; Corbitt et al., 2011; Amos et al., 2013).

(30) Sections 3.4 and 3.5 **These sections are long-winded, rather confusing (too many acronyms) and contain information which, given the number of studies on general circulation patterns and the long-range transport of numerous pollutant species with varying lifetimes, does add much to the sum of knowledge concerning Hg.**

REPLY: Thanks for the comments. Many of the acronyms are abbreviated names of the eleven continents. We used them to reduce the page length.

(31) **It also misses the point that deposition to oceans is of extreme importance.**

REPLY: Please see the response to the question (3).

(32) **p25199 l18 onwards, as the authors note Lin uses a different emissions database, at this point surely the most obvious thing to do would have been to compare the results of this study with observations.**

REPLY: Please see the response to the question (2).

(33) **The difference in the N. American deposition found by Philip et al., could simply have been checked by summing the same region as Philip et al., to see if the values are comparable. It would certainly have been preferable to the rather offhand last sentence in this paragraph. The last line of section 3.4.1 does rather seem to be stating the obvious.**

REPLY: Sorry for the confusing. Our purpose here was not to make comparisons between the results in this study and the results from Philip et al. (2007) and Lin et

al. (2010). We attempted to show the deposition fluxes in some selected regions (such as East Asia, North America) in this study. This paragraph will be revised to only show our deposition fluxes. Comparisons are not needed here because more valuable comparisons of TGM concentrations and budget will be added. These two last sentences in the two paragraphs will be deleted in revised manuscript.

(34) **Overall the article is simply the description of what happened when the authors ran somebody else's model and tagged the emissions from some areas of the globe. The results are as is to be expected given the lifetimes of the Hg species in the model.**

REPLY: Although we ran somebody else's model, there are significant novelties in this study. First, the study covers all the continents and the world is divided into eleven regions. Some regions have never been considered before (eg, Russia, Africa, Southeast Asia). Second, both air concentrations of the three mercury species and mercury deposition are considered in this study. Third, the source contributions are divided into primary and secondary contributions, and readers can get valuable information from secondary contributions. Fourth, fluxes of some important processes, which have not been considered in previous studies (Holmes et al., 2010; Soerensen et al., 2010; Amos et al., 2012), such as partitioning between Hg(II) and Hg(P), adsorption by sea-salt aerosol, the deposition and burden of Hg(P), were given in our budget. Previous studies (Philip et al., 2007; Jaffe and Strode, 2008; Lin et al., 2010; Corbitt et al., 2011; Zhang et al., 2012) have focused on selected continental regions (e.g. East Asia, North America) and were not systematic and comprehensive worldwide source-receptor relationships studies. The results of this study showed quantitative and comprehensive source–receptor relationships among major continental regions worldwide and provided a timely picture of current mercury fluxes before the implementation of the UNEP Treaty on mercury.

(35) **There is no comparison of model results with observations. The emission inventory used is out of date, and the authors change the Hg species emission**

ratios following a study which used a different emission database and was a regional rather than global study.

REPLY: Please see responses to the question (2) (8) (15).

(36) **There were no experiments to assess the impact of increasing or decreasing emissions in any of the anthropogenic Hg source regions.**

REPLY: Thanks for the comments. Sensitive scenarios are good points for model predict simulations. However, given the page limit of one paper, we didn't make it in this manuscript.

(37) **This article lacks any significant new scientific contribution, and contains no results which are particularly new or interesting.**

REPLY: As indicated above, this study is the first work to establish systematic and comprehensive source-receptor relationships on intercontinental scales. There are significant novelties in this study (see responses to the question (6) (34)).

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Note: Followings are the results of our additional work after receiving the comments from the reviewers. We hope they are helpful for further review.

Fig. 1. Comparisons between modeled values and observations under three scenarios: (a) In-plume reduction used globally with 2005 inventory; (b) No in-plume reduction but 2005 inventory used; (3) In-plume reduction used globally with 2005-2010 inventory (Based on linear interpolation, the two inventories were used to interpolate into yearly inventory during 2005-2011). Observations are from CAMNet (http://www.ec.gc.ca/natchem/default.asp?lang=En&n=BFF7F7EE-1), AMNet (http://nadp.sws.uiuc.edu/amn/), EMEP (http://www.nilu.no/projects/ccc/index.html) and East Asia (Literature data: Wang et al., 2007; Fu et al., 2008a; Fu et al., 2008b; Fu et al., 2010; Wan et al., 2009a; Wan et al., 2009b; Chand et al., 2008; Ci et al., 2011a; Ci et al., 2011b; Kim et al., 2009; Zhang, 2011). We selected the observational data during 2005-2011 and averaged the data through the period for each site.

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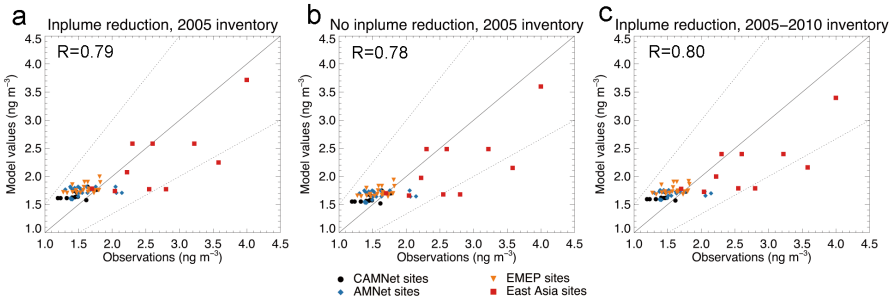

Fig. 1. Comparisons between modeled values and observations under three scenarios