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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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

ACPD
13, C13403–C13408, 2014

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. (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). 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. 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.

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. Section 2.2: As mentioned above the emissions database used by the authors is from AMAP/UNEP not GEIA. It was not valid for the later part of the period simulated by the model in this work. 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

ACPD
13, C13403–C13408, 2014

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.

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. 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.

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.?The period 2005 – 2011 is not representative of a decade as three different global emission databases are available for 200, 2005, and 2010. Section 3.1: the emission inventory is from AMAP/UNEP. 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. The interhemispheric gradient is described as stronger, stronger than what? 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). The rapid oxidation of Hg(0) by Br in polar regions depends on the season. Comparing regional model output which has a

ACPD
13, C13403–C13408, 2014

predominantly continental domain, with global model output is not valid. Section 3.2: The global budget is similar to that from previous studies using the same model. As would be expected, I imagine. 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. 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. Last line; sequestered? Buried with sediments in the deep ocean?

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. It also misses the point that deposition to oceans is of extreme importance. 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. 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.

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. 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. There were no experiments to assess the impact of increasing or decreasing emissions in any of the anthropogenic Hg source regions. This article lacks any significant new scientific contribution, and contains no results which are particularly new or interesting.

ACPD
13, C13403–C13408, 2014

It is not suitable for publication in ACP.

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