Geosci. Model Dev. Discuss., 7, C2674–C2682, 2014
www.geosci-model-dev-discuss.net/7/C2674/2014/

Geoscientific
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Interactive comment on "GNAQPMS-Hg v1.0, a global nested atmospheric mercury transport model: model description, evaluation and application to trans-boundary transport of Chinese anthropogenic emissions" *by* H. S. Chen et al.

Anonymous Referee #2

Received and published: 16 December 2014

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,

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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 don not think there is much reason to publish this article in GMD.

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 The GEOS-Chem reference is out of date there are a number of more up to date publications, with various improvements on Selin et al.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/longdistance sources of Hg to the US, perhaps it should be cited earlier.

2.2 Model Description

2.2.1 Mercury Chemistry

All the Hg(II) produced by the reactions between Hg and O_3 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 $PM_{2.5}$. 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.

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.

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.

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?

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.

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 5800 Mg 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.

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?

2.3.4 Oxidized mercury

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

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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).

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.

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.

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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.

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