

Interactive comment on “Multi-model study of mercury dispersion in the atmosphere: Atmospheric processes and model evaluation” by Oleg Travnikov et al.

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Received and published: 3 March 2017

The authors are thankful to the reviewer for the critical comments and suggestions. They resulted in significant improvement of the manuscript. Below we present point-by-point answers to particular comments (original comments of the reviewer are in italic font).

1) The main issue is that the 4 models used are completely different (see paragraph 2.2) in terms of spatial resolution (1 to 2.8 degrees in lat. and log.), upper boundary domain (from 30 km to 80 km), primary and secondary emission parametrization, chemical scheme utilized. Under this heterogeneity of the models it is hard to find a process or a chemical scheme that is better than the other or useful to explain or to

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better understand how mercury is dispersed in the atmosphere.

We cannot completely agree with this statement. The models used in the study are significantly different but not completely as it is mentioned. Indeed, in terms of spatial resolution they differ from 1 to 2.8 degrees. This is a typical range of spatial resolution for global scale models used for this purpose. Significantly higher resolution (down to few kilometres) is commonly applied in regional models, whereas much lower resolution (tens of lat/lon degrees) is characteristic of global compartmental or box models. It means that in spite of quantitative difference in resolution the models relate to the same class of chemical transport models and should provide comparable results.

The difference in the upper boundary height (from 30 km to 80 km) is also mostly quantitative. Computational domains of all the models cover the entire troposphere (10-15 km) and, at least, significant part of the stratosphere (up to 50 km). It is enough to take into account the majority of processes governing mercury dispersion in the atmosphere.

The models utilize exactly the same inventory of primary anthropogenic emissions with somewhat different emission speciation in the base case. The parameterisation of natural/secondary emissions does differ considerably among the models. However, in many cases it is a part of a model set-up reflecting mercury cycling between the surface and the atmosphere, which cannot be easily modified without disturbing the entire model formulation.

Probably, the largest difference among the models is associated with the chemical schemes applied in the BASE case. However, other numerical experiments of the study (Table 2) were formulated in such a way to study particular processes when all the models used the same (to the extent possible) chemical mechanisms.

The aim of the study was to reveal similarities in simulation results of the models taking into account the mentioned above differences. Therefore, the results when the models agree either in reproduction of observed dependencies (e.g. spatial or temporal

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variation) or in failing to reproduce them appear to be particularly valuable.

The discussion on the model differences has been appropriately extended in Section 2.2 of the updated manuscript (page 5, lines 8-25).

2) Page 9, lines 12-19. In describing the comparison of model simulations and observations reported in figure 2, authors report: “models predict similar spatial pattern of the observed Hg with pronounced gradient between Southern and Northern Hemisphere”. It is correct that qualitatively all the models reproduce the north to south gradient, but the absolute values are completely different from model to model and also the spatial distribution. The question is: Which one is better? Would be interesting to explore the reasons of these differences between models and give the reasons that can be useful to other modeler. Moreover this will help to understand the process and the chemistry important to predict mercury with models.

We agree with this critical comment. This part of the paper was completely rewritten including detailed analysis of the simulated patterns and comparison with observations (see the revised manuscript, Section 3.1, page 10, lines 9-35, page 11, lines 1-5). Besides, a new figure (Fig. S5) was added to the Supplement to support the analysis.

3) Page 9, lines 22-25: Authors report: “models demonstrate lower spatial variation of annual GEM concentrations than the measurements do. This can be partly explained by relatively low spatial resolution of the model grids (1-3 hundreds of kilometers) that can hardly allow them to reproduce local meteorological conditions at measurement sites”. First of all, if this poor spatial resolution of the model explain only partially the disagreement between model and observation spatial variation, explain the other reasons. On the other hand, from this sentence is evident that a model with high spatial resolution is required to explain the observations more quantitatively, otherwise with models we can have just a general picture.

This statement mostly relates to sites compactly located in North America and Europe. They are often affected by regional or local emission sources. Besides, even rural mea-

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surement sites in these regions are representative for limited surrounding area that can hardly exceed tens of kilometers in size. On the other hand, chemical transport models that applied on a global scale commonly relatively rough resolution (1-3 degrees). It is quite enough to characterise concentration levels over the globe and major physical and chemical processes governing dispersion in the environment. To characterize detailed concentration and deposition patterns over particular region or continent regional scale models are commonly applied. They have much finer spatial resolution (down to a few kilometers) but also require definition of boundary conditions.

Another reason of the lower variability can be associated with possible overestimation of GEM residence time in the atmosphere that leads to additional smoothing of the simulated concentration patterns. Current model estimates of the GEM residence time vary from 0.5 to 1 year. But recent studies show that it can be scaled down to a few months (Horowitz et al., 2017).

Appropriate changes has been done in the text of the revised manuscript (page 10, lines 1-8).

4) Page 10, lines 9-11: Authors report: “The models predict some decrease of concentration further northward, which is not evident from the observations. It can be connected with overestimation of the oxidation chemistry in the Arctic or with underestimation of Hg re-emission from snow and seawater”. This is really important to know: is it the oxidation scheme in the model or underestimation of the Hg re-emission from snow and seawater? Addressing this point is something that help to go further in our understand of mercury chemistry, and is mandatory to give some evidence of the process responsible for the model-measurement discrepancy, otherwise which is the added value of using 4 models?

It is mostly connected with underestimation of Hg re-emission. More detailed analysis of the modelling results in the polar regions and their comparison with measurements performed in the Arctic and Antarctica is given in the companion paper (Angot

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et al., 2016). In particular, it is shown that three of the four models successfully reproduced the stringtime minimum of GEM concentration caused by occurrence of the atmospheric mercury depletion events (AMDEs). However, the models tend to underestimate the summertime maximum that is determined by revolatilization from snow-pack/meltwaters and evasion from the ocean.

This clarification and the reference has been added to the text (page 11, lines 14-20).

5) Page 10, lines 33-34 and page 11, lines 1-2: Authors report: “Simulations with different chemical mechanisms (BrCHEM1, BrCHEM2, O3CHEM, OHCHEM) do not lead to considerable changes of both spatial distribution and temporal variation of GEM concentration in the surface air. Somewhat better spatial correlation was obtained for the oxidation reactions with Br (BrCHEM1) and OH radical (OHCHEM) and worse for the reaction with ozone (O3CHEM)”. This conclusion are a bit confusing: if as reported at the beginning, different chemical mechanisms do not influence the models performance, this means that chemistry is not an issue. On the other hand if is correct that somewhat better spatial correlation between measurements and simulations can be achieved with Br and OH oxidation reactions and worse results with O3 oxidation, then it is this the direction to explore: why Br and OH chemistry improve the performance of the models and O3 chemistry does not?

Agree. Use of different chemical mechanisms do provide different spatial patterns of GEM concentration. The difference is not too large in terms of the comparison with measurements but still requires consideration. We have revised this section including additional analysis of the influence of chemical mechanisms on simulated GEM patterns (page 12, lines 4-15 of the revised manuscript).

6) Page 13, lines 16-18: Author report: “The deviations between the modelled and measured RM/GEM are again mostly within a factor of 5 and the model-to-model difference is probably resulted from application of somewhat different reaction constants”. If the differences are probably due to different reaction constants used by the models,

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this is another variable to take into account and this further issue does not help to understand the problem. What happen if all the models use the same reactions constants? This test could be useful to exclude one of the uncertainties of the model simulations.

The model experiments were originally formulated to use the models in their standard configurations (including particular parameterizations and rate constants). However, we agree that the use of different reaction constants by the models introduces additional uncertainty to the analysis. Therefore, results of the mentioned experiment (OHCHEM) have been recalculated by one of the models (GLEMOS) with the common reaction rate to harmonize conditions of the experiment. The new results demonstrate much better agreement between the models (Fig. 7 of the revised manuscript). All figures containing the OHCHEM results have been updated accordingly.

7) Page 14, lines 2-8 and figure 8: *The simulations of RM/GEM is a disaster for the ECHMERIT model that show a maximum during summer with all the chemical schemes, whereas the observations show a maximum in March and a minimum during summer. Also the other models do not do a good job in reproducing the observation of RM/GEM, only GEOS-Chem is quit fine with the BRCHEM1 scheme. This is a big issue that need to be explored and the explanation can help to go further with our knowledge of the mercury chemistry.*

An additional analysis has been performed to address this issue. High levels of oxidized Hg in spring are evidently caused by high Br concentrations in both the free troposphere and the boundary layer in the Northern Hemisphere. Only GEOS-Chem reproduces correctly the time of the maximum in March. Two other models move the highest RM/GEM ratio to April and May. This discrepancy can be explained by partitioning the GEM oxidation products. GEOS-Chem is the only model that uses dynamic gas-particle partitioning of RM. This means that oxidized Hg originated from GEM oxidation is dynamically fractionated between GOM and PBM. This equilibrium is sensitive to air temperature shifting to GOM under warmer conditions. On the other hand, GOM

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is more efficiently removed from the atmosphere than PBM by both precipitation and surface uptake. This phenomenon leads to higher deposition and lower RM concentrations in late spring (April and May). Two other models predict a fixed share of the products of GEM oxidation by Br, thus missing changes in RM concentration during the season. The fourth model (ECHMERIT) predicts the highest ratios during late summer independently of the chemical mechanism used, a phenomenon that is determined by meteorological conditions and removal processes. In particular, the model simulates extremely low precipitation in late summer and early fall. This leads to significant underestimation of wet RM removal and ultimately to overestimation of oxidized Hg concentrations during this period.

Appropriate discussion has been added to the revised manuscript (page 15, lines 14-27).

8) Page 15, lines 7-12 and figure 10: It is not a good idea to exclude one model (ECHMERIT) results from the statistics only because excluding the results of this model improves the overall performance of the models simulation. This exclusion does not help to understand what is going on, and if one model is excluded only one time the results show in figure 10 are not comparable with those of the other figures since here the statistics includes 3 models and not 4. On the other hand, this exclusion of one model indirectly says that one model is worst the others: is this an indirect conclusion of Authors?

Agree. Results of the model have been returned back to Figs. 11-12 along with appropriate description (page 16, lines 29-34, page 17, lines 26-28, page 18, lines 7-10).

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