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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling of mercury with the Danish Eulerian Hemispheric Model" *by* J. H. Christensen et al.

J. H. Christensen et al.

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The authors would like to thank the reviewer for the very detailed and useful comments to the paper.

General Comments:

Reviewer: "The manuscript describes a mercury atmospheric transport model and some results obtained by the model. Significant part of the text is devoted to an attempt to simulate Arctic mercury depletion phenomenon, which can be responsible for elevated concentrations of mercury in Arctic ecosystems. Mapping of deposition fields over the Northern Hemisphere and particularly over the Arctic is extremely important for assessment of environmental pollution by persistent toxic pollutants like mercury. Creation of mercury monitoring network of more or less high density is absolutely unreal on global scale (or even in the Arctic). Hence, the only way to obtain the information is

usage of reliable mathematical models."

Answer: The authors agree.

Reviewer: "Unfortunately, a reader can get no idea about reliability of the model described."

Answer: The knowledge about mercury in the atmosphere is still very uncertain. During the last 3-4 years it has become possible to measure Reactive Gaseous Mercury (RGM) in the atmosphere, it is however still unknown what RGM exactly is. This implies also that knowledge about mercury chemistry is poor. Based on that, the model consists of two parts: a well documented and well performing transport model which has been validated for other species (see e.g. Barrie et al., 2001, and Christensen, 1997), together with what is available of suitable chemical mercury schemes. At the time of the model development, only one suitable chemical mercury scheme was available (Petersen et al, 1998). The focus of the paper is the Arctic, and especially the performance of the model in the Arctic. In the paper three figures presenting a comparison of calculated mercury concentrations and measurements are shown. The authors believe the important way to obtain more detailed knowledge about mercury in the atmosphere is to combine measurements and process studies with models like the one described in the present paper. This paper is the first of many steps, and more papers will follow with combined model results and measurement. The Barrie et al. reference is now added to the paper.

Reviewer: "The mercury model description is very curtailed. No attempts are made to explain choice of the modeling parameters. "

Answer: The transport model is described in detail in Christensen (1997) and the chemistry is described in detail in Petersen et al. (1998). There was no reason to repeat these descriptions.

Reviewer: "No information is given about verification of the mercury model."

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Answer: The focus of the paper is the Arctic, and especially the performance of the model in the Arctic. In the paper three figures for three different measurement stations are shown, where the calculated mercury concentrations are compared to measurements. This is a verification of the model.

Reviewer: "The paper focuses on the problem of mercury pollution in the Arctic. It would be better to reflect this interest to the Arctic in the paper title."

Answer: The authors agree with the reviewer: The title has been changed to: Modeling of Mercury in the Arctic with the Danish Eulerian Hemispheric Model.

Reviewer: "It is difficult to read the text. Only one example - let us look at the first three phrases of the second paragraph of the Introduction. 1. The Model (DEHM) is a well tested model. 2. The purpose of the model is 3. The model is based on the DEHM Can a reader understand what model is described?"

Answer: The authors have tried to improve the text in order to make it more easily read.

Specific comments:

Reviewer: "Page 3529, lines 12-14. It is possible to assume that the authors do not consider both re-emission and natural emission of mercury. For a hemispheric model the boundary conditions are realized on the Equator only (1.5 ng/m3). Atmospheric exchanges though the Equator is slow. Only anthropogenic emissions and weak atmospheric transport through the Equator can not provide maintenance of more or less stable mercury concentration on the level of 1.5 ng/m3 in the Northern Hemisphere. So, the model must reveal permanent long-term decline of the background mercury concentration."

Answer: The time scale of exchange between the two hemispheres is about 1 year. Because the boundaries in the model are located between 10N (in the corners) and 25N, the time scale is lower for the exchange between the boundaries and the model domain. The lifetime for elemental mercury in the gas phase reaction with ozone is

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approximately 2 years when using a hemispheric ozone mean concentration for the troposphere of 30ppb and taking into account the vertical variations of the density. There is therefore no long term decline of the background concentrations. Investigation of the calculated concentrations at e.g. Station Nord located in NE Greenland reveals a decline from May to September followed by an increase due to the enhanced transport from lower latitudes.

Reviewer: "Page 3529, lines 16-17. There is scientific information that mercury chemistry can involve radicals, chlorine and other reactants (e.g., Shia et all, 1999). What reason is to ignore such reactions? "

Answer: It is correct that there are other reactants participating in the Mercury chemistry. During the development of the mercury model, the chemical scheme obtained from Petersen et al., 1998 was applied, because it was at that time a state-of-the-art mercury chemical scheme. Currently a new scheme with a more comprehensive treatment of the chemical reactions is being implemented for the next generation of the model.

Reviewer: "Page 3529, line 19. As I understand the model starts to take into account the depletion (fast oxidation) at the moment of polar sunrise. What is the moment of polar sunrise? Appearance of a small piece of the sun over the horizon for few minutes after the polar night? Appearance of the full disk of the sun over the horizon? Sunrise is the Arctic is very slow process. By the way, ÿ polar sunrise or ÿ Polar Sunrise ? "

Answer: The additional fast oxidation rate is depending on the sun angle with a maximum rate at 0.25 1/hr, when the sun angle is higher than 20 degrees;. This information has been included in the new version of the paper. Polar Sunrise with capital letters have now been applied throughout the paper.

Reviewer: "Page 3529, lines 19-20. Fast oxidation of Hg(0) to HgO. Why HgO? Is there any theoretical or empirical evidence? Any reference?"

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Answer: It was the best guess when the model was originally developed. Currently the chemical reactions between elemental mercury and bromine are included, resulting in the end products HgBr2 and other HgBr species.

Reviewer: "Page 3529, line 22. The depletion can be realized only over sea ice. What information on sea ice was used in the model?"

Answer: Information about sea-ice is obtained from NCEP re-analysis. This information is now included in the paper.

Reviewer:" Page 3529, lines 21-22. Additional oxidation rate. Additional to what rate?"

Answer: The fast oxidation rate is added to the oxidation rate of elemental mercury due to the oxidation by ozone. This information is now included in the paper.

Reviewer:" Page 3529, line 22. Extremely important!!! What is a reason to choose the additional rate equal to 0.25 1/hr? Any theoretical consideration? Any reference? It would be possible to obtain much more intensive mercury deposition fluxes in the Arctic if one assumes the rate which is 10 times higher. The value used must be explained."

Answer: There is no theoretical consideration, because the rate is unknown. The value was based on a model fit only.

Reviewer: "Page 3529, lines 22. Temperature of depletion termination is assumed to be -4C. Why? Is it momentary value? Is it daily mean temperature? It is extremely important parameter, which determines the total value of mercury deposition over the Arctic. Any scientific support must be done for choice of this value."

Answer: The -4C is based on experience from Pt. Barrow (Steve Brooks, pers. com.). This information has now been included in the paper. It is not the daily temperature, but the instantaneous surface temperature as calculated by MM5.

Reviewer: "Page 3530, line 15. Effect of the depletion is very pronounced in the region of Hudson Bay. However, on the level of 60 degree latitude there is no Polar Night. So,

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there is no Polar Sunrise there. How could the model simulate the strong effect of the depletion here? At Barrow the model predict the beginning of the depletion by the end of February the beginning of March (Fig. 8). However, the sun appears on this latitude in the middle of January. Again, the question arises what does it mean Polar Sunrise ?"

Answer: Because the parameterization is dependent on sea-ice, surface temperature, sun-angle (and cloud cover) only, this phenomena will also be modeled for Hudson Bay. Observations of BrO from the GOME satellite (see http://www.doas-bremen.de/bro_from_gome.htm#Data) indicate that mercury depletion is in fact an observed phenomenon at Hudson Bay.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3525, 2003.