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Interactive comment on "ECHMERIT V1.0 – a new global fully coupled mercury-chemistry and transport model" *by* G. Jung et al.

Anonymous Referee #1

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The ECHMERIT model described by Jung et al. presents a new model with atmospheric chemistry embedded within a general circulation model (GCM). The authors highlight its application to mercury chemistry and transport, which is the key area in which they have extended beyond past work on coupled chemistry-climate simulations. The ECHMERIT model is surely a significant contribution to modeling science within the scope of GMD and I recommend publication with some revisions.

Overall, this is a strong paper, and very interesting to read. The prose and topic were sufficiently engaging that I found myself spending significant time reviewing this paper and therefore noticing aspects that could be improved. These are largely matters of clarifying the text. I look forward to reading a revised manuscript and future studies involving the ECHMERIT model.

MAJOR CONCERNS

I want to raise a few suggestions that I think would enhance the strength of this paper. The authors state at the beginning of section 4 that "complete model validation and statistical analysis" is beyond the scope of this paper. I understand that such an evaluation is a major task which is better suited to a second paper. However the authors do devote section 4 of this paper to model testing. Within the context of this brief evaluation, there are several simple figures and numbers which I believe would provide a stronger foundation for the credibility of the model.

- The authors spend some time evaluating the performance of the model's ozone simulation (sections 4.2 and 4.4). This is very useful as a foundation of their analysis of mercury cycling, since ozone is generally thought to be a major atmospheric oxidant for mercury. However the analysis is limited primarily to surface data from Europe (EMEP). The statistical measures of model bias (section 4.4) have very limited significance for a global model, since the authors do not evaluate their ozone simulation against any observations outside of Europe, nor do they use vertical profiles. Comparisons with seasonal means from a few ozone sondes, NOAA CMDL or WMO GAW locations would be very helpful.

- The authors do not describe or evaluate the OH concentrations within their model. OH is a important reactant for Hg, at least within the reaction set they have chosen, in addition to being a valuable check on plausibility of the model in general. At the very least the authors should state the global mean OH and associated methane lifetime within their model and compare it with the accepted range (c.f. Shindell et al. 2006).

- What are the model's mean concentrations of TGM in the northern and southern hemispheres? Figure 2 shows very little gradient between the northern and southern hemispheres, while observations suggest a value around 1.4 (Lindberg et al. 2007). The authors comment on the discrepancy on page 408 (lines 1-14) but do not give any quantitative comparison. Similarly, what is the atmospheric lifetime of TGM in the

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model and how does it compare with the literature cited in the introduction?

- What is the time step for operator splitting, chemistry, deposition, and emission? The authors refer to this time step on page 395 line 24. Separately, they also discuss operator splitting in section 2.7. If the photochemistry and transport time step is the same as the 2 h short wave radiation time step, (page 391 line 17), then this could be a major model deficiency. If it is much smaller, then this simply needs to be clarified in the text.

SECONDARY ISSUES

- What concentrations of ozone, OH and other constituents does the ECHMERIT model assume in the stratosphere? Apparently the model uses a climatology of ozone (page 405 lines 9-13), but the source and reference are not given. Does the model include cross-tropopause transport of ozone and other species? Oxidation of Hg(0) is fast in the upper troposphere and lower stratosphere (Lin et al. Atmos. Environ. 2006; Holmes et al. GRL 2006; Selin et al. 2007), so these assumptions will have a significant impact on the mercury simulation.

- Streets et al. (2009) released an inventory of mercury emissions which deserves some comment and comparison in the introduction to the Pirrone and Mason (2009) inventory that the authors have chosen.

- Does the model include ship, aircraft or lightning emissions? These will affect the ozone in remote regions and upper atmosphere (Eyring et al. ACP 2007; Wild ACP 2007).

- Second 2.4.3 requires some clarification. Nearly 90% of the wet deposited mercury is scavenged below precipitating clouds (page 410 line 9), but this wash out process is not described. Equations 5 and 6 appear to assume that wet deposition is generated from the model layers with the greatest cloud liquid water content, since they will have largest C_k (assuming relatively uniform Hg(II) distribution). It's unclear to me that this

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is consistent with the liquid water tendency in the GCM.

- The authors find that elemental mercury dry deposition is negligible (page 410 line 14). This conflicts with previous field studies (e.g. Lindberg et al. Atmos. Environ. 2002) and model studies (e.g. Selin et al. Glob. Biogeochem. Cycles 2008) which found dry deposition velocities around 0.1cm/s over vegetated surfaces. The discrepancy warrants some discussion.

- In section 6, the authors write, "modelling uncertainties due to interpolation of meteorological input variables on the CTM model grid and also uncertainties due to the lack of representation of high-frequency meteorology features are reduced" (page 412 line 5). I do not think this conclusion is adequately supported earlier in the paper. Perhaps some literature review of the error or bias introduced by CTMs could support this.

- I am somewhat confused by the paper's final paragraph. I'm not sure what the "former" and "latter" cases refer to. It is also unclear what aspects of regional models respond "linearly" to boundary conditions (line 26). More importantly, I disagree with the "abject lack of available measurement data" (line 21) since there are multiple published studies of long-range mercury transport, albeit not enough (e.g. Jaffe et al. Atmos. Environ. 2005; Swartzendruber et al. JGR 2006; Slemr et al. ACP 2009).

MINOR AND EDITORIAL ISSUES

- Figure 8: Ozone concentrations over Siberia and Arctic Canada appear to be < 10 ppb during DJF. Concentrations in this region during this season are typically 30-40 ppbv. (e.g. Simpson et al. ACP 2007). What explains the discrepancy?

- Page 406 line 10 says that the ozone dry deposition velocity "is further investigated". Perhaps the authors mean that it "will be further investigated", since this is the last mention of it in this paper.

- World maps in the figures have extensive lake boundaries that tend to obscure the colors and maps, making it difficult to read numerical values.

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- Figures 4 and 5 are presumably "dry" deposition velocity, but do not say this.

- A log scale would enhance the readability and usefulness of some figures (especially 14 and 15)

- Figure 16 does not have any labels on the y-axis and the categories on the x-axis are not explained.

- The writing in this paper is very clear and accurate. I noticed a few scattered typographical errors: "ECHMA" instead of "ECHAM", "arctic" instead of "Arctic" and "antarctic" instead of "Antarctic".

A few references which might not be clear from the context above: W. Simpson et al. Atmos. Chem. Phys., 7, 4375–4418, 2007

D. Shindell et al. JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 111, D19306, doi:10.1029/2006JD007100, 2006

D. Streets et al. Environ. Sci. Technol., 2009, 43 (8), 2983-2988. DOI: 10.1021/es802474j

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