Atmos. Chem. Phys. Discuss., 9, C7108–C7110, 2009
www.atmos-chem-phys-discuss.net/9/C7108/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating mercury emission outflow from East Asia using CMAQ-Hg" *by* C.-J. Lin et al.

Anonymous Referee #2

Received and published: 13 November 2009

General: This is a useful contribution to a very important subject and it merits publication, subject to revisions requested below. Specific: Three points need to be addressed before it is published. These are, in (roughly) increasing order of importance: 1) In the context of reducing global health hazards, one of the most useful results obtainable from this paper is the anthropogenic contribution to mercury outflow, because this can be changed, whereas the natural contribution cannot. The study by (Shetty, et al., 2008) on which the natural emissions in this work were based, concludes that in East Asia, on average, the natural emissions are comparable to (∼80 % of) the anthropogenic emissions and exceed the latter considerably in the summer a result that is reflected in Figure 6. The abstract and conclusions, however, discuss only the total outflow and deposition in the context of global values of these parameters, emphasizing the very large absolute amounts involved. I think it would be helpful to provide a

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

better breakdown of natural vs. anthropogenic outflow and deposition in these more prominent parts of the paper. 2) BC/IC: The method purportedly is independent of the BCs because it relies on a subtraction (equation 3) that has BCs in both sides of the equation. It is useful to compare the outflow calculated by this method (i.e. equation 3) with the net flux at the boundaries obtained from the GEOS/Chem BCs. Are these different? 3) It is curious that the model has such a large over-prediction of the PHg and RGM at Seoul and Cape Hedo, but at the same time gets the GEM in these locations right. The authors postulate that this might be caused by too much GEM leaving the mainland (and getting oxidized in transit) or too little deposition of the reactive species to the water. They also refer to the possibility of incorrect oxidation rates by NOx/VOC chemistry, which raises an important source of error: The emissions of criteria pollutants such as VOCs and NOx in this region are not well known, so the possibility is very real that the oxidation rate of GEM to RGM and PHg might be wrong. If the model over-predicts RGM and PHg at a remote site due to a oxidation rates that are too high because of poorly known emissions of VOCs and/or NOx, then the oxidation rates might be too high in the domain as well. This would lead to a high removal rate of GEM, which is consistent with the observation that nearly all mainland measurements are higher than the model predictions. This is a different explanation than the common one that the emissions must be diluted over the grid square. While the latter is certainly true, it is difficult to quantify and thus one should use it cautiously and not as a catch-all for model failures. Moreover, this point can be tested easily by comparing the ozone levels obtained from the model with measurements. Such a comparison is extremely useful as a reality check for any work that is sensitive to atmospheric oxidation, which is the case here. We read on page 21297 "In the absence of mercury emission input, the mercury mass entering the model domain from the boundaries is readily removed due to chemical oxidation of GEM followed by dry and wet deposition." This shows that oxidation in the domain is an important process and will influence the outflow significantly. Thus I agree with the last line in the paper: "Major uncertainties of this assessment include mercury chemical mechanisms and mercury speciation of

ACPD

9, C7108–C7110, 2009

the anthropogenic emission estimates. Further understanding of mercury chemistry and emission processes will greatly reduce the uncertainties." Since both the authors and the referee agree that this is a problem, I suggest that a comparison with ozone be included in the work to assess the importance to the result of oxidation due to lack of information about the NOx/VOC emissions. Technical: The grammar should be reviewed and improved. Also, there seems to be a confusion between "removal" and "export". On pg 21291, line 25: "A positive value of transport budget indicates a net removal of mercury mass in the domain (what's coming in is greater than what's going out); while a negative value indicates a net export of mercury from the domain. The mercury outflow caused (or enhanced) by the mercury". This is confusing. I guess it means a removal of mass from air that is coming into the domain, not a removal IN the domain? This occurs later as well.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 21285, 2009.