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Atmospheric Chemistry and Physics Discussions

Interactive comment on "GEM/POPs: a global 3-D dynamic model for semi-volatile persistent organic pollutants – Part 2: Global transports and budgets of PCBs" *by* P. Huang et al.

P. Huang et al.

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We would like to thank the anonymous reviewer for the detailed review of our manuscript which gives us the opportunity to clarify some points. In the following we quoted each review question in the square brackets and added our response after each paragraph.

Specific concerns

[As indicated above, I miss a critical comparison of predicted model outputs with related studies that have been carried out in the past. Specifically, it is my opinion that the paper has a limited recognition of valuable studies that could and should have been used to evaluate model predictions (beyond the model evaluation detailed in the first paper). The authors are thus kindly requested to discuss and evaluate their findings of the 2nd paper on the basis of the following studies: a) Global soil data: Meijer et al 2003 Environ Sci Technol 37: 667-672. b) Dry and wet deposition estimates to the global oceans: Jurado et al. 2004 Environ Sci Technol 38: 5505-5513 and Jurado et al 2005 Environ Sci Technol 39: 2426-2435, respectively.]

First of all, we have added a section in our paper 1 with scatter plots of model vs observations, which gave more quantitative evaluation of our model results. However, we value very much of the suggestions here for our paper 2 and have added following discussion in section 4.2 of this paper:

"Initial soil concentrations play a very important role in the atmospheric concentrations of PCBs. When the original initial soil concentrations of selected PCB congeners from the MSC-East model were used, the global air concentrations of PCBs produced by the GEM/POPs were too low to compare with observations. The global soil burden of PCBs was 16819 kg for PCB28 and 75857 kg for PCB180, which was about one order of magnitude less than the previous study (Meijer et al., 2003). After a 3D-VAR data assimilation scheme was applied to the initial soil concentrations by using the observational data from Meijer et al. (2003), the air concentrations of PCBs were improved greatly and comparable with observations (Gong et al., 2007). GEM/POPs estimated the zonal surface soil burden of PCB180 as 374594 kg between 30N-60N and 73924 kg between 60N-90N, agreeing reasonably well with the estimates by Meijer et al. (2003). Surface soil burden of PCB28 was overestimated 36%, but its distribution in the two zonal regions, 30N-60N and 60N-90N, matched the results of Meijer et al. (2003) very well. Due to the lack of observational soil data set in the zonal area between 90S-30N, no substantial improvements were made."

and in the section 4.3:

"The dynamic processes of emissions, transport and deposition determine the atmospheric loading of PCBs. Consequently, an accurate dry and wet deposition velocity

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and flux is needed to realistically predict the budgets of PCBs. Compared to other studies (e.g. Jurado et al. 2004), GEM/POPs captured the general pictures of global PCB depositions. For example, it is noticeable that the region E, i.e. Atlantic region, was a receptor and the deposition was an important process for the heavier PCB congeners in that region. Relative high dry deposition flux was found around 30 ON to 86 ON in the Atlantic area and the highest value was 500 pg/m2/day at 53 ON. This distribution was associated with the high air concentration in that area and consistent with the distribution and magnitude estimated by Jurado (2004). Wet deposition flux of PCB180 also presented a correlation to its air concentration: a peak value was predicted from 30 ON to 86ON. Particle scavenging dominated the wet removal process (Jurado 2005), which agrees with the prediction by GEM/POPs that 92% of the total deposition flux for PCB180 in the meridional region of Atlantic Ocean between1žE - 20žE was due to wet scavenging"

[The authors are furthermore encouraged to look out for additional studies that may provide additional information of relevance. For example, the model results suggest that re-emission from soils is a significant source of PCB-28 into the air on a global scale. However, there has been a debate in the scientific literature on the relative importance of primary and secondary sources in controlling contemporary atmospheric levels of PCBs for more than a decade with obvious implications for control strategies (see e.g. Harrad et al 2004 Env Pollut 85:131-146, Jaward et al. 2004 Environ Sci Technol 38:34-41; Hung et al. 2005 Atmos Environ 39:6502-6512).]

Some statements and citations about this issue have been added in this paper. Following paragraph will further address the re-emission issues.

[According to Meijer et al 2003, the global surface soil burden of PCB-28 is estimated to be 190 tonnes, whereas the GEM/POPs model predicts that the annual net (re-)emission of PCB-28 from soil should be approximately 41 tonnes (Table 1). Thus, please elaborate and discuss potential implications of the model outputs in the context of other studies with respect to this specific example and beyond.]

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We have added following discussion in section 4.2 to address the re-emission issues:

The global soil burden of PCB28 from GEM/POPs was about 250 tonnes and the annual cumulative emission from soil was 41 tonnes. About 16% of soil burden of PCB28 re-volatilized to the atmosphere, which is almost close to the magnitude of anthropogenic emission of PCB28 in year 2000 (Table 1). Other modeling studies (e.g. Malanichev et al 2004) have also discussed about the relative importance of re-emission vs. primary emissions of PCBs. This modeling result indicates that emission of lighter PCBs (e.g. PCB28) are more probable to be dominated by re-emissions rather than primary emissions. For heaver congeners, PCB153 and PCB 180, although their global soil burden calculated by GEM/POPs were 573 tonnes and 457 tonnes, their net soil-atmosphere exchange in the year 2000 were 6.5 tonnes and 0.9 tonnes toward to the soil compartment, respectively. This is consistent with a study by Hung et al (2005) who concluded that the primary emissions of heaver congeners (e.g. PCB153 and PCB180) were still the dominant source to the atmosphere (Hung et al. 2005) in the year 2000.

[Given the high temporal and spatial resolution of model outputs, I think it would be nice if the authors could offer some brief thoughts regarding current and future air monitoring strategies. For example, current air monitoring strategies under the Stockholm Convention of POPs seems to advocate for the use of passive air sampling devices (which captures mainly the gaseous fraction). Indeed, studies aiming to support the work under the convention have already been published (e.g. Pozo et al. 2006 Environ Sci Technol 40: 4867-4873). Relevant questions to be discussed in the context of critical knowledge/monitoring gaps could be: Is it feasible to use data from GAPS and related efforts to evaluate predicted spatial patterns? Will a passive air sampling approach be sufficient to evaluate model predictions, or is there a need for alternative monitoring strategies or targeted sampling campaigns? Are there regions for which there may be specific needs for further measurements (given the model outputs)?]

The comparisons between modeling results and the passive sampling results of global

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POPs are emerging right now. Initial data set of passive POPs measurements has been used to evaluate some modeling results (Shen et al., 2006). However, due the nature of the passive sampling, longer sampling time is needed than the active samplers and consequently temporal resolution of the data is rather coarse, i.e. one data per 2 to 7 months. Since a passive net work provides far more special coverage than the active networks, the results from the passive network should be very useful to evaluate the long-term spatial distributions of various POPs. However, how to combine the active and passive sampling methods to validate modeling results still needs more study, which is outside the scope of current paper.

Minor issues

[Page 3838, line 25: What is meant by “insoluble”?]

This word is deleted.

[Page 3839, line 7: It is somehow misleading to cite an old paper dealing with HCHs, when PCBs have been studied as well using global non-steady state multimedia fate models. See Wania and Daly, 2002 Atmos Environ 36:5581-5593; Wania and Su YS, 2004. Ambio 33: 161-168. Macleod et al 2005. Environ Sci Technol 39: 6749-6756 and Hung et al. 2005 Atmos Environ 39:6502-6512.]

Citations about PCB studies have been added in this paper.

[Page 3840, line 4: A verb is missing at end of line. This paper is (devoted?) to ˇ E?]

Typos have been corrected.

[Page 3840, line 6: Please be more explicit or rephrase what is meant by “current PCBs”?]

The word “current” is deleted

[Page 3844, line 8: Please add latitude of Arctic Circle at first mention and not on line

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19. Besides, I thought the Arctic Circle was found at 66.33 N and not 66.50 N?]

We added the latitude of Arctic Circle with 66.5žN in section 3.2 and deleted “66.50žN” on page 3844. The southern limit of the arctic region is commonly placed at the Arctic Circle (latitude 66 degrees, 32 minutes North), that is about at 66.5 žN (not 66.50žN).

[Page 3846, line 7: favourable conditions (not favourite)?]

Typos have been corrected.

[Page 3849, lines 1-10 and last sentence of abstract: The finding that long-range transport of PCB-28 is limited by OH-radical degradation and heavier PCBs by atmospheric deposition has been recognised previously (e.g. Wania and Daly, 2002 Atmos Environ 36:5581-5593).]

We have deleted the last sentence of abstract, added some statements and citations in this part:

Those quantitative results calculated by GEM/POPs’ are in agreement with the findings presented in previous studies (Wania and Daly, 2002).

[Page 3849, lines 12-13: Please rephrase sentence starting with “The area ˇ E” (to many “as”). I assume it would be OK if “as” is deleted in front of “half” and “twice”.]

Typos have been corrected.

[Page 3849, line 20: What is meant by “More features”?]

These two words are rephrased as: More processes of PCBs in environment.

[Reference list: Ref Gong et al. JGR is given with year 4007.]

Typos have been corrected.

[Figure 3: Inner figure presents PCB-28 as PCB028.]

It is changed as “PCB28”

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