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Fractionation and current time trends of PCB congeners: Evolvement of distributions 1950-2010 studied using a global atmosphere-ocean general circulation model

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Review

The paper addresses relevant scientific questions within the scope of ACP. The fractionation of 4 PCB congeners is investigated with the multicompartment chemistry transport model (MPI-MCTM), which is based on the coupled general ocean-atmosphere general circulation model (ECHAM5-MPIOM), and has embedded sub-dynamic models for atmospheric aerosols (HAM), marine biogeochemistry (HAMOCC5), and air-surface exchange processes with soils, vegetation and the cryosphere.

The approach and the results are novel in that fractionation of PCBs (and POPs) is modeled with a global 3-D model, which allows for variability in both the longitudinal and latitudinal directions. Previous models have been averaged in the longitudinal direction and fractionation in the latitudinal direction only has been investigated, see, e.g. Scheringer (2008), Gouin and Wania (2007).

The authors present some important and interesting conclusions that result from using their 3D model.

The study is very interesting and has important conclusions. In principle, the article is fit for publication in ACP. However, the language and the grammar need to be substantially improved throughout the manuscript, and the quality of the graphics is fair at best. Therefore, I have to say that the manuscript requires a major revision before it can be published in ACP.

I would be happy to accept the manuscript for publication once the authors have addressed these issues.

Please see some specific comments below.

1. Methods
    a. The modeling approach is sound and the authors appear to have a very good handle on the numerical/scientific system.
    b. Page 6, line 9: A model validation is very quickly presented in the supplementary material. Model results are generally an order of magnitude greater than observations. Please state briefly in the text what makes the model results reasonable.

2. Results and Discussion
   The results are very interesting and are well presented and discussed.
   Just a few points:
   a. Page 11708, line 7: The northward shift of the COG …. Etc. Please expand on this statement.
   b. Page 11709, 2nd sentence: 94-98% of ……. Please provide a figure describing this
   c. Page 11709, 2nd paragraph: please expand further on why your ACP predictions can be expected to be an order of magnitude greater than those of Wania (2006).
   d. 3.3 Page 11710-11714: The results are interesting but the results can read more like a reporting of numbers. Try to split it into more paragraphs and improve language and grammar, to read more easily, and make it flow more easily for you and the reader.
3. Conclusions
   4.1., 2nd paragraph: There is no mention of ecosystems in the results and discussion (sedimentation is briefly mentioned). Please address this.
4. The abstract is OK but the language is vague.
5. The language needs to be improved somewhat throughout the manuscript. The grammar can be poor. Some sentences are poorly written, and many sentences are not sentences at all. For example:
   a. Page 11699, beginning line 8: sentences beginning on lines 11, 12 and 13 start with They, their and it, respectively. This language gets vaguer with every sentence.
   b. Page 11700, beginning line 11. This is not a sentence. Perhaps something is missing after ‘transport’
   c. The passive form of verbs is often used when active form is correct, e.g. PCBs are undergoing …. Should be … PCBs undergo; … are migrating southward …. Should be … migrate southward …. This reappears throughout the text, please change.
   d. Page 11702, lines 13-16. This sentence is not clear.
   e. End of page 11702, line 27: this sentence is not clear. Do you mean …. of PCBs on their global …
   f. Page 11703, lines 9-11. Please rewrite this sentence.
   g. Page 11704, lines 5-6 … the one of ….. this appears throughout the text. Replace with … that of …..
   h. Page 11706, lines 3-8: please rewrite this sentence.
   i. Be careful with articles (the, a); sometimes they appear when they shouldn’t be there and vice versa.

j. Page 11707: replace ‘In consequence’ with ‘Consequently’; rewrite lines 15-20.
k. Page 11708, line 7. Please change the language ‘goes in line ’ to , for example, ‘is consistent with a decrease in the …’
l. Page 11708, line 21. Please change the word ‘migrate’ to something like ‘advection’/’transport’/’flux’/’shift’
m. Page 11709, line 19: ‘The metrics are not ….’
n. Page 11709, line 26: Please rewrite this sentence: ‘The mean number ….’ – bad grammar after the equation.
o. Page 11714, line 13, new paragraph: ‘The model suggests …’ , no, ‘The model results suggest …….’
p. Page 11714: change the language of ‘the shares of PCBs …’ to something like ‘The fraction of the various PCBs contained in the total mass’

These problems persist throughout the text.

6. The figures are of poor quality and need to be improved. Specifically,
   a. Fig 2 is illegible; I cannot read the legend and I cannot tell one line from another.
   b. Titles, labels and axes in Fig 3 are illegible. Please put the figure over two pages. The blue lines are barely legible. In Fig 3b, I would like to see an extra set of black/grey figures showing the fraction of each of the PCBs in ocean, soil, vegetation, ice and snow (Perhaps you could colour it and divide each of the colours, one per PCB, into different shades for each of the compartments). Fig 3 could be also displayed over 2 pages.
   c. Fig 4 is barely legible, see b. above.
   d. Please be more careful and complete with figure captions. They are vague ans sometimes poorly worded.

   I have also suggested the authors provide a few additional figures, since there are now only 4.

7. The reference list is excellent.