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Interactive comment on “PCBs in the Arctic atmosphere: determining important driving forces using a global atmospheric transport model” by C. L. Friedman et al.

Anonymous Referee #2

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General comments

Friedman and Selin have made a model study using an adjusted version of the global atmospheric transport model GEOS-CHEM to study the most important drivers of Arctic atmospheric concentrations of seven PCB congeners. The authors describe the new model version with a comprehensive and thorough supplementary material, although three references are not cited in the main manuscript. The model set-up is well described and the scientific methods and assumptions are valid and clearly outlined. The results are generally sufficient to support the interpretations, although some of them could be discussed in more detail. Substantial conclusions are reached and the

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study is in general transparent and traceable. The title clearly reflects the contents of the paper and the abstract provides a concise and complete summary. The manuscript is well structured and clear and the language fluent and precise. Proper credit is given to related work.

A similarly thorough model sensitivity analysis of the most important processes determining the atmospheric transport of PCBs to the Arctic has not been published previously, and the manuscript provides valuable new insight within the field. I think this will be of great interest to the readers of Atmospheric Chemistry and Physics and I suggest to publish the manuscript subject to minor revisions. I have added a few specific comments below that the authors should take into account in their revised manuscript.

Specific comments

Partitioning to particles

Page 30862, line 17-18: “there is no such conclusive evidence that PCBs adsorb more strongly to BC than they do to OM”. Has this been investigated and can the authors supply references for this claim? If not, please discuss why you expect PCBs to behave differently to PAHs with respect to adsorption to BC.

This is mentioned again on page 20870, line 21.

I think there may be a general problem with the parametrization of the gas-particle partitioning in the model, where the fraction sorbed to particles appears to be quite low (0.02%-1.4%). Although the particle fraction of PCBs has not been reported in any model studies, this has been studied in several measurement experiments, e.g. Simcik et al., ES&T, 32, 1998 reported particle fractions of 0.2%-0.7% for CB 28 and 5%-10% for CB 180 in July for Lake Michigan, depending on the wind direction, 1.1%-1.4% and 16.2%-21.2% for the two congeners in July and 2.9% and 49.4% in January in Chicago. Lohmann et al., ES&T, 34, 2000, reported PCB particle concentrations up to 70% for an urban site in the UK during winter, and in a more recent study Li et al., Aerosol and

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Air Quality Research, 15, 2015 reported 6-44% in Tianjin, China. The higher fractions for the urban site can be due to larger particle concentrations in the urban area, which is not reflected in the GEOM-CHEM model due to the spatial resolution, but even at the remote sites the measured fractions are well above the modelled fractions in this study. A larger particle phase fraction will also influence the loss from oxidation due to the neglected particle phase oxidation (the discussion on page 30874, second paragraph). Please comment on this, and if possible, introduce a sensitivity study of the effect of gas-particle partitioning in the model.

Other specific comments

Page 30868, second paragraph/Table 3. I think the statistics should be revised in this section. What is of interest in a model evaluation is how well the predicted concentrations fit to the measured concentrations on a 1:1 line. Instead the authors show the best possible linear fit, which is of no use in the interpretation of the results unless discussed further. This results in larger correlation coefficients than with a 1:1 fit. I therefore suggest that the authors replace the calculated correlation coefficients with a fit to the 1:1 line and also include a proper calculation of the bias and/or the mean error. This will improve the interpretation of how well the model predicts atmospheric concentrations. Please also specify if this is seasonal mean or monthly mean.

Page 30869, line 24 and Page 30870, line 1: It is not quite clear to the reader that the “temporal average” in this case refers to the “monthly concentrations averaged over time” like in the previous paragraph. Please specify this.

Figure 3 & 4. It is interesting to note that there is a clear decreasing trend in the simulated monthly concentrations of CB 28 at both sites that appear to be declining more rapidly than the observations. The results for CB 153 (Figure 4) show the same pattern although as clear. Please comment on this (something with the emission input?).

Table 2. Another interesting result that is not discussed is the similar Sim/Obs ratios for the Arctic and the NUML sites as well as for the Antarctic site, with higher simulated

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mean concentrations for CB 28 and 118 and lower for the other congeners. This could also be related to the emission input. Please comment on this. There are 3 references in the supplementary material that are not cited in the main manuscript (Gouin et al., 2013; Li et al., 2003; Mackay et al, 2006). Proper credit should be given to these studies.

Technical corrections

Page 30868, line 18: Please insert “seasonal” into this sentence: “. . .the model predicts seasonal observed concentrations. . .”

Page 30869, line 22: Please change “minimum” to “maximum” and vice versa.

Table 2, line 3: Please insert (NUML): “. . .and non-urban mid-latitude (NUML) sites. . .”

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 30857, 2015.

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