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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source regions of some Persistent Organic Pollutants measured in the atmosphere at Birkenes, Norway" *by* S. Eckhardt et al.

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We thank reviewer 1 for her/his comments on our manuscript. In the following, we repeat the reviewer's comments in italic font and respond point-by-point.

My most fundamental problem with the paper is the author's assumption that emission fluxes in each grid cell are linearly dependent on temperature (Equation 5). Presumably, emissions vary with temperature because of variability in the vapor pressure of PCB-28 and γ-HCH. And, the ideal gas law dictates that the logarithm of vapor pressure varies in direct proportion to 1/T, with the proportionality constant being the heat of vaporization. Therefore, in assuming that emissions vary linearly with temperature,

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Interactive Discussion

Discussion Paper

the authors have failed Environmental Chemistry 101!

The aim of introducing a linear dependence of emissions on air temperature is mainly to get a "reasonable" seasonal variation of the emissions because only annual emissions are available. In fact, we did not apply any temperature-dependent correction of the emission fluxes for γ-HCH because we had monthly emission data available for that species. Now, even for PCB-28, the primary emissions do not only depend on air temperature but also on many other factors. For instance, significant atmospheric emissions are expected to originate from indoor environments. If we assume indoor temperatures to be constant throughout the year and a constant "pool" of PCBs within buildings, then primary atmospheric emissions to the outdoor environment are expected to be a function of building ventilation rates. Building ventilation rates do not necessarily follow the ideal gas law, but are nevertheless likely to be affected by ambient air temperature because people will, for instance, open windows on hot days. Furthermore, significant PCB emissions may occur when waste containing PCBs is subject to combustion. Some of these combustion sources, like backyard barrel burning, are likely to have a seasonal component. Even for other outdoor emissions, the emissions will not follow directly grid-cell average surface air temperatures because different surfaces (soils, buildings, etc.) will heat up differently during daytime and their temperatures will not follow air temperatures closely. Since it is impossible for us to develop physically-based temperature relationships for this myriad of different sources, we sought a simple relationship, which should give higher emissions in summer than in winter. We also intended to keep the relationship between emissions and temperature conservative, i.e., to rather underestimate the temperature dependence of the emissions than to overestimate it, in a situation where little is known about the true temperature (or seasonal) dependence. A linear functional relationship is very appropriate to obtain such a conservative estimate. Finally, we also need to consider that even annual emissions are uncertain by an order of magnitude and even the best characterization of temperature dependence will not yield more accurate total emissions. This also calls for a rather simple approach to address the seasonal variability of emis-

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sions. In the revised version of the paper, we describe the motivation for the choice of the temperature relationship of the emission fluxes.

A second problem with the paper is that the work is interpreted within only a very narrow review of other research that addresses similar issues. The most glaring omission, in my opinion, is the lack of references and discussion of the Potential Source Contribution Function (PSCF) modeling work that has been done to interpret long-term monitoring data from the IADN sites in the Great Lakes. One reference to this work is: Hafner, W.D., and Hites, R.A. Environmental Science and Technology, 2003, 37, 3764-3773. Like this approach, PSCF modeling also combines long-term monitoring data with a trajectory model run in reverse to estimate the location of source regions. I think it is necessary for the authors to acknowledge this previous work and state differences of their approach. I suspect that the PSCF approach is exactly what the authors of this paper have in mind in their outlook in the last sentence of the paper prior to the acknowledgements.Page 3 (last paragraph): In this paragraph the authors use a very narrow survery of the literature to justify their modeling approach. Their approach is novel and interesting, but there are other tools available that are capable of something similar. For example, multimedia box models have evolved a long way since MacKay, 2001, and there are now models that consider meteorological data in their parameterization, most notably the BETR-Global model (MacLeod et al. Environmental Science and Technology, 2005, 39(17): 6749-6756. This study is novel and interesting because of the close marriage of modeling and monitoring data, not because the model is somehow superior to other models.

Many different model- and measurement-based methods have been used in the past to study the source-receptor-relationships of POPs. Since our paper is not a review, it is very difficult to give a fully comprehensive overview of all this past work. Our paper already contains more than 70 references, most of them regarding work done by others. However, we are grateful for the additional (and very appropriate!) references provided by the reviewer. These papers are now cited in the revised version of the

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paper.

Page 3, "Depending on temperature and the concentrations..." The "potential" to undergo reversible exchange is not a function of temperature or concentration.

We agree that this wording is unfortunate. Instead of “Depending on temperature and the concentrations in the various media, some POPs have the potential to undergo reversible exchange between terrestrial or aquatic surfaces and the atmosphere.” the sentence in the revised version of the paper now reads simply: “Some POPs have the potential to undergo reversible exchange between terrestrial or aquatic surfaces and the atmosphere.”

Page 5: Here, the ES and EC concepts are introduced. These are important for interpreting the results that are presented, and I would like to see this section revised to try to communicate these concepts more clearly. Again, I believe there a precident in the literature for these ideas that is not cited in the manuscript. Don Macky (Environmental Pollution, 2008, 156, 1196 - 1203 1182 - 1189) has introduced the concept of "distant residence time" as the mass of contaminant in a distant region divided by the emission rate. I believe this is the same concept as the "ES" that is introduced here, and at least a reference is warranted in the paper.

Notice that we do not really introduce these concepts. They are in broad use in other areas of air pollution research. For instance, adjoint chemistry transport models are nowadays often used to improve emission estimates. Furthermore, we have used the same model concept for establishing source-receptor relationships for carbon monoxide or aerosols and references to this past work are given in the paper. Examples are Stohl et al. (2003) and Seibert and Frank (2004), where also a complete description of the theory is given. Still, since this is the first application to POPs, we have slightly extended the description of ES and EC in the revised version of the paper. We now also cite the paper by Mackay, which is indeed somewhat related.

Mackay, D., and Reid, L.: Local and distant residence times of contaminants in multi-

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compartment models. Part I: A review of the theoretical basis, Environmental Pollution, 156, 1196-1203, 10.1016/j.envpol.2008.04.012, 2008.

Seibert, P., and Frank, A.: Source-receptor matrix calculation with a Lagrangian particle dispersion model in backward mode, Atmospheric Chemistry and Physics, 4, 51-63, 2004.

Stohl, A., Forster, C., Eckhardt, S., Spichtinger, N., Huntrieser, H., Heland, J., Schlager, H., Wilhelm, S., Arnold, F., and Cooper, O.: A backward modeling study of intercontinental pollution transport using aircraft measurements, Journal of Geophysical Research-Atmospheres, 108, 4370 10.1029/2002jd002862, 2003.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12345, 2009.