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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 2 for the valuable comments on our manuscript. In the following we repeat the remarks (or a summary thereof) of the reviewer in italic font and add our comments:

In Abstract and Introduction the authors mentioned that FLEXPART was ran in a timereversed (adjoint) mode to identify the source regions responsible for the chemicals loading at measurement site. It is interesting to know how this adjoint mode works. As a Lagrangian model the FLEXPART can run in backward trajectory for detecting source – receptor relationships. In adjoint mode comprehensive measurement data are needed

to modify predicted concentrations. For toxic chemicals, I am afraid those measurement data are not always available. Or I may misunderstand the notation of "adjoint" in this paper. Is adjoint mode used in this study similar to what has been used in Eulerian models?

The backward mode is described in detail in several papers, for instance Stohl et al. (2003) and, in particular, Seibert and Frank (2004). We have extended the description of the backward mode somewhat, following also the comments of reviewer 1. However, it is out of the scope of this paper to present the theory (again) in detail. The calculation of emission sensitivities and emission contributions is independent of the measurement data. Only if the model results were used for inverse modeling of the emissions, measurement data would be needed. However, this was not done in the present paper, with the exception of a simple scaling of regional contributions with the ratio of the measured to modeled concentrations in Figure 12, which can be seen as the most simple inverse modeling possible. We agree that currently there are not enough measurement data available for a full inverse modeling exercise but hope that this will be possible in the near future, as our institute is planning to perform POP measurements at more stations.

It will be helpful to mark Birkenes site on, for example, Fig. 1.

We think Figure 1 is too small to show the location of the station. However, we have already marked it in Figure 12 which shows a map of Europe and we will point the reader to this figure.

Pg. 12352, line 10. The authors estimate "dry gaseous deposition". It is interesting to know how the bulk surface resistance rc is defined in the paper, or at least a reference is perhaps necessary. For dry gaseous deposition, the surface resistance is likely the dominant resistance and is most difficult to be parameterized. Wesely's model appeared applicable in trace gases. How is it extended to POPs? How is rc parameterized for PCB28 and γ-HCH?

Depending on meteorological conditions and the magnitude of resistances r_a and r_b, r_c can indeed sometimes be the dominant resistance. According to Wesely's model, the surface resistance r_c depends on the landuse type and on the physico-chemical properties of the substances considered, i.e., the water solubility, reactivity relative to ozone and diffusivity. Diffusivity can be directly related to the stomatal resistance in a plant and depends on the molar weight of a substance, so it can be directly calculated. See our comment to the next question for details on the diffusivity values used. The water solubility in Wesely's model is calculated using Henry's law constant, which is known for both PCB-28 and γ-HCH. We have chosen values of 33.1 and 0.24 Pa m^3 mol^{-1} for PCB-28 and γ-HCH (references are given in the manuscript), respectively. Regarding the reactivity relative to ozone, we have assumed that both have a low reactivity (0.1). With this information, the Wesely (1989) model can be used also for PCB-28 and γ-HCH. In an unpublished study we compared the deposition velocities for PCB-28 calculated by FLEXPART using the Wesely (1989) method with values in the literature and found good agreement. Earlier, the Wesely method used also in other studies for simulation PCB transport and deposition (e.g. Semeena and Lammel, 2005; Su and Wania, 2005)

Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical-models, Atmospheric Environment, 23, 1293-1304, 1989.

Semeena, V. S., and Lammel, G.: The significance of the grasshopper effect on the atmospheric distribution of persistent organic substances, Geophysical Research Letters, 32, L0780410.1029/2004gl022229, 2005.

Su, Y. S., and Wania, F.: Does the forest filter effect prevent semivolatile organic compounds from reaching the Arctic?, Environmental Science and Technology, 39, 7185-7193, 10.1021/es0481979, 2005.

Pg 12352, line 13 (right after eq. 1), is same diffusivity applied for PCB28 and γ-HCH?

Yes, we applied the same values for the diffusivity of PCB-28 and γ-HCH, as was done

in Cousins et al. (1999). Alternatively, the diffusivity can also be calculated from the ratio of the diffusivity of water and the substance considered, which is related to the square root of the ratio of the respective molecular weights. The values derived by this latter method would have given nearly identical values as the molecular weight of PCB-28 and γ-HCH are very similar (258 and 291).

Cousins, I. T., Mackay, D., and Jones, K. C.: Measuring and modelling the vertical distribution of semivolatile organic compounds in soils. II: Model development, Chemosphere, 39, 2519-2534, 1999.

Figure 1. High γ-HCH emission in Canadian Prairies in January is apparently unrealistic. There was neither application nor reemission for that period in that region.

For the γ-HCH emission inventory, it was assumed that no fresh use of lindane occurred in the year 2005 worldwide, including Canadian Prairies. According to our emission model (Li et al., 2008), however, the emission did happen due to the historically heavy application of this pesticide in the region.

Li, Y. F., Ren, N. Q., and Tian, C.: China-North America project on reduction of lindane usage in china and its impact globally and on North America, in, Report to North America Commission for Environmental Cooperation (CEC), Environment Canada and US EPA, 2008.

Figure 2. Usually, January is the coldest month in the Northern Hemisphere. Fig. 2 shows higher γ-HCH emission in January, even higher than April. Does this higher emission in January suggest an anomalous higher monthly air temperature?

The monthly emissions of γ-HCH were calculated from its soil concentrations by using our emission model (Li et al.,2008) In this model, the emissions of γ-HCH depend on many factors, including temperature and the amount of soil residues of this pesticide. The results show that, for the entire northern hemisphere, the emissions in January were lower than the emissions in the following months (see Figure 1). However, for

Europe the January emissions were higher than the emissions in February, March, and April in Europe (see Figure 2), mainly because temperatures in Europe were slightly higher in January 2005 than in the following months (see Figure 3). We have added this explanation also in the revised version of the paper. One other factor is that the emissions also reduce the amount of soil residues, so the residues in soil are reduced every month and, given otherwise identical environmental conditions, emissions would decrease from one month to the next.

Li, Y. F., Ren, N. Q., and Tian, C.: China-North America project on reduction of lindane usage in china and its impact globally and on North America, in, Report to North America Commission for Environmental Cooperation (CEC), Environment Canada and US EPA, 2008.

Pg. 12357, line 4, "positive temperature anomalies are correlated with transport: : :" It is not clear how the authors defined "positive temperature anomalies". Temperature anomalies usually indicate the departure of temperature from its means.

Indeed, this is also our definition of the anomaly. What we mean is that when transport is from the south, there will usually be a positive temperature anomaly (warm air advection) but at the same time the air also tends to pass over high-emission areas, which are mostly located in Central Europe, to the south of the station.

Figure 9. Authors should indicate different scales are used in x- and y-axis of Fig. 9.

In the revised paper version, we have added in the Figure caption: Notice that the scale on the ordinate extends to ten times larger values than the scale on the abscissa.

Pg. 12361, line 6-8, "The decrease with distance is stronger for summer than for winter, due to both slower transport and : : :" Given that ES and EC maps shown in Figs. 10 and 11 are seasonal averaged, even "slower transport" could travel a long distance in air. During the summertime, atmospheric long waves in the wintertime break down to short waves and local atmosphere circulations dominate atmospheric transport. This

renders long-range transport seldom occurring in summer.

We generally agree with the reviewer but we do not fully understand this comment. Certainly, also "slower" transport can lead to transport over long distances on the time scales considered but on average these distances will be shorter than in winter. The reviewer has given a good meteorological explanation why this occurs but we don't think this is the topic of this paper.

Figure 12. Authors should put a color bar in Fig. 12 showing contribution percentage and label a- and y-axis (lat/lon) of upper panel of Fig. 12.

In the revised paper version, we have added a label to the axis in the first panel. This panel does not show the percentage of the contribution but simply the color code use for the various regions. We have modified the figure by adding labels and have extended the figure caption to make this clear.

The figure caption reads now as following:

Panel a) shows the different regions for which relative source contributions for the Birkenes station (marked with a white circle) have been calculated (1, Western Europe . . ..). Panels b) and c) show the regional source contributions (in percent of the total) for simulated PCB-28 and γ-HCH, respectively, with the colors indicating the contributions from the different regions as colored in panel a). The grey bars show the contributions when they are weighted by the ratio of the measured/modeled concentrations.

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

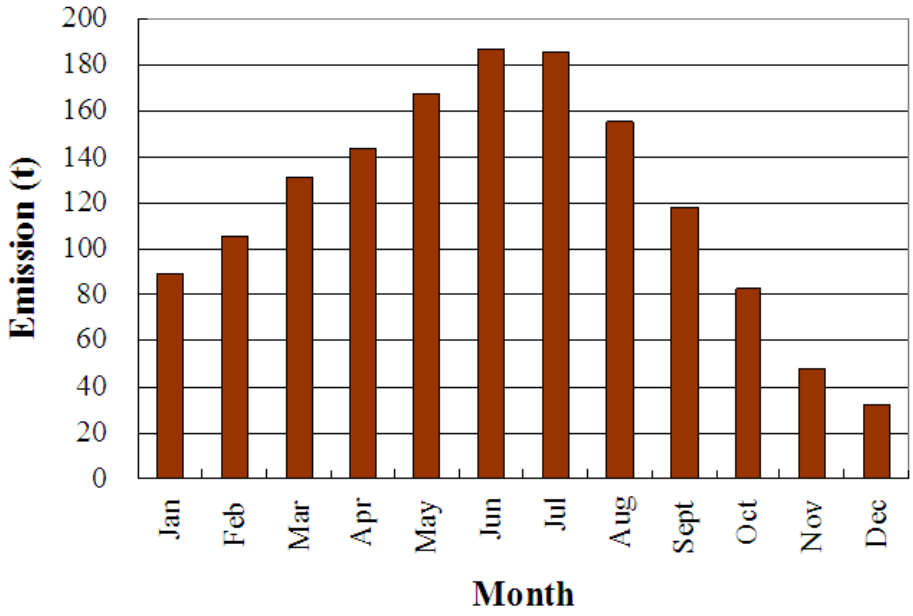

Fig. 1. Monthly emission of gamma-HCH for 2005.

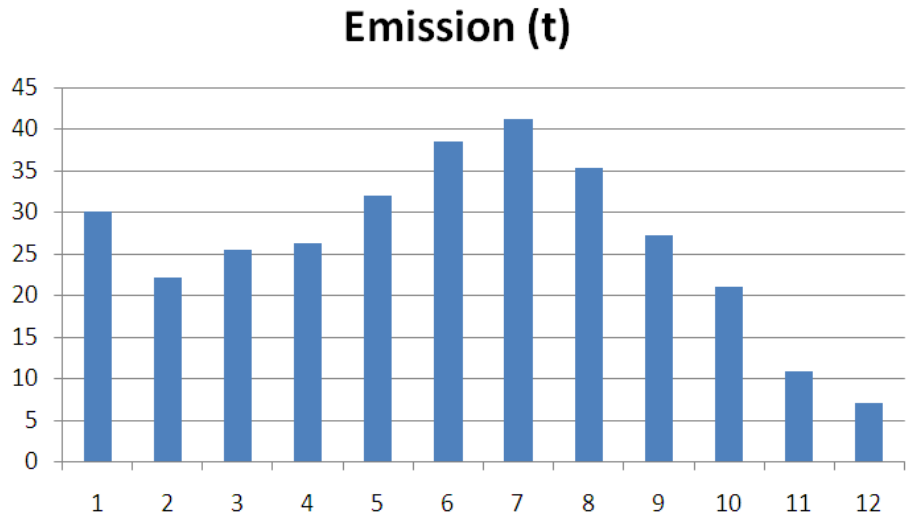

Fig. 2. Monthly emissions of gamma-HCH integrated over a box over Europe (10 deg W to 50 deg E and north of 20 deg N).

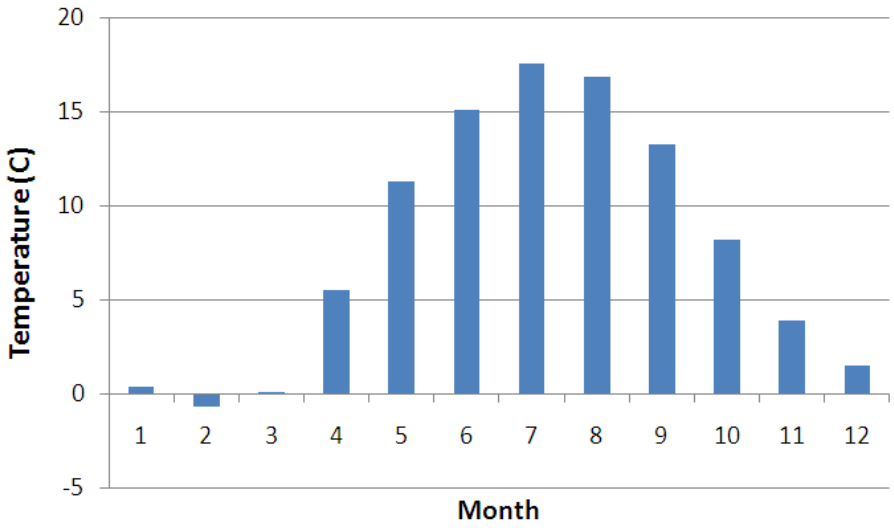

Fig. 3. Monthly temperature in 2005 in Europe.