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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatial variability of POPs in European background air" *by* A. K. Halse et al.

Anonymous Referee #1

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Manuscript evaluation criteria

- Scientific significance: 1 – Excellent
- Scientific quality: 2 – Good
- Presentation quality: 3 – Fair

Recommendation:

Accept subject to minor revisions

General comments:

The authors present results from an ambitious study in which passive air samplers (PAS) were deployed at EMEP monitoring sites around Europe for four months, then collected and analyzed for persistent organic pollutants (POPs). As described by the authors, this study is important because at present the spatial coverage of air monitoring for POPs within the EMEP program is highly biased towards sites in Northern and Western Europe, where active air samplers are used. Expanding this coverage to more EMEP sites using passive samplers would be a marked improvement over the current situation because it would provide data that is required to better evaluate fate & transport models and emission inventories. However, there are challenges associated with the use of passive samplers, since they provide only semi-quantitative information about concentrations of POPs in air.

The study appears to have been quite successful. Where it was possible to make comparisons, the PAS-measured concentrations are in reasonable agreement with active samplers. As discussed in the paper, the spatial variability in concentrations of a range of POPs measured in the campaign is consistent with current understanding of the sources and atmospheric fate & transport of these substances. The most valuable new contribution provided by the paper are the measurement data themselves, which provide a snapshot of spatial variability of concentrations of POPs in air in the European domain, and which will be invaluable to model developers working within the framework of the CLRTAP convention. The success of the study is also highly significant finding because it points the way to improving EMEP's monitoring program. As such I believe the paper will be of interest to readers of *Atmospheric Chemistry & Physics*, and that it should be published.

That said, prior to publication I strongly suggest that the authors add a full reporting of the raw data collected in the study to the Supplementary Material. This should include

estimated sampling rates at all sites, and details of the method and field blank levels for all substances analyzed, preferably as an Excel spreadsheet or in annotated text files. Modelers will require these raw data and will not be interested in summary statistics such as presented in Table 1, or the semi-quantitative information presented in Figure S3.

In addition, I believe there are many opportunities to improve the readability and clarity of the paper. The presentation and discussion of the data lacks polish throughout, and conveys the impression that the authors rushed this work to completion. This is a shame because the paper reports highly significant findings that have resulted from the concerted efforts of numerous collaborators and volunteers. I have provided several specific suggestions to improve the manuscript in the section below, with a focus on the first part of the paper. But, this is definitely not an exhaustive list of areas where improvements are possible, and I encourage the authors to critically edit the entire manuscript to address issues similar to those raised below.

Specific comments:

P. 22587, Line 6: Are POPs really 'common'? In what sense? Certainly only a small fraction of all chemicals in existence have properties that make them POPs. What is 'common' about that?

Line 9: Why use the word 'components' here when you are talking about discrete chemical substances that are not components of anything!

Line 11: I don't believe the goal of either the Stockholm Convention or the CLRTAP is 'to reduce future environmental burdens in remote areas'. Why be imprecise about what the goals really are?

P. 22588, Line 11: delete the word 'potential' that is modifying 'similarities'.

P. 22591, Line 11: change were → where.

P. 22597, Line 25 – 26: 'greater higher'?

P. 22598, Section 4.2: What happened to the results from Aspvreten in the comparison between passive and active sampling in this section? It is listed on Line 23 along with other sites, but no results are shown for any compounds. Also, the spelling of Råö is inconsistent between this section of the text and Figures 2 & 3.

P. 22599, Line 10 – 15: The discussion in this section is quite confusing. Differences in windspeed are proposed as a possible explanation for different concentrations of PCBs measured in duplicate samplers. But in the next paragraph, it is stated that results for the HCHs show no evidence that different windspeeds affected the samplers. The same samplers were used for PCBs and HCHs, so they should have experienced the same windspeeds! Couldn't the whole question of effective windspeed be resolved by comparing the rate of depletion of the performance reference compounds in the duplicate samplers? This is not even discussed in the paper at present.

P. 22601, Line 6: Again a passing reference is made to problems at the Aspvreten site that are not explained. Why is there only one PAS from this site?

P. 22601, Line 10 – 20: Here, the possibility that the active samplers and the passive samplers at Kosetice sampled different air masses is raised. I believe the authors could evaluate this possibility quantitatively using the results from their FLEXPART modeling. Why not do so?

P. 22608 & Figure 4: The agreement between the model and measurements is discussed in terms of logarithmic discrepancies (ie, 'within a factor of 2'). I believe this is appropriate, but it implies that Figure 4 should be presented as a log-log plot rather than on a linear scale.

P. 22609, Line 0 – 5: What effect would secondary sources of PCB 28 have on the EC maps that are presented here? There is a recent paper by Harner and Li in *ES&T* that suggests air-soil fugacity ratios of the less chlorinated PCBs indicate near equilibrium distribution. If secondary sources are important, aren't the EC maps derived from primary emissions going to be highly misleading?

Fig. 1: Panel E shows the value '0' twice on the vertical axis. I would prefer to see a logarithmic scale used in all panels. In both Figure 1 & 2 I think some statistics should be used to determine whether differences are significant or not.

Fig. 2: Panel F says 'Hivol' where it should say 'AAS'.

Figure S6 – S20: The two versions of the ES and EC maps for some sites should be better explained in the figure captions. I presume these are for the active versus the passive samplers? But, if this is the case, why aren't there two versions presented for Birkenes and Kosetece in Figure 5. As noted above, separate analysis using the model for the active and passive samplers could be used to discuss the possibility that different source regions influenced levels measured in the two types of samplers.

The scales on all of these Figures should be adjusted so that the colored portion of the ES map extends over all possible source regions identified in the EC map. It is frustrating to not be able to read quantitative information about the ES for most sites from anywhere in North America, but then to see that the east coast of North America makes a higher contribution in the EC map than many areas of Europe.

Finally the map in Figure 14 does not show the location of the sampling site!