

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

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General comments in this study, authors collected air samples with PUF-passive air samplers from 86 European background sites. Various kinds of persistent organic pollutants (POPs) were analyzed and the spatial distributions of them were investigated. In addition, simulation results were also considered. The data presented in this study are quite valuable and this topic is highly relevant to Atmospheric Chemistry and Physics. However, this manuscript needs some revision before the publication. See below specific comments.

We thank Referee #2 for helpful comments on this manuscript.

Specific comments:

Page 22586 1. The length of Abstract is unnecessarily long. Lines 10-17 can be removed from the Abstract, because the introduction of passive air sampling and EMEP programs is well described in the main text. 2. I think the result for the co-deployed PAS samples is not the main result of this manuscript, thus I would delete the relevant sentence.

The abstract have been shortened and modified accordingly.

Page 22590 1. For easier understanding, why don't you add a map of EMEP sites where PAS were co-deployed with AAS to the manuscript or the supporting information (SI)?

A map of EMEP POPs stations is now included in SI (Fig. S1) and reference to this figure has been included in the text whenever relevant.

Page 22591 1. I wonder effective sampling volumes for each target chemical were calculated when the default sampling rate was used.

Yes, the effective air sampling volumes for each target chemical was calculated based on the default sampling rate. This is now clarified in the text as well (section 2.4). However, the resulting air concentrations for these sites should be interpreted with caution (as commented in the manuscript).

2. In this study, 8 performance reference compounds (PRCs) were used for the calculation of sampling rates. This means that 8 sampling rates could be calculated for one sample. However, there is one sampling rate for each sampling site in Table S1 in the SI. I wonder if averaged values of sampling rates were used. Please describe the method used in this study.

Yes, an average sampling rate was used. The average sampling rate was calculated only on the basis of those PRCs which experienced sufficient loss (>40%). The last column in Table S1 in SI lists how many PRCs were used to estimated an average sampling rate for each site. This average sampling rate was in turn used to estimate

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the effective sampling volume for each substance. The method is now clarified in the text (section 2.4).

Page 22592 1. What is the definition of ‘emission sensitivities (ES)’?

An ES map illustrates the residence time for the arimass released at the sampling location during the sampling interval, and furthermore exemplify where the air mass has ability to collect sources of pollutants. This is now further clarified in the text of the main manuscript.

Page 22593 1. The half of the MDL from the field and method blanks was used to calculate air concentrations. It seems quite arbitrary to use the half values. They can be simply expressed as BDL (below detection limit).

We agree that the selection of using 1/2 MDL is somehow arbitrary. Yet, we chose this approach as it is consistent with former studies of relevance, such as GAPS (Pozo et al 2009 ES&T 43: 796-803), and because a numerical estimate is needed in order to calculate the statistics presented in Table 1. Secondly, while we agree it may be a proper suggestion to use below detection limit for values that fall below the method detection limit, we cannot see that this notation will increase the readability of Table 1. Actually, we suspect that the interested reader may want to inspect the numerical values used for statistical treatment. Finally, in order to minimize the risk of additional confusion by having to introduce and define BDL, we prefer to maintain the use of MDL with explanatory footnotes as given in Table 1. We hope the referee finds our decision agreeable.

Page 22594 1. There is no description how to estimate the uncertainty in the chemical analysis. How was the number of 35% calculated?

Further details on how the uncertainty in the chemical analysis was estimated is now included in the text towards the end of section 3.4.

Page 22598 1. In section 4.2, a map for EMEP sites will be helpful.

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Discussion Paper



Reference to the new figure (Fig S1) is now provided in the start of section 4.2.

Page 22599 1. The whole section 4.2.1 is about method validation. This section does not give any scientific information for monitoring data. It can be moved to the SI.

Section 4.2.1 has now been revised and moved to SI (new section S1.6) along with the corresponding Fig 1 (now Fig. S2).

Page 22600 1. Passive air sampling using PUF-disk has been already confirmed to be a complementary technique to active air sampling. Considering this, the key objective of this study mentioned in section 4.2.2 is not proper.

Our stated objective was not to assess the potential of PAS as a complementary technique to AAS in general terms, but to assess the potential of PAS as a complementary technique to AAS within the EMEP program. This is an important distinction, as AAS has so far been the recommended method for air monitoring of POPs within EMEP. We are fully aware that past studies have previously studied the complementary features of AAS and PAS, and we cite a few relevant studies in the beginning of this paragraph in the text. To further accommodate the concern and to better recognize relevant efforts carried out in the past, we have adjusted the first sentence of section 4.2.1 (former 4.2.2) to read; “One of the objectives of this study has been to further assess the potential . . . within the EMEP monitoring program”. Finally, we have also cited a number of studies in the introduction which have previously illustrated the complementary feature of PAS vs AAS (lines 51-54).

2. Passive air samplers are known to collect gaseous compounds exclusively. However, there is no mention that the active air sampling data are for gaseous data or the total data (i.e., sum of gaseous and particulate concentrations).

We agree that this is worth mentioning for those readers who may be less familiar with AAS and have now included a sentence in the text which states that “AAS and PAS sampling strategies are also fundamentally different as data obtained on the basis of

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AAS typically represent the sum of the particulate and gaseous air concentrations.”

Page 22601 1. The frequency of active air sampling was not well described in this section, but it was listed in Table S5.

We have now added further details in the text to help clarify differences in AAS sampling frequencies, durations and coverage for the various EMEP sites.

Page 22609 1. What is the main conclusion of this study? It would be better to add conclusion section.

We have added a new section at the end of the manuscript which now outlines the main conclusions from this study.

Table 1: Use ‘BDL’ when the concentration is below method detection limits.

Please find our response to this issue included above (Re: Page 22593 1).

Figure 4: Add units of X and Y axis.

Units have been added to Figure 4 (now Figure 3).

Figure 5: Explain the unit of ns/m³ (what is ns?).

The units for the emission sensitivity are nanoseconds per volume (m³). This is the residence time of air masses normalized by the volume. An explanation is now offered in the text (section 2.5).

Supporting Information Page 2: What is M.a.s.l.? I guess it is above sea level (m) or a.s.l. (m).

Correct. A footnote has been added to Table S1 which spells out the abbreviation.

Page 4: Remove one blank between 3) and Adjusted.

Corrected.

Page 7: The target ion of PAHs may be also [M]⁺.

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The target ion [M]⁺ has been added to PAHs in Table S3.

Page 9: Give an equation for the calculation of deviation between PAS and AAS.

The equation for the calculation of deviation between PAS and AAS which was included in section 4.2.1., is now also repeated in the title of table S5 for clarity.

Page 9: Remove a parenthesis for -77).

Removed.

Page 10: Remove 0, below the table.

Removed.

Page 11, Line 13: I could not calculate the spiked amount of PRC, because there is no concentration data for PRC.

The concentration data for the PRCs have now been added in section S1.1

Line 27: ml -> mL (Check it throughout the whole text.)

Corrected.

Page 12, Line 38-39: Sometimes, there is a blank between number and C, and sometimes there is no blank.

For consistency, the blank between the number and ĚŽC has now been removed.

Line 54: μl -> μL (Check it throughout the whole text.)

μl has been replaced with μL throughout manuscript.

Give information about the amount of internal standards injected.

Please find all information concerning the added standards in SI section 1.3.

Page 13, Line 77: 25m -> 25 m (Check it throughout the whole text.)

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Discussion Paper



Corrected.

Page 14, Line 88: 1mL -> 1 mL (Check it throughout the whole text.)

Corrected.

Line 104: 57-116% is averages from 8 PRCs?

The range in percentage recovery (57-116%) is the range in recovery for each PRC (N=8) in the field and method blanks individually (Table S2). As described in the SI (S1.5): The second recovery values were for the added mixture of PRCs. These values lie in the range of 57-116 %. The range in percentage recovery is governed from each PRC (8) in the field and method blank individually (Table S2).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 22585, 2010.

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