

Interactive comment on “Increasing and decreasing trends of the atmospheric deposition of organochlorine compounds in European remote areas during the last decade” by L. Arellano et al.

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This published ACPD version has been revised significantly based on the comments received from the first round quick reviews. While one reviewer posted more comments on this ACPD version, another review chose not to provide further comments. I have a few minor points for the authors to consider in the final version of this paper.

P3417, L23. Using “high solubility” instead of “low Henry’s law constant” may be more straightforward. This is because different units are used in literature for Henry’s law constant, and in some literature where a different unit is used than the one referred

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here, high H is equivalent to high solubility. In the main body of this paper where Henry’s law constant is first used, the unit needs to be provided and a statement that “low H generally means high solubility” is preferred.

Response. We usually prefer to work with H because when referring to organic compounds, which show low solubility in water, speaking about high solubility may be confounding. H is more precise in the context of atmospheric deposition because it defines the capability of the compound to be deposited by wet and dry deposition as well as by air-water exchange. H takes into account two physical-chemical properties, volatility and solubility, that are both important in the atmospheric deposition of POPs. For example, pp'-DDT has a relatively low H (1.1 Pa m³/mol) similar to α -HCH (0.741 Pa m³/mol), but the water solubility of the two compounds is very different (0.149 mg/L for DDT and 96.85 mg/L for α -HCH). In the revised version of the manuscript, we have provided the units of the H the first time that it appears (Page 7, Line 13)

P3418, L23. Some species have short lifetime, but can reemit from the surface after deposition and are then transported further down the wind (the so-called grass-hopper effect). Is this mechanism considered in this study?

Response. Actually the occurrence of the grasshopper effect has been confirmed and constitutes the base of our studies in high mountain areas (see for example: Grimalt et al 2001, ES&T, 35, 2690-2697, Arellano et al 2011, ES&T, 45, 9268-9675, among others). In our bulk atmospheric deposition devices, samples remained in the collectors during 15 or 30 days. In these conditions we assume that what was measured was the net amount of each compound that would be really deposited in these ecosystems during the considered sampling period, therefore, the influence of the grasshopper effect would be already included in our data.

P3420, L24. In general, bulk deposition collectors only collect a portion of dry deposition besides collecting all of the wet deposition. The magnitude of this collected dry deposition portion depends on species gas-particle partitioning, among many other

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factors (solubility, meteorological conditions, materials used in the collectors). Can the authors provide a brief discussion on how much portion the dry deposition was collected (should be species dependent)? Such information might be important, because in reality, dry deposition contributes more than wet deposition on annual basis for many pops (this is especially the case in non-snow season over vegetated surfaces).

Response. It is difficult to estimate the proportion of dry deposition collected by our bulk atmospheric deposition sampler. In initial experiments performed in 1997 in Redòn, we compared the efficiency of collection of our bulk deposition sampler with another sampler prepared for collecting dry and wet deposition separately. Focusing on the particle amounts, in some occasions the efficiency of the bulk deposition collector encompassed between 30 % and 50 % of the particles collected in the dry+wet deposition sampler, but in other cases the particle fluxes measured in the bulk atmospheric deposition samples were higher than those measured in the dry + wet samples. Despite its limitations, bulk atmospheric deposition is considered a good approach to estimate total deposition of organic compounds. In the context of our study, it is not so clear that dry deposition contributes more than wet deposition (on annual basis) to the deposition of the studied compounds. It has to be taken into account that our sampling sites are located in remote mountain areas situated above the tree line, with low vegetation (catchment areas constituted basically by bare rock). Furthermore, they receive significant amounts of precipitation in form of snow and rain (more than in low altitude sites).

P3423, L16-22. Are these concentrations referred to ambient air concentrations? For all classes of POS, deposition and air concentration need to be discussed together to better explain the trends and patterns.

Response. No, they aren't. We have not measured air concentrations in this study. These are deposition fluxes calculated from concentrations measured in deposition samples. As it is explained in P3423, L3-6, atmospheric deposition fluxes ($\text{ng m}^{-2} \text{ mo}^{-1}$) were calculated by multiplying the OC concentrations (ng L^{-1}) measured in each

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sample by the collected volume and dividing by the surface area of the collection funnel and the sampling time, around one month for most of the samples.

PP3425, L4. Also see a more recent and quantified study of snow scavenging (Zhang et al., 2015, ACP 15, 1421-1434)

Response. Thanks for informing us about the work by Zhang et al, from which we were not aware when submitting this manuscript. This work deals with different organic compounds than those considered in our study but confirms that snow is more efficient than rain for organic pollutants scavenging from the atmosphere. This reference has been added to the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 3415, 2015.

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