

Interactive comment on “Snow scavenging and phase partitioning of nitrated and oxygenated aromatic hydrocarbons in polluted and remote environments in central Europe and the European Arctic” by Pourya Shahpoury et al.

Anonymous Referee #2

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Shahpoury et al. present data on nitrated and oxygenated polycyclic aromatic hydrocarbons (N/OPAHs) from snow samples at different European sites. They report concentrations from particulate and liquid (melted) snow phases and estimate the fraction of N/OPAHs removed by precipitation through particle scavenging based on predicted particulate mass fractions and observed snow phase partitioning. Such data are sparse and could in principle be helpful to better understand wet removal of hydrophobic and less hydrophobic organic compounds from the atmosphere. I have, however, a number of major issues with the applied methodology and data processing which need

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clarification before the paper can be considered for publication. In addition, the paper lacks important details and explanations to be able to fully understand what has been done. The structure of the paper and the clarity of the results' presentations need also be improved. I recommend re-consideration after major revision.

1) The authors aim at measuring the phase partitioning of N/OPAHs in snow by analyzing both the snow particulate and aqueous phases. To do so, they pass the melted snow samples through a filtration-extraction system. My concern is that the observed distributions between snow particulates (retained on a filter) and snow water (extracted on a sorbent) might be strongly biased for compounds with some water solubility, which upon sample thawing will dissolve from the particulates into the melt water. This potential artefact needs to be thoroughly addressed as it might render many of the presented results useless. One of the main findings of the study is the unexpected behavior of highly water soluble NMAHs (p11 l29-30). Considering the potential experimental bias, I wonder if this finding really holds.

2) Snow particulate concentrations are given in ng L⁻¹, which will strongly depend of the final volume during sample extraction. As this final volume is an arbitrary choice of the authors (and not even reported), it is not clear to me how these concentrations can be used in any reasonable way beyond comparing between different samples. Even the comparison with snow aqueous phase concentrations seems difficult to me. Adding them up to a total snow concentration, as done in Fig. 1 and Table S4A seems hard to justify as well to me.

3) To make my confusion complete, in Table S4B the authors calculate dimensionless total scavenging ratios from the sum of snow particulate and aqueous phase (in ng L⁻¹) and the sum of particle and gas phase concentrations (in ng m⁻³), obtained just before the snow events. This ratio seems to be derived by multiplying ng L⁻¹ concentrations by a factor of 1000 and then dividing by ng m⁻³ concentrations, ignoring that the denominator in the unit is referring to solvent volume in one case and air volume in the other. This seems very odd to me and needs explanation.

4) The conclusion that the phase partitioning of NMAHs is determined by an interplay between GPP, particle mass size distribution, and dissolution during in- or below-cloud scavenging (abstract I17-19 and p12 I13-19) is weak and not convincing. Even assuming the applied methodology was artefact-free and the calculation of the total scavenging ratio can be justified (see above), I cannot follow the authors' reasoning why the observations would indicate an importance of particle mass size distribution and dissolution. This needs to be much better laid out in the discussion and all data in support need to be shown (p12 I14-15). Much of this conclusion seems to originate from one single sample, where additional measurements have been made. The poor robustness of results that would follow from this needs to be addressed as well.

5) On p7 I13ff and p12 I6ff the authors discuss acid dissociation in particles in relation to pH. However, they seem to not be aware of sample pH being a different thing than particle pH, i.e. pH of particle liquid water. With a melted snow sample being highly diluted in comparison to aerosol particles, the measured sample pH cannot be used to describe acid dissociation in particle liquid water. This needs to be corrected appropriately.

6) A "dissolved" phase is not a thing. There are particulate, aqueous, or gas phases, for example, but a dissolved phase does not exist and the term needs to be replaced throughout all the manuscript.

7) The experimental section lacks many details, e.g. flow rate during extraction (p4 I23), volume of ethyl acetate (p5 I8-9), volume of buffer (p5, I32-33), concentration of EDTA (p5, I33), duration of ultrasonication (p6 I1), volume of buffer (p6 I1-2), etc. Please make sure any reader would be able to fully repeat your experiments with the information given.

8) Data presentation could be improved. All the abbreviations for the different sites are impossible to remember. I suggest removing them completely from the discussion text and referring to the name of the sites instead. Other abbreviations are used without

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explanation, e.g. the indices WSOM and OP (p6, l29). Different panels in the Figures are sometimes difficult to compare, because the order of compounds changes. Figure 3 contains empty brackets in the y-axis label, Figure 4 even contains only empty brackets as y-axis label. The discussion of air mass back trajectories in 3.1 is odd, as it is nowhere else in the manuscript referred to. If still important, it should be substantiated.

9) Correction factors for fOM (p7 l1-2) are taken from a study in the 1980s done in Los Angeles, USA. I wonder if this is really the best reference for the sites sampled by the authors. Also, these factors are likely to differ substantially both between sites but also between different meteorological situations. A note on the sensitivity of the results on such highly uncertain parameters would be helpful.

10) Sections 3.2 and 3.3 are tedious to read with all the abbreviations and very detailed concentrations. I suggest discussing the main observations in these measurements in a more compact way and leaving much of the numbers to the corresponding Figures and Tables. Also, some paragraphs might be moved to the Introduction (e.g. p8 l14-22). The space gained here should be used to discuss main findings of the paper in a more substantiated way (see comment above).

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