

## Response to Reviewer 1

*This manuscript makes a strong contribution to advancing our understanding of passive air sampling by critically evaluating the existing paradigms of sampler deployment and, importantly, identifying a strong source of uncertainty that is being introduced into the data. The overview of the state of passive air sampling is accurate and correctly captures the uncertainties in passive sampling sorbents, configurations, and uptakes that impact PAC quantification by PAS.*

*The authors take a step-by-step approach in testing a series of hypotheses - this structure is very clear, and very comprehensively integrates the results from the different methods that were combined in the study. The scientific methods and interpretation are very good, and only a few points require further clarification.*

We appreciate the reviewer's endorsement of our work.

Specific points for the authors to clarify:

*Please clarify hypothesis 1 "Can Uptake of Particle-bound Compounds Explain Decreasing Trends in the Amounts Accumulated in PASs?" While I fully agree with the methodology used to test the hypothesis and the conclusions the authors have made with respect to this hypothesis, I question how this phenomenon could lead to decreases in chemicals in the samplers over time. I rather think this would lead to irregular/non-linear uptake. Does this hypothesis assume that particles can be "blown-off" the sorbents in over time?*

The reviewer is correct that the uptake of particle-bound compounds does not provide an explanation for the amount of a chemical quantified in the sampler sorbent decreasing with deployment time. This hypothesis is rather suggesting that because the uptake of particles can be influenced by the size, type, and concentrations of particles in air and the wind exposure of a PAS during deployment, it could result in inconsistent and unpredictable uptake of PACs, including in uptake curves that could be suggestive of compound loss from the sampler sorbent. We agree that this was not clearly articulated. We have rephrased the introduction of that hypothesis as follows:

The first hypothesis posits that the observed uptake behaviour was caused by the ~~potentially inconsistent and unpredictable~~ uptake and accumulation of atmospheric particles in the XAD-PAS. This hypothesis is based on previous studies showing that some PASs do not efficiently block wind from carrying particle-bound substances to the sorbent, as is, e.g., observed in the case of the PUF-PAS with double bowl shelters (Wania and Shunthirasingham, 2020; Chaemfa et al., 2009; Bohlin et al., 2014b; Harner et al., 2013). Even though there is currently no evidence of the uptake of particle-bound SVOCs in the XAD-PAS, if such uptake were to occur, the accumulated amount of a chemical in the XAD-PAS would be the sum of the amount taken up from the gas phase and the amount derived from sampled particle phase. **While this admittedly cannot explain why the amount of a chemical quantified in the sorbent would decrease with deployment time, it could result in inconsistent and unpredictable uptake of PACs, because such the uptake amount could be influenced by the size, type, and concentrations of particles in air and the wind exposure of a PAS during deployment. Some such inconsistent uptake could conceivably be suggestive of compound loss from the sampler sorbent.**

*Please consider adding more specificity to the naming of the hypothesis related to degradation (hypothesis 3) to make it clear that this is related to photooxidants, largely ozone. In fact, the outcome of the study seems to be that some other unknown degradation pathway is contributing to the observed patterns, but that aspect is not tested under the 3rd hypothesis.*

We agree with the reviewer and have changed the name of hypothesis 3 to "Can Reactions with Photooxidants Explain Decreasing Trends in the Amounts Accumulated in PASs?"