Response to Reviewer 2

General comments

Review of "Seasonal Air Concentration Variability, Gas/Particle Partitioning, Precipitation Scavenging, and Air-Water Equilibrium of Organophosphate Esters in Southern Canada" by Li et al.

Li et al. present extensive long-term measurements of organophosphate esters (OPE) in air, particles, precipitation and bodies of water. Measurements of OPEs across different environments are quite rare and merit publication. The results show OPEs are ubiquitous and show moderate seasonal trends in some cases. In addition, the authors discuss partitioning between these environments and whether they fit their theoretical or estimated partitioning coefficients. The authors did an excellent job discussing the data, citing relevant references and discussing the limitations of these challenging measurements. The article should be accepted after addressing the following small comments

We appreciate the positive feedback.

Specific comments

Figures S1 and S2 can be moved to the main text for readability purposes.

While we appreciate the suggestion, we still prefer to leave Figures S1 and S2 in the supporting information, considering (i) their large size and (ii) that the data in these two figures are displayed differently in Figure 1.

Line 201: ***"We are no comparing…"*** *no should read not.*

We will correct this typo by replacing "no" with "not".

Section 3.2: The measured concentrations vary significantly both on a species level as well as a function of location. Could the authors expand upon why some OPEs show higher concentrations in places like Antarctica vs Toronto or similar concentrations in urban and rural environments?

OPE concentrations in Antarctica that are higher, or within the same range as, concentrations we measured in Toronto are difficult to explain by long-range atmospheric transport. Local sources could be responsible for the OPE concentrations reported by Wang et al. (2020a), considering that their Antarctic sampling location was close to the Chinese Great Wall station. Sühring et al. (2016) also attributed higher OPE concentrations at Resolute Bay in the Arctic to the influenced of local sources.

Concentration levels of OPEs on atmospheric particles on Saturna Island and in suburban Toronto were similar. We do not have a clear explanation for this observation, although there is the possibility of local sources influencing the deployment location of the active air samplers on Saturna Island. For example, this site was close to a weather station and a ferry terminal.

Line 277-278: ***"the dispersion plume of the Montreal waste water treatment plant enters the river at 45 40' N, 73 28' W and stays on the north side of the river"*** *Perhaps this feature can be added to the figure as a marker.*

While the scale of the map in Figure 1 is too small to indicate this feature graphically, we will move this sentence to the caption of Figure 1.

Line 368-370: ***"However, the unexpectedly low fraction observed in the particle phase may suggest that TPhP and EHDPP are emitted at higher temperatures and are not in a state of equilibrium between gas and particle phase"****What would prevent these species from achieving equilibrium within the timescales of the measurements?*

We refer to the study by Zhao et al. (2021a) for a detailed explanation. Zhao et al. (2021a) performed simple model calculations that show that compounds with very low volatility emitted at high temperature do not reach equilibrium because "the time scale for chemical exchange between the gas and particle phases [is] longer than the deposition

removal time scale of the sorbed chemicals". Simply speaking, the particles are not airborne for long enough to reach equilibrium.

Zhao et al. (2021b) had previously observed gas-particle partitioning of OPEs that appears to indicate non-equilibrium. They suggested that this could be a result of the operationally defined gas phase which includes very fine and ultrafine particles that might pass through glass fiber filters. This would cause an overestimation of the fraction of OPEs in the gas phase. We will now add a sentence indicating this as an alternative explanation for the apparent non-equilibrium: "Alternatively, the fraction of TPhP and EHDPP in the gas phase may have been overestimated if very fine and ultrafine particles containing these OPEs passed through the glass fiber filters (Zhao et al., 2021b)."

We note that earlier studies often observed the opposite behaviour, namely much larger particle-bound fractions of relatively volatile OPEs than might be expected.

Line 461-463: Specific industries that use OPEs could also be more active in the summer months e.g. construction.

Thanks for pointing this out. We will modify the original sentences by adding the potential influence of seasonally variable industrial activities: "(i) an increased release of OPEs at higher temperatures from outdoor materials to which they have been added (Kemmlein et al. 2003), (ii) a faster ventilation of OPE emitted indoors (Stamp et al., 2022, Han et al., 2024),), **(iii) more active industrial activities, such as construction, using products containing OPEs in the summer months,** and (iv) the atmospheric oxidation of organophosphite precursors (Liu et al., 2023)."

All references cited in this response can be found in the submitted preprint of the manuscript.