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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Non-equilibrium interplay between gas-particle partitioning and multiphase chemical reactions of semi-volatile compounds: mechanistic insights and practical implications for atmospheric modeling of PAHs" *by* Jake Wilson et al.

Anonymous Referee #2

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General comments: This paper summarizes a computational study investigating the effects of chemical losses (reactions with O3 and OH) on gas-particle partitioning of PAHs on soot particles. This study is important because it reiterates the idea that equilibrium partitioning is not an accurate model of the gas-particle distribution of PAHs, but it further demonstrates that a number of factors can affect the non-equilibrium distribution of PAHs between gases and soot particles and how far from equilibrium these impacts perturb the system. I think it should be published after addressing my comments

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below, and also an overarching question I was left with: how much do the authors predict these findings will impact the results of current global and regional CTMs? They conclude by saying that the model is too computationally intensive to be deployed, and that another algorithm summarizing the results could be created, but even if that is done, do they anticipate a large change in results?

Specific comments: It would be useful to add a short section at the end of paragraph 2 in the Introduction that explains why gas-P partitioning is important assessing human health hazards – a short description of the differences between gas-phase and particle-phase deposition behavior and also uptake into lungs.

Line 62: I suggest explicit definitions of SOA and “particles” or “aerosol particles” to aid readers.

Line 76: Does PRA stand for something? If so, would help to define the abbreviation.

Line 78: Quantification of equilibrium processes for six PAHs is mentioned, but this is a bit confusing because the abstract only notes using pyrene as a case study. I suggest clarification in both the abstract and the end of the intro on which parts of the study employ six PAHs and where pyrene is used as a model PAH.

Line 97 through 106: It would be helpful to put the diffusion flux terms on Fig. A1 and to very briefly explain their direction in the main textâĂŤit is not clear without reading the appendix that the gas diffusion flux is between the gas phase and the near-surface gas phase. Another thing that kind of confuses me is the representation of the gas phase a finite layer in Fig. A1. It made me wonder why equation 1 didn’t contain a term for the influx of gas-phase particles, because I was thinking of it as something separate (as part of the particle) from the infinite bulk gas phase of the atmosphere. Is there a way to graphically represent it as a layer that extends into space on the top side in A1? Finally, the term LS is used in equation 3, but the term LP is defined in the text (line 104).

Line 113: Why the choice to use the same O3 rate coefficient for all PAHs observed?

Line 122: Same question for the rate coefficient for PAHs with OH.

Since the abstract states that the equilibration timescale depends on factors including temperature (and the influence of temperature is mentioned several times throughout the introduction), it is surprising that the temperature dependence of kinetic rate coefficients is not taken into account.

Section 3.1 of the results and Fig. 2 in particular nicely outline the approach of the paper, and though this was hinted at in the intro, I feel that it would be helpful to have something a little more detailed like this earlier on, like in the intro. I couldn't really picture the goals/approach well until reaching this point in the article.

Line 180: There seems to be an entire section missing on PAH molecular structure!

Lines 204-205: The description of the shading in Figure 3 is backwards.

Throughout: the concentration unit of cm-3 is kind of confusing to me. At the beginning of the results, it is explained that PAH concentration is kept at 5 x 105 cm-3. Is that molecules per cm3? And if so, why is not stated? [Also, as an aside, this seems like a detail that should be placed in the methods rather than the results.]

Figure 8: Delta t = 1 min and Delta t = 30 min. should be overlayed on plots a) and b) respectively.

Also Figure 8: It would be interesting for the authors to highlight the area on these plots where they feel most CTMs reside. . . are desorption kinetics too fast? What about O3 concentrations? If modeled, these usually vary based on simulated chemistry. Also, do most models even include O3 oxidation? It seems to me that many only consider OH. Is there an opportunity here to compare the relative importance of OH and O3 oxidation as a whole to PAH lifetimes?

Technical corrections: Line 70: Replace "accounting" with "account".

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