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Atmospheric Chemistry and Physics Discussions

Interactive comment on "PAH concentrations simulated with the AURAMS-PAH chemical transport model over Canada and the USA" *by* E. Galarneau et al.

Anonymous Referee #2

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This manuscript summarizes the construction and evaluation of a regional (Canada and USA) atmospheric PAH model. The manuscript adds significantly to the literature on this topic, as there are no other North American regional atmospheric chemical transport PAH models in existence at this time. The authors test the model's ability to simulate both total atmospheric PAH concentrations as well as the distribution of concentrations between the gas and particle phases using two different gas-particle partitioning parameterizations. The evaluation of model ability to predict gas-particle partitioning against observations is not often investigated in similar modeling exercises, further adding the study's novelty. I found no technical corrections to be made (other than perhaps limiting the abstract to one paragraph; in my view, an abstract is too long if it needs to be separated into paragraphs). Therefore, I would like to see the following questions/comments addressed before final publication (i.e., somewhere between minor and major revisions):

Please define AURAMS at first use within the main text.

Please explain the motivation for the choice of gas-particle partitioning schemes tested (i.e., why was organic matter sorption alone omitted?).

Dry deposition: I would imagine that the cuticle and ground resistances for PAHs are actually quite different from those of acetaldehyde and C3 carbonyls, as they are much larger compounds with greater aromaticity. Can the authors comment on how the scaling factors used may impact results?

Since loss via oxidation by hydroxyl radical is a major sink for atmospheric PAHs, can the authors comment on the potential influence of using the same OH reaction rate constant for the four larger PAHs?

What is the effect of assuming fully reversible partitioning? What is the justification for this, considering the primary author has previously published data suggesting that the partitioning process may not be fully reversible?

Which particle size bins end up with the greatest fractions of particulate PAH?

I find line 25 of page 18424 to be a bit confusing. Do Jonker and Koelmans report values for sum PAH?

Can the authors comment on the possible influences of including snow scavenging?

Line 5, page 18429: Please specify that annual means are being discussed, rather than “overall means”.

What are some other possible reasons for the smaller temporal RSDs in model results compared to measurements besides temporal variability in emissions? Have OH

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and/or particle concentrations simulated by AURUMAS been compared to observations within the same model domain, and do the temporal RSDs of these quantities vary less in the model compared to observations as well?

Line 17, page 18428: Please use “total concentrations” rather than “values”.

Line 6, page 18429: There are more recent references for particle-reactive BaP (e.g., Poschl et al., 2001, J. Phys. Chem. A; Shiraiwa et al., 2009, Atmos. Chem. Phys.; Kwamena et al., 2004, J. Phys. Chem. A).

Lines 13-26, page 18429: I would argue that the results in fact DO show that there is a trend toward underestimation of more volatile PAHs, indicating surface volatilization is an important process. The authors state that the results are ambiguous, but in general, the more volatile PAHs are underestimated, the mid-volatility PAHs show little bias, and the low volatility PAHs are overestimated. I would like to see this section rewritten to acknowledge that revolatilization may in fact significantly contribute to atmospheric concentrations. Also, line 13: do the authors mean “water and soil” rather than “air and soil”?

It would be useful to have some statement at the end of Section 3.1.1 that hits on the utility of a regional model that simulates concentrations generally within an order of magnitude. What are the model strengths versus other models (e.g., global) that could simulate North American concentrations at more coarse spatial resolutions? Later there is some discussion of finer resolution hopefully providing more accurate results, but can the current version of the model be employed to answer outstanding questions regarding PAHs in North America?

Deposition: Do any of the measurement stations included in the study provide deposition measurements in addition to concentration? It would be interesting to see how well the model simulates deposition, particularly for the less volatile compounds. This would also help determine whether revolatilization does indeed have a substantial effect on atmospheric concentrations or not (perhaps deposition of volatile PAHs is

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overestimated compared to particulate PAHs). I ask that the authors please comment on this.

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