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PAH Concentrations Simulated with the AURAMS-PAH Chemical Transport Model over Canada and the USA

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The authors have revised the manuscript according to the comments/queries raised during my original review. Importantly, they have considered my key points below and provided clear and satisfactory responses including key amendments to the manuscript marked by additional/modified text and a figure (Fig 4) concerning surface-to-air volatilisation sources on atmospheric PAH levels. This has also resulted in an amended/improved abstract. The authors have clearly thought about this issue at some length and their response to my comments have been thorough and systematic. This study is one of a handful of high-resolution CTM studies to tackle semi-volatile chemicals like PAHs. Most of these studies neglect or discount the air-surface exchange component on chemical concentrations in air. However, this manuscript now goes to some length in assessing/quantifying this issue on model results and as such is a valuable addition to the literature.

I have no reservations in recommending publication.

First review:

The authors present a revised manuscript detailing a modelling study that simulates PAH air concentrations across North America using a Eulerian CTM model. The simulation is based on the year 2002 and includes 7 PAHs that cover a relatively wide range in physical-chemical properties. This is one of only a handful of studies that has attempted to predict air concentrations at high spatial-resolution for these chemicals and considerable effort has been expended on gas-particle partitioning given the semi-volatile nature of these compounds. The primary emissions of PAHs appear to be very well handled and detailed. These build on a methodology previously published by the lead author. Similarly a thorough model description is provided on the various fate/behaviour processes, including the use of two gas-particle partitioning approaches.

The results are usefully presented including a thorough comparison with measurement data (modelled/measured ratios). I believe the paper is of publishable quality for ACP. However, the authors need to do more to address the concerns raised by a previous reviewer regarding the contribution of temperature-driven re-emission from secondary sources. Below are two key points which the authors need to highlight/address:

(1) For the 7 PAHs studied here secondary sources (re: air-surface exchange) are most pertinent for phenanthrene (PHEN). Its isomer anthracene (ANTH) is probably too reactive for this processes to be fully significant (i.e. its half-life in air and soil is relatively short, plus its presence in air is erratic and can be driven in the main by notable point/primary sources). For the higher MW 4-ringed PAHs of fluoranthene

and pyrene the magnitude of temperature—driven re-emission from secondary sources will be low relative to primary emissions (particularly over a single summer season re: 2002). Secondary emissions are negligible for the high MW PAHs (e.g. B[a]P). The authors need to make these points clear in the manuscript and cite studies that have observed this phenomenon and provide a few quantitative details (i.e. the observed seasonality in low MW PAH concentrations brought about by secondary re-emission) (there are plenty of good studies in the literature for soil, water bodies, vegetation and urban surfaces).

(2) Section 3.1.1. para (line 419-434). The authors discuss the model results with regards to the exclusion of air-surface exchange in their model. This paragraph is essential but I disagree with the flow of their discussion as they state that the model summary results provide indefinite evidence. Far from it(!) – the model appears to be working very well - and from the evidence provided both in Table 1 (summary comparison between model and measured) and Fig 3, the model appears to fit with PAH behaviour as described in point (1) above. For PHEN, the model clearly under predicts the air concentrations. There is nothing ambiguous about this and the underestimated PHEN concentrations in the model must presumably be due to the re-emission component which is missing from the model. This is fine and not a problem for the manuscript, but the authors need to state this clearly and examine this issue more carefully and include this observation/finding in the abstract. For example, I would like to see a map of the spatial distribution of model/measurement ratios for PHEN, for both winter (i.e. Dec – Feb) and summer (i.e. June-Aug). The hypothesis here is that the ratios will be closer to 1 in the winter and deviate further from 1 in the summer (i.e. as the re-emission component becomes more relevant during the warmer part of the year). The authors should 'play up' this part of the paper and not try to discount discrepancies as ambiguous or uncertain. Another case in point is ANTH. As ANTH is relatively volatile it is not surprising that the model average is below the measurement average (again akin to PHEN re: re-emission component) but the data are variable and skewed (hence similar model/measured medians). This is just what I would expect for ANTH released from notable point sources but with a relatively short half-life.

In short, the authors need to highlight the role played by air-surface exchange on PHEN and ANTH concentrations, and use the model results to demonstrate/quantify this process rather than discount it as model ambiguity. In fact, I believe this could be a real strength to this paper, more so than the gas-particle (g-p) partitioning story (especially as the two g-p approaches show little difference).