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Geoscientific Model Development Discussions

Interactive comment on "GEM-MACH-PAH (rev2488): a new high-resolution chemical transport model for North American PAHs and benzene" *by* Cynthia H. Whaley et al.

Anonymous Referee #1

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General comments:

This discussion paper addresses the high-spatial resolution modelling of PAHs, which is relevant within the scope of GMD and important for the estimation of air quality. The presented model improves upon existing PAH prediction capabilities, making novel improvements to an existing tool's performance. The methods and assumptions are generally clearly outlined and valid with the conclusions supported by the results, with the exception of some specifics discussed below. The specific model improvements discussed are precisely and clearly presented, and therefore should be reproducible. To the best of my knowledge, the authors give proper credit to related work and clearly

indicate their own contribution. The title clearly reflects the contents of the paper and the abstract provides a complete and relatively concise summary. Overall, the paper is very well structured and clear, and the language is completely fluent and precise. The number and quality of the references is appropriate, as is the supplementary material. The authors provide a link to model source code, but do not include a user manual or compilation/run instructions and dependencies.

This paper represents an advance in PAH modelling, where the model appears to be limited by the availability of inputs, particularly emissions; the inability of such a high-resolution model to capture daily variability appears to be likely due to unresolved variability in emissions. However, the ability to make even seasonal-scale PAH estimates comparable to highly local measurements given emissions scenarios is an important asset for air-quality science.

While the abstract, conclusion, and body of the paper include ambiguous use of "statistically indistinguishable" which overstates the performance of model, the actual performance of the model represents a sufficient advance for PAH modelling.

Specific comments:

Adjusting Ksw to measurements taken in 2002 is a good way to navigate their high uncertainty, but some of the adjustments are incredibly large. E.g. PHEN and PYR Ksw increases by almost 2 orders of magnitude. The authors should discuss the justification of such a large change in the context of the prior uncertainty and/or possible missing mechanisms.

Equation 6 holds only if m_i_gas is equal to the total mass of PAH in the parcel of air considered; i.e. prior to partitioning, all of the PAH is gas-phase. Does this mean that once the PAH partitions to water it is considered lost? Is partitioning to cloud-water irreversible? A clarification of the fate of PAH that undergoes water uptake but not precipitative loss from the atmosphere is warranted.

ca. L460 Biases of BaA and BaP indicate NO3 reactions that are noted in the literature. Here a few more lines of discussion of this point would be helpful. A back-of-the envelope estimate of the effect of these reactions would increase confidence that these are the reason (or not) for the remaining bias.

ca. L560 The R-values in these site-by-site comparisons are fairly low even after the anomalous sites are removed. Discussion of the causes of this low correlation is warranted. At the considered time and spatial scales, is unresolved time-variability in emissions too large? The conclusions should highlight further the reliance on accurate emissions and their time and spatial distributions for daily and small-scale predictions.

The inability of the model to reproduce short-timescale PAH concentrations is a weakness considering its high resolution.

The number of aspects of the simulation that are compared to observations is a major asset for this work. The comparison to Kp, particulate fraction, wet deposition, and concentrations across many sites and PAHs allows a very detailed and transparent assessment of the model.

The winter/summer differences in wet deposition show that snow-initiated wet deposition is a definite weakness of the model. In the conclusions, while the authors mention that snow scavenging is new to the model, it should be acknowledged that it requires improvements going forward, along with possibilities of what these improvements might be.

ca. L675 “but it is at least promising to see that there are no particular sites where the model is consistently too high or too low, rather the errors in spatial distribution are haphazard and may be due to propagation of error, rather than any major error with the PAH scavenging scheme itself” I think that this paragraph should be re-written. The difference in the model compared to the observations is definitely due to the propagation of error, in all cases. The fact that these errors are different for different PAHs and sites is a result of the complexity of the processes involved, as the authors write, but I

do not think that this makes them more or less promising.

ca. L705 “we have determined from our sensitivity test that the GEM-MACH-PAH model has a linear response to a 50% variation in mobile emission factors, simulating concentrations that vary up to 30%.” In the results section, the authors present a /non-linear/ response of concentrations to variations in mobile emission factors. Increasing emission from 50% to 100% yields $\sim$10% difference in concentrations, while increasing from 100% to 150% yields $\sim$30% difference in concentrations.

Figure 8) Further displays non-linearity of mobile source factor. What is the reason that it is not linear? What change in total emissions results from the mobile source change?

Abstract, L694 and ca. L570 “with concentrations statistically indistinguishable from observations, at 2.5-km resolution”. If I understand the analysis, this phrasing highly overstates the performance of the model. Firstly, the 2.5 km resolution is not a significant part of the model-measurement comparison. The sites are a few dozen distributed all across the Northeastern U.S. and Southern Ontario, and are aggregated, and therefore the comparison is not testing the high spatial resolution. Secondly, by grouping all of the measurement-model pairs for the whole domain and season, a more accurate statement of the agreement would be “Over the domain as a whole and on the seasonal time-scale, the model is unbiased with respect to measurements.” The above phrasing is misleading and ambiguous, and must be changed to at least clearly state the statistical test performed.

Technical corrections:

Equation 1: “b” in equation, but “B” in text

L165 “amoung” should be “among”

L222 “In order to investigate whether these U.S. values would be representative of conditions in Canada and whether only have those two fuel-type categories are adequate,

. . .” I believe should read: “In order to investigate whether these U.S. values would be representative of conditions in Canada and whether having only these two fuel-type categories is adequate, . . .”

Equation 4: the “reduced” and “i” labeling is confusing; indicating a forward time-stepping would be clearer.

L269 “. . .for the seven PAHs is a linear relationship with inverse temperature.” Should read “. . .for the seven PAHs are proportional to inverse temperature.”

L380 “NATTS is an a U.S.” should read “NATTS is a U.S.”

L444 “. . . slope of the best-fit line is very close to 1.” Here it is preferable to simply quantify the slope and remove the qualitative phrase “very close”.

L648 “gages” should read “gauges”

Figure 4: The white circles are difficult to see on difference maps. A more visible color should be used. In all 4 map panels, the grey color of some of the dots is not on the color scale.

Figure 5 a) text too small Figure 5 b) great figure, but purple overlay hard to discern.

Figure 6 a) Should remove irrelevant labels on each y-axis (<=1e4 on left, >1e3 on right)

Figure 10 a) remove meaningless negative particle fraction axis labels Figure 10: b) does not exist but is mentioned in Figure 10 caption.

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