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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Top-down estimates of benzene and toluene emissions in Pearl River Delta and Hong Kong, China" *by* X. Fang et al.

**Anonymous Referee #4**

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General comments: This paper presents one of the first estimates of benzene and toluene emissions in China using atmospheric measurement data from a rural site in the region and inverse modeling. This paper conveys important information about the toxic pollutants in China. In particular, this paper suggests the need for more measurements to better constrain these emissions.

Specific comments:

Although the result is promising, the authors need to address a few important issues before publication. First, with regard to transport model evaluation, I don't see enough description or result on this. In terms of the methodology, it should be addressed in Section 2.2, but there is no information about potential model biases. The simple

question I can ask is how reliable the NCEP meteorology is in this region of China. This is not about random errors we consider in the inversion. If the transport model is biased, then we get the biased mean estimate for the posterior emissions. The authors need to evaluate the transport model only for less than a month (pretty short period compared to other many studies). I suggest that the authors evaluate the transport model for potential biases (e.g., wind bias, planetary boundary layer height).

With respect to observation–model mismatch errors, just using the RMSE value of model minus observation is a convenient way, but not a correct way. I assume that "model" in L22 of P24845 means the model prediction for mixing ratios based on the a priori emission. The way inversion works is that the difference between predictions and measurements are minimized in a statistically consistent way with the two error covariance matrices. Thus if the error covariance is not properly defined, then the results are not reliable. What fraction of the observation-model mismatch is from the transport error? Was the background error included in the total mismatch error? If so, what fraction? After inversion, the RMSE is usually reduced compared to that before inversion. If I read the description in Section 2.3 as it is, the authors assign all of the average difference between "model" and observation to the random errors (i.e., the mismatch error). However, the difference between (prior) "model" and observation is also due to the lower or higher emission inventory (used as the a priori emission) than the true emission. Since the detail is not provided here, this is how I interpret L21 - L22). This important issue should be addressed.

I also wonder how the authors evaluate the uncertainty of the OH simulations from GEOS-Chem. Ideally, this uncertainty should be explicitly estimated and included in the observation–model mismatch errors

Now, with respect to background, the authors set the background to zero. However, the initial values at the domain boundary, which represents the atmospheric background, are not zero. I agree that these background concentrations (which contribute to the receptor) will dissipate in 10 or 20 days. However, what is not clear is how large the



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modeling domain is. If the authors used 20-day backward simulations to remove the effect of the background, then the back trajectories would have gone outside China. I wonder if the authors used a large enough domain such that the background effect is negligible. From the figures in the paper, the domain is very small, all regional.

Minor comments:

P24841, L25: too much use of 'crucial'

P24842, L2 - L3: sudden jump in the logic. Why spatial and temporal distribution all of sudden? Is this related to the main conclusion of this study?

L11: typo in "the most the most"

P24843, L15: From the perspective of inversion, one day in December would not make a big difference for the less than a month-long dataset. It is weak reasoning that the authors say "Data from 1 December 2010 were not used, since we focused on mixing ratios and emissions in November". There should be a better explanation to not to use the December data (e.g., poor measurement accuracy). The measurements are not enough here.

Also, in the abstract it says "a rural site". Is it only one site? In Section 2.1, it says "atmospheric measurements of benzene and toluene at two sites were used," It looks like the Mt. TMS site was not used for inversion. The authors need to be clear about this at the beginning of this measurement section.

P24848, L18 - L20. How small? Is there a reference or supporting material?

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 24839, 2015.