

The authors thank the referee for the constructive comments. Our replies to the comments and our actions taken to revise the paper (in blue) are given below in yellow color.

Anonymous Referee #3

The authors have conducted inverse modeling to estimate benzene and toluene emissions in PRD and HK for November 2010. The inversion was based on the PRIDE---PRD2010 campaign. They extrapolate the November estimate to the annual and compare with existing bottom---up estimates. The paper is well written and considering the limited number of inverse modeling studies focused on these species, I recommend the publication after some minor revisions. I have several specific comments, followed by some minor ones.

Specific comments

Echoing one of the other reviewers, why one measurement site (Heshan) was chosen for inversion, while the other one (Mt. Tai Mo Shan) was used for validation needs to be explained better in the text. I also feel that what is suggested by this reviewer (including both measurements as well as using only the Mt. Tai Mo Shan measurement in inversion) would be extremely helpful for the reader.

Response: We also tried a 2-site inversion, and we found the a posteriori benzene emissions for PRD was 4.6 Gg/month, compared to the 4.0 Gg/month from the only-Heshan inversion. Thus, the difference is only about 15% which is within the a posteriori uncertainty. The reasons why Mt. TMS was not used as well in the inversion are those 1) measurements at two stations are calibrated in difference scales so that using two data sets in one inversion is not a scientific approach, according to the review paper by Weiss and Prinn (2011), 2) the number of measurement data at TMS (totally 75 in Nov. 2010) is much less than that at Heshan (totally 419), which means limited help from adding TMS station into inversion, and 3) TMS is relatively close to central Urban Hong Kong (~7 km given by Guo et al. (2013)) so that the TMS is likely of a high risk to be influenced by relative local sources.

Authors have used the term "RCP 3PD", but in my opinion this scenario is better known as "RCP 2.6". I would recommend using RCP 2.6 instead of RCP 3PD in the paper. I believe the authors should also explain why they have chosen this specific scenario versus others (RCP 4.5, etc.). It would also be interesting to check if using different emissions scenarios as their prior has any impacts on posterior estimates, especially because background mixing ratios are considered to be zero. For some species, RCP 2.6 provides the highest emissions estimates in 2010 and that might be the case for the species considered here.

Response:

We agree that "RCP 2.6" is better known than "RCP 3PD", although "RCP 3PD" is also a correct way to call this inventory. Thus, in our revised manuscript, we have changed it to "RCP 2.6" in texts and figures. Yes, there are four scenarios in RCPs. We just chose one of them. The choice of RCP specific scenarios influences our inversion only marginally, since the RCP emissions were only used as emissions outside of China (emissions in PRD and rest of China use MEIC v1.2 and Yin et al. (2015)). Benzene and toluene emissions outside of China could hardly reach observation stations in our study.

In terms of the prior emissions, why did the authors use the averaged emissions using MEIC v1.2 and Yin et al. (2015) for toluene for the PRD region? Did the posterior emissions change much if only MEIC

v1.2 was used instead? And why was this approach not used for benzene? I feel that a similar type of sensitivity analysis using different emissions inventories might be more interesting, in addition to changing simulation length and chemical loss as currently done in the paper.

Response: According to the comments, we tested the impact of the choice of a priori emissions. A posteriori emissions for PRD from inversions using MEIC v1.2, Yin et al. (2015) and the averaged emissions using MEIC v1.2 and Yin et al. (2015), respectively, were 13.8 Gg/month, 11.1 Gg/month and 12.0 Gg/month. Thus the difference is less than 15% which is not a big value and within the a posteriori uncertainty reported in this study. We also did the tests for benzene, and we found the difference of a posteriori emissions is about 10% using different a priori emissions. The estimated emissions for toluene by MEIC v1.2 and Yin et al. (2015) are quite different, thus we used the averaged value as the a priori emission; while the estimated emissions for benzene by MEIC v1.2 and Yin et al. (2015) are relatively close, thus we used one of them (MEIC v1.2 used in our study).

I am also curious if the inversion is conducted for the two species together or separately. I would also like to see a better justification as to why 100 and 70% were chosen for prior emissions uncertainty for benzene and toluene, respectively. Also, it would be good to include an equation showing how the posterior uncertainty is calculated.

Response: The inversion is conducted separately for benzene and toluene. As for the a priori uncertainty, we determined it by looking at the variations of different bottom-up estimates for that particular species. For benzene, our priori emissions were 3.1 Gg/month, while the RCP 2.6 was 3.7 Gg/month, Yin et al. (2015) was 4.4 Gg/month, REAS v1.1 was 0.7 Gg/month. Thus, the largest deviation is about  $1 - 0.7/3.1 = 80\%$ , and then we set the a priori uncertainty as 100%. For toluene, our priori emissions were 11.5 Gg/month, while the RCP 2.6 was 3.6 Gg/month, Yin et al. (2015) was 5.2 Gg/month, REAS v1.1 was 3.8 Gg/month and MEIC v1.2 was 14.9. Thus, the largest deviation is about  $1 - 3.6/11.5 = 69\%$ , and then we set the a priori uncertainty as 70%.

Authors wrote on p. 24848 “Notice that systematic errors in the simulated chemical loss would lead to smaller errors (i.e., compared to the extreme case of no loss) in the a posteriori emissions retrieved by the inversion,” but I am not sure where I can see this change in the systematic errors.

Response: Systematic errors were not analyzed in our paper and in our revised manuscript, we have deleted this sentence.

The authors mentioned in the introduction that the two major source sectors of benzene and toluene are industry and road transport. Is it possible to assess which emissions estimates need improvement in the bottom-up estimates from your inversion? Is it possible to conduct inversion at the source sector level?

Response: it is not possible to assess the emissions at source sector level, since the emissions estimated by the inversion is at grid cell scale. Emissions from different sector could not be distinguished within one grid cell.

Minor comments

1. l. 11, p. 24842. “the most the most” the most

Response: We have incorporated this change in our revised manuscript.

2. The dark blue color used for PRD is very difficult to see on Figure 1

Response: It is fine for reading.

3. The authors state on p. 24849 that “Figure 4 shows the benzene a priori and a posteriori emission fields, their differences and uncertainty reduction. The a priori fields show that emission hot spots are located in Guangzhou, Shenzhen and Hong Kong megacities.” I think it would be helpful if you can show these locations on the map.

Response: In our revised manuscript, the cities are labeled in Figure 1 with a larger text font size.

4. Why is the correlation so much worse for toluene compared to benzene?

Response: The reasons are complicated and not clearly known. The reasons may be those 1) toluene has relatively much shorter lifetime and more complicated atmospheric reactions that may not be well simulated by the model, 2) more variations of diurnal and weekday/weekend emissions, 3) some emissions sources not far from the observatory which could not be well distinguished by the 0.25 degree grid cells.

## Reference:

Guo, H., Ling, Z. H., Cheung, K., Jiang, F., Wang, D. W., Simpson, I. J., Barletta, B., Meinardi, S., Wang, T. J., Wang, X. M., Saunders, S. M., and Blake, D. R.: Characterization of photochemical pollution at different elevations in mountainous areas in Hong Kong, *Atmos. Chem. Phys.*, 13, 3881-3898, 10.5194/acp-13-3881-2013, 2013.

Weiss, R. F., and Prinn, R. G.: Quantifying greenhouse-gas emissions from atmospheric measurements: a critical reality check for climate legislation, *Philosophical Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences*, 369, 1925-1942, 10.1098/rsta.2011.0006, 2011.