

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 #1

General comments

This paper provides estimates of Benzene and Toluene for the Pearl Delta Region and Hong Kong in China, using an inversion method. To the author's and my knowledge, these are the first top-down estimates produced for this region. The author uses data from two sites in this study: data from the Heshan site was used within the inversion, whereas data from the Tai Mo Shan site were used for model validation. Emission estimates were made for the month of November 2010. Estimates were compared to various bottom-up estimates for this month and also for annual estimates. I would recommend this paper for publishing once to comments below have been addressed. The paper is well structured and written clearly. Some figures are too small to be able to see the details discussed in the paper however. More detail about the uncertainties associated with the setup and with the emission results are needed. This study could be strengthened further with an additional discussion into where the bottom-up inventories could be limited / inaccurate (see comments below) – if this information is available to the author.

Response:

In our revised manuscript, the figures have been enlarged and made clearer, especially the Figure 1.

More detail about the uncertainties associated with the setup and with the emission results have been added, e.g., "The a posteriori uncertainty of the emissions in each grid cell was calculated as described by Seibert et al. (2011), and the uncertainty reduction in each grid cell represents the difference (as a percentage) between the a posteriori and priori emission uncertainties in the corresponding grid cell" in the section 2.3.

As for the "inventories could be limited / inaccurate", right now, it is hard to say "which sources may be incorrectly", mainly because the activity data, emission factors, source profile data behind the bottom-up estimates are not shown. Thus, we cannot say which source is incorrect for sure. Nevertheless, our study gives the light that bottom-up estimates need to be improved/investigated.

Specific comments

Page 24842 Lines 15-17: Are the minor sources for both benzene and toluene or just toluene? I think this just needs to be made clearer.

Response: They are for both benzene and toluene, according to the study by Ou et al. (2015). In our revised manuscript, to be clearer, we have changed to "while minor sources for both benzene and toluene include stationary combustion, gasoline evaporation, biomass burning, etc. (Ou et al., 2015)"

Page 24843-4 Section 2.1: I would describe both sites using m.a.s.l as well as describing that the Heshan site is 60 m.a.g.l. It is easier to understand the differences between the two sites. Is the TMS site also rural?

Response: We agree with the comments. In our revised manuscript, we have added “; ~100 m above sea level” in the section. The TMS site is at an elevation of 640 m above sea level and rural.

Page 24844 Section 2.2: I think the relationship between the FLEXPART model and how emission sensitivities are produced needs to be described in further detail here. Otherwise, it’s difficult for readers to understand how this is produced. For example, how do particles being measured backwards in time, driven by meteorological data, produce the emissions sensitivities with units $\text{s g}^{-1} \text{m}^2$? I realise this is explained in Stohl’s paper but this is a key part to understanding how your modelled observations are synthesised and should therefore be explained within this paper.

Response: In our revised manuscript, we have added “In FLEXPART, the trajectories of tracer particles are calculated using the mean winds interpolated from the analysis fields plus random motions representing turbulence (Stohl and Thomson, 1999). The emission sensitivity value in a particular grid cell is proportional to the particle residence time in that cell (Seibert and Frank, 2004). Residence time is specifically for the layer from the surface up to a specified height in the planetary boundary layer (100 m used by this study and previous studies, e.g., Stohl et al. (2009))” in section 2.2.

Page 24844 Lines 22-23: Is there any particular reason why you chose a RMSE cost function? How are modelling uncertainties incorporated into this cost function?

Response: Observation-model mismatch errors (including transport model errors) are determined as the root mean square error (RMSE) of the model-observation mismatch (Stohl et al., 2009; Stohl et al., 2010)

Page 24848 Line 25: ‘between the simulations and the observations’ – do you mean the agreement between the a posteriori simulations and the observations are better than the a priori simulations and the observations? If yes, make this clearer.

Response: Yes. According to this suggestion, in our revised manuscript, we have added “the agreement between the a posteriori simulations and the observations are better than the a priori simulations and the observations” into this sentence.

Page 24849 Line 13: The term ‘Uncertainty reduction’ should be further explained to how these values are calculated. How are uncertainties calculated within this inversion model?

Response: Thanks for this suggestion. In our revised manuscript, we have added “A posteriori uncertainty of the emissions in each grid cell was calculated as described by Seibert et al. (2011), and uncertain reduction in each grid cell represents the difference (%) between the a posteriori and priori emission uncertainties in the corresponding grid cell”.

Page 24850 Lines 10-21: Out of interest, was there a particular reason why the Heshan site data was used in the inversion and the Mt. TMS was chosen to verify? Did you try the inversion the other way

around and compare the a posteriori emissions produced? Did you try a 2-site inversion? If you have specific reasons for doing it this way, I think the readers would be interested in these reasons.

Response: We also tried a 2-site inversion from which we found that the a posteriori benzene emissions for PRD were 4.6 Gg/month, compared to the 4.0 Gg/month from the only-Heshan inversion (used in our study). 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 the TMS site (totally 75 in Nov. 2010) is much less than that at Heshan site (totally 419), which means limited help from adding measurement data at the TMS site 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.

In our revised manuscript, we have added these sentences in section 2.1.

Page 24853 Line 6: Suggested re-phrase – ‘have not changed much’ to ‘have remained relatively stable’

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

Page 24853 Paragraphs 2-3: A suggestion, if you know how the bottom-up inventories differ to each other you could draw some potentially useful comments about which sources may be incorrectly represented.

Response: Right now, it is hard to say “which sources may be incorrectly”, mainly because the activity data, emission factors, source profile data behind the bottom-up estimates are not shown. Thus, we cannot say which source is incorrect for sure. Nevertheless, our study gives the light that bottom-up estimates need to be improved/investigated.

Page 24854 Section 3.6: Would be worth also suggesting multiple site inversion analysis, with more thorough measurement data, to better constrain the inversion. For example, many US and global studies have incorporated more than one measurement site within their analysis.

Response: Yes, we agree with this suggestion. In our revised manuscript, we have changed to “then the major emission sources in PRD and HK regions could be “viewed” from different angles (multiple-site inversion)”. Given the short lifetime of toluene, to optimize fluxes over a region, the sites need to be sufficiently close so that they receive a significant signal, e.g. sites in the US probably would not help optimizing fluxes in China.

Figure 1: I think the place labels are too small and should be made bigger, especially as you refer to these place names throughout the paper. The units of the scale are also small and should maybe be increased to the same size as the equivalent units in Figure 2.

Response: We have incorporated these changes in our revised manuscript.

Figure 3: The two maps under the time series are too small to read the text clearly.

Response: In the ACPD, the figures are small. They are fine to read in a relative large figure, if this paper is finally published in ACP.

Figure 6: The figures here are too small to look at in detail. I would suggest orientating them differently. This comment applies to the equivalent Toluene figure in the supplementary material.

Response: In the ACPD, the figures are small. They are fine to read in a relative large figure, if this paper is finally published in ACP.

Figure 8: The arrows needs to be explained in the figure caption. Explain how their gradients are determined and what they are showing. If they are not scientifically determined (i.e. through regression) then I do not think they add much to the figure and would suggest removing them.

Response: In our revised manuscript, we have added an explanation of arrows in Figure 8.

Technical corrections

Title: 'in Pearl River Delta' to 'in the Pearl River Delta'

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

Page 24841 Line 6: 'in Pearl River Delta' to 'in the Pearl River Delta'

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

Page 24842 Line 7: 'Guangzhou, Shenzen in PDR, and Hong Kong are' to 'Guangzhou and Shenzen, located in the PRD, and Hong Kong are' : : : Otherwise it reads like Guangzhou is in a separate regions to the PRD and HK.

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

Page 24842 Line 19: add a comma (,) after 'For example'

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

Page 24842 Line 14: 'In PRD' to 'In the PRD'

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

Page 24842 Line 11: delete 'the most' as it's written twice

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

Page 24843 Line 15: 'period at Heshan site' to 'period at the Heshan site'

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

Page 24849 Line 9: 'of PRD' to 'of the PRD'

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

Page 24849 Line 12: 'of PRD' to 'of the PRD'

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

Page 24850 Line 1: 'in PRD' to 'in the PRD'

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

Page 24850 Line 3: 'for PRD' to 'for the PRD'

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

Page 24852 Line 1: 'in PRD' to 'in the PRD'

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

Page 24852 Line 10: 'in PRD' to 'in the PRD'

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

Page 24853 Line 18: 'in PRD' to 'in the PRD'

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

Page 24853 Line 22: 'in PRD' to 'in the PRD'

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

Page 24854 Line 5: 'in PRD' to 'in the PRD'

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

Page 24854 Line 17: Remove 'in the future' It is not needed when using the term 'are urgently needed'

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

Page 24854 Line 19: 'in PRD' to 'in the PRD'

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

Page 24854 Line 21: 'in PRD' to 'in the PRD' and 'of PRD' to 'of the PRD'

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

Page 24855 Line 6: 'in PRD' to 'in the PRD'

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

Page 24855 Line 10: 'in PRD' to 'in the PRD'

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

Page 24868 Line 6: 'for PRD' to 'for the PRD' and 'within PRD' to 'within the PRD' (Figure 6)

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

Reference:

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- 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.