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Interactive comment on "Evaluating BC and NO$_x$ emission inventories for the Paris region from MEGAPOLI aircraft measurements" *by* H. Petetin et al.

H. Petetin et al.

hervepetetin@gmail.com

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Answer to referee #2

We would like to thank the referee for its good appreciation and its useful general comment about the organization of the paper. In the following, we explain the major changes applied to the table of contents, and provide answers to the specific comments. Note that, due to a significant rearrangement of the paper, not all (but most of) small modifications are indicated here, and we refer the reader to the new version of the paper.

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General Comment:

The study have performed nice analysis method and followed all the precautions for evaluating the model with observations. In principle, they have done extensive work. The content of the paper and novel methodology is worth publication in ACP. The short-coming of the paper is that manuscript is not easy to read and contain too many details about the model observation comparison, too many subsections which many time confusing and lengthy and should be avoided. I am not sure how to reduce so many sections and subsections but composition of paper need signiïňĄcant overhaul and rearrangement.

ANSWER : In its new version, the paper is largely rearranged in order to simplify its reading :

- Section 3.5 : To our opinion, the content of Sect. 3.5 ("Black carbon/elemental carbon terminology") is worth staying in the paper as modeling studies usually do not pay enough attention to this point. For clarification, the section is removed, and its content is moved to previous sections : the first paragraph (reminding the recommendations of Petzold et al. (2013)) is moved in Sect. 3.1 ("Measurement data base"), and the second one (reminding that emission inventories are expressed as EC which does not exactly correspond to the observed EBC) is moved in Sect. 3.2 ("Emission inventories")

- Section 4.1 : Sect. 4.1.1 ("Surface observations") and 4.1.2 ("Observations in altitude") are changed into (bold) paragraph titles (which simplifies the reading, to our opinion) and introduced by : "In this section, meteorological input data used in CHIMERE simulations, with both MM5 and WRF models, are evaluated against observations at surface and in altitude.".

- Section 4.2 : For clarity, Sect. 4.2 is renamed into "Approach n°1 : emission evaluation from surface measurements". In this section, the CHIMERE evaluation of BC, NOx and BC/NOx (p29254/L19-p29255/L28) and the Table 4 (statistical results) are moved into the Supplement, in order to focus on the evaluation of BC emissions relatively to

NOx ones. The initial text is replaced by : "Details on the evaluation of CHIMERE against observations and statistical results are given in Sect. S.3 in the Supplement. In a few words, BC is strongly overestimated, in particular with the TNO inventory and during BL transitions, the use of WRF reduces biases mainly during the late afternoon (after 18:00 UTC). NOx is also overestimated, but mainly during the end of the day. BC/NOx ratios are rather constant (0.06 μg m-3 ppb-1 in average) in July except during some nights, but with a diurnal pattern showing lower values around 5 UTC and higher ones around midnight. CHIMERE also simulates rather constant ratios but with a positive bias with TNO and to less extent in EMEP inventories, while bias with TNO-MP emissions is rather small (< 13%).". Next paragraphs dealing with the emission evaluation are introduced by : " We now evaluate in some more detail BC emissions relatively to NOx ones. " (p29256/L1)

- Section 4.3 : For clarity, Sect. 4.3 is renamed into " Approach n°2 : emission evaluation from airborne measurements ". The discussion in Sect. 5 is lightened and moved into Sect. 4.3, after the discussion on the variability in observations. Fig. 15 is moved to the Supplement. Sect. 4.3.2 and 4.3.3 are combined into one section (Sect. 4.3.2) : "Results on emission errors factors" with bold paragraph titles. A section 4.3.3 entitled "variability in observations" is created, that includes the discussion on the regional background heterogeneities (p29260/L21-28) and the time window of emission sampling (previously in Sect. 4.3.4)

- Section 4 : We added in the introduction of Sect. 4 (p29251/L23) : "In this section, we first evaluate meteorological input data (Sect. 4.1). A first simple approach is then applied to evaluate BC emissions against NOx ones, based on ground based measurements at the urban background LHVP site in Paris (approach n°1, Sect. 4.2). We then describe the procedure to evaluate BC emissions based on airborne measurements in the Paris plume, and present the corresponding results (approach n°2, Sect. 4.3). We finally discuss discrepancies between both methods (Sect. 4.4)."

- Section 5 : see comment on Section 4.3

Finally, the table of contents is as follows :

Concerning tables and figures :

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- Figures 2, 3, 5, 10 and 15 are moved to the Supplement.

- Figures 13 and 14 are gathered into one (and become Fig. 9a and 9b).

- Table 1 (description of domains) is removed and its content simply added in the text : "Two nested domains of increasing resolution - CONT3 (0.5 x 0.5°, i.e. ∼50 x 50 km, 67x46 cells) and MEG3 (0.04 x 0.027°, 120x120 cells) - are considered (see Fig. S4 in the supplement)."

- Tables 3, 4 and 6 are moved into the Supplement.

Some other modifications are applied (related to this rearrangement):

- In the introduction (p29244/L2) : "Results from both ground and airborne measurements are shown and discussed in terms of representativeness in Sect. 4. The various uncertainty sources are discussed in Sect. 5." is replaced by "Results from both ground and airborne measurements are discussed and compared in Sect. 4."

- In the conclusion, (p29272/L20-22) "However, these results are judged as representative only for an area surrounding the LVHP site in a few kilometers of distance." is moved and replaced by (p29273/L17) "Results obtained at a ground based site in Paris are not consistent with those obtained in the plume, due to the fact that surface measurements are representative only for an area surrounding the LVHP site by a few kilometers while emissions from the whole agglomeration are sampled in the Paris plume."

Specific Comments:

The paper covers an important and interesting topic. Evaluating BC and NOx emission inventories from the urban center using aircraft measurements. This study evaluates BC and NOx emissions from the Paris city using aircraft measurements across the city plume as well as using measurements at ground site. Authors have considered the emission inventories EMEP, TNO and TNO-MP. Further they have used CHIMERE chemical transport model to simulate the emission Plume over the Paris region to eval-

uate the BC and NOx emissions from these emission inventories. Finally authors have shown that BC emissions in EMEP and TNO, and NOx emissions in TNO-MP, is over-estimated over the Paris region. This paper is definitely a first step in achieving the objectives the authors have set up to achieve. My overall recommendation is accep-tance after careful revision of the text and queries as under.

1. The manuscript is not easy to read and contain too many details. I would suggest to cuts-sot some of these fine details (or move in supplementar y material) and focus on the objective of the manuscript.

ANSWER : See answer to general comments.

2. In addition to advantage, author should also discuss the demerit of this approach in the abstract as well in conclusion section to put a transparent balance picture to readers.

ANSWER : We add the following sentence in the abstract (p29239/L14) : "Large uncer-tainty values are determined in our results, which limits the usefulness of the method to rather strongly erroneous emission inventories."

3. Authors have shown that compared to MM5, WRF meteorology shows better agree-ment with the observation shows. Why authors have done 16 tracer experiments with MM5 meteorology? This need clarification in text and justification required.

ANSWER : The main differences between MM5 and WRF concern the representation of the boundary layer, and more specifically : (i) the higher underestimation of MM5 in the afternoon maxima, and (ii) its much too early evening transition. It should be men-tioned that discrepancies between MM5 and WRF are moderate between 00:00-10:00 UTC, i.e. the period of interest in this tracer analysis. They start to increase at 10:00 UTC, but this is likely not so important in our analysis since we are investigating the rel-ative (and not absolute) contribution of each tracer. We add in the paper (p29262/L14) : "Note that the use of WRF is not expected to substantially modify the results ob-

tained here with MM5 since major discrepancies between both meteorological model outputs only concern the BLH starting from 10:00 UTC and that emission tracers are here investigated relative to each other."

4. Page 29243, L25: The aim of the paper has not come out properly. Composition of the paper is discussed but the major objective of paper should come out in 1-2 sentences to sum-up the introduction section.

ANSWER : We replace the first sentence of this paragraph (p29243/L18-20) by : "The aim of this paper is to evaluate emission inventories at the scale of a large city. In this frame, it presents an original methodology based on airborne measurements in the city plume and chemistry-transport simulations."

5. Are different PBL schemes are used during MM5 and WRF simulations? Which PBL scheme is used? Uncertainty, will also be introduced due to PBL scheme, authors should discuss the same in text with quantification?

ANSWER : Two different PBL schemes are used in MM5 and WRF, and we agree with the referee that these schemes play a major role in the model uncertainties. Two distinct meteorological input data are used in this study in order to investigate how it influences the emission evaluation results, but it is beyond the scope of this study to discuss in detail the parametrization of boundary layer. We add in the paper (p29251/L3) : "Note also that MM5 and WRF have distinct boundary layer schemes : Medium Range Forecast (MRF) for the first, and Yonsei University (YSU) for the second."

6. Section 3.5: Black Carbon/Elementar y Carbon Terminology- This section can be merged with introduction and there is no need to make it a separate section.

ANSWER : See answers to general comments.

7. P. 29254 and Fig. 6: Flight timings are different for different days and taking single day for diurnal profiles of BC, NOx and BC/NOx ratio is inhomogeneous. How it is analyzed? Few days or whole July month?

ANSWER : We are not sure to understand properly the question of the referee. Concerning the approach based on surface observations, BC versus NOx slopes were determined over the 05:00-08:00 UTC time window in order to minimize the potential influence of imports from outside the Paris megacity. Results are given for both the set of flight dates and the whole July month, and show very similar slopes (see Table 5). Concerning the flight timings, they indeed vary from one day to the other, with a shift that remains below two hours (around one hour most of time). The tracer sensitivity test shows that the time window of sampled emissions mostly depends on the wind speed. For the BC versus NOx slopes calculations at surface, it would have been possible to consider different time windows for each flight date in order to match the emissions sampled by the plane, but (i) we do not know the time window on which emissions are really sampled (the information is only available for the model), and (ii) considering a time window outside the morning rush hours (when local emissions are maximum) increases the uncertainties associated to the regional background of BC and NOx (that is removed in the approach based on airborne measurements). Note however that considering the whole July month and all hours of the day, rather similar slopes are obtained (0.039 μg m-3 ppb-1 for observations, and 0.096, 0.138 and 0.070 μg m-3 ppb-1 for EMEP, TNO and TNO-MP with the WRF meteorology).

8. 5. Uncertainty of the inversion methodology: This section should have come before the results. The result and discussion section can be combined.

ANSWER : See answers to general comments.

9. Conclusions cannot run for 2 pages. It must be shortened. I think authors can delete first para except first 2 sentences.

ANSWER : We agree with the referee that conclusion can be shortened. The first paragraph is replaced by (p29272/L2-16): "Black carbon (BC) emissions are still highly uncertain, and very few studies have attempted to evaluate their inventories. This paper presents an original approach, based on airborne measurement across the Paris

plume, developed in order to evaluate BC and NOx emissions at the scale of the whole agglomeration. It is applied to three emission inventories (EMEP, TNO, TNO-MP)."

10. Page 29273: Remove first sentence of line 28-29. ("To our knowledge, this study is one of the most comprehensive ones to evaluate BC emissions at the scale of a large megacity.")

ANSWER : Without any suggestion of other studies dealing with the BC emission evaluation at the scale of a megacity, we do think that this sentence is worth staying in the conclusion.

11. Section 3.2: Provide details of resolution of inventories in each scenario.

ANSWER : Inventory resolutions are already mentioned in Sect. 3.2 (p29247/L18 and p29248/L9), but we agree that they may be indicated more clearly. We add in the text (p29247/L13) : "The EMEP inventory (Vestreng et al., 2007), with a longitude-latitude resolution of 0.5° x 0.5°." and remove (p29248/L9) : "whose resolution is 0.5° x 0.5°" ; and we add the following precison (p29247/L23-25) : "A third inventory based on the TNO inventory and with the same 1/8° x 1/16° longitude-latitude resolution, but incorporating bottom-up emission data over the four European megacities (Paris, London, Rhine-Ruhr and Po valley)."

12. Figures and Tables: It is way too many. There are repetitive information between Figures and Tables. Author should try to combine information in various tables in 1. As for example, all error and uncertainty related information can come in one table. In my opinion, Figures 11, 12 and 15 can be avoided as information is well covered in text as well as in tables.

ANSWER : See answers to general comments. Concerning Fig. 11 and 12, they show the main results of this study and are thus worth staying in the paper as results are easier to understand on a graphical way, but we moved Table 6 (BC/NOx mean results) into the Supplement as values are discussed in the text.

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13. Figure 2: Unit of Y-axis missing.

ANSWER : The Y-axis parameter is a ratio, thus unitless. We add in the legend : "Integrated BC, NOx and BC/NOx emissions at various distances from the LHVP site, relatively to the TNO-MP inventory (unitless)." (this figure has been moved into the Supplement).

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 29237, 2014.