Atmos. Chem. Phys. Discuss., 14, C13163–C13171, 2015
www.atmos-chem-phys-discuss.net/14/C13163/2015/

Atmospheric Chemistry and Physics Discussions

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

Received and published: 18 May 2015

Answer to referee #1

We thank the referee for its positive appreciation of our work and we provide here answers to its comments.

Petetin et al. present a novel approach to evaluating BC and NOx emissions from a whole large city (Paris) based on airborne measurements of the large-scale downwind plume. The BC and NOx concentrations observed during the level flights (about 600 m a.g.l.) across the pollution plumes and an atmospheric chemistry-transport model

driven by BC and NOx emission inventories are used in this approach. To minimize several errors in the model, the integrated values of the excess BC and NOx above the background concentrations and the BC/NOx ratios are compared between the observation and the model simulations. From the comparison the BC and NOx emission inventories are evaluated. BC and NOx concentrations observed at ground site in Paris (LHVP) are also examined, and it is confirmed that the ground observation, predominantly influenced by the local emissions, is not appropriate to detect the emissions from the whole city. Petetin et al. carefully examine the sources of the uncertainties including meteorological data, vertical mixing, and analytical uncertainties, and finally find the significant biases in the BC and NOx emission inventories used in the model simulations. Although there are still relatively large uncertainties in the estimations, the proposed approach is considerably useful to constrain the whole emissions from the large city. I found that the paper is well written, the approach is excellent, and containing material that should be published. I strongly recommend this paper for publication in Atmospheric Physics and Chemistry with minor revisions described below.

General comments:

If the BC and NOx sources are collocated, BC/NOx ratio would better constrain the emission ratio because the errors associated with the atmospheric transport are minimized. However, there are some difficulties in simulating the atmospheric NOx concentrations due to the dry/wet deposition and the chemical processes. I think CO is more appropriate to constrain the BC emissions because CO is also burning process-related species, is more conservative for the relevant time scale, and is more accurately measured than NOx. Actually, the measurements of CO were conducted at LHVP (Lopez, et al., 2013, ACP, 13, 7343-7358) and during the MEGAPOLI airborne measurements (Freney et al., 2014, ACP, 14, 1397-1412). There is no need to add the CO data and the discussion in the revised manuscript, but if the authors agree with this comment, I think it would be better to mention briefly the possibility to use another species to constrain the BC emissions. If there are associated studies on the BC emissions using

CO and so on, it would also be better to add the information.

ANSWER : We agree on the fact that CO is also an appropriate (and, on some points of view, a better) candidate for the evaluation of BC emissions and should thus be mentioned in the text. However, as mentioned and discussed in Sect. 4.2, it is worthwhile noting that in the methodology based on ground measurements, some uncertainties may arise from an erroneous simulation of the regional background (even focusing on the morning rush hours). This is particularly true for CO that is characterized by high background concentrations contrary to NOx, and thus a lower contribution of local emissions to urban concentrations in the city. We thus propose to add in Sect. 4.2 (p29257/L3) : "It is worthwhile noting that, as a burning process-related species of long lifetime, carbon monoxide is another appropriate candidate for the evaluation of BC emissions (Zhou et al., 2009). However, it should also be mentioned that, due to its significant background concentrations, higher uncertainties (compared to NOx) may arise from errors in the simulation of the regional background around Paris, even considering only rush hours.". Concerning the methodology based on airborne measurements, despite a high regional background, the CO Paris plume remains well distinguishable (as illustrated in Freney et al., 2014), which would allow the evaluation of its emissions with this methodology.

As the authors pointed out, how well the model reproduces the vertical profiles is one of the important error source for the emission estimation. Although only lateral observations are examined in this study, it is mentioned that the vertical profiles up to 3 km a.g.l. were measured at the end of the several flights (P. 29257, L. 24-25). If so, I think it would be better to compare the observed vertical profiles with the simulated vertical profiles because such comparisons could allow us to more directly validate the model performance and to evaluate the model uncertainty.

ANSWER : The referee points the benefits of using the vertical profile observations performed by the aircraft for evaluating the ability of CHIMERE to reproduce vertical gradients. Indeed, in all flights of the MEGAPOLI campaign, vertical profile samplings

up to 3 km a.g.l. were collected by the aircraft, and we agree with the referee on the potential interest of such observations for estimating the uncertainties related to the vertical mixing representation. However, such an analysis has not been conducted in this study because most vertical samplings are actually performed outside the Paris plume, as illustrated in Fig. 8 (of the discussion version) for the 10 and 13 July (where vertical profiles are performed at the furthest point from Paris, on the symmetry axis of the flight trajectory). In addition, these vertical profiles were performed at the end of the flight, and thus cannot provide information on the vertical mixing closer to Paris, where uncertainties are the most important.

Specific comments

1) P. 29245, L. 3: Is "horizontal variability of the boundary layer height over the aircraft trajectory" discussed in this paper?

ANSWER : Indeed, this point has not been investigated due to the absence of measurements outside the Paris agglomeration. The sentence is modified as follows (p29245/L2-4) : "the boundary layer height that directly affects the level of concentrations".

2) P. 29245-29246, Section 3.1: Several analyzers were used to detect EC, NOx, and so on. If those analyzers are commercially available, it would be better if you clarify the model of the instrument and the manufacturer.

ANSWER : We add in the text (p29245/L21-22) : "by a Multi-Angle Absorption Photometer (MAAP, Model 5012, ThermoScientific)" ; (p29245/L24) : "chemiluminescence monitor (AC31M, Environment SA)"; (p29246/L19) : "Particle Soot/Absorption Photometer (PSAP) instrument (Radiance research)" ; (p29246/L9) : "Ecophysics (CLD 780 TH)"

3) P. 29249, L. 10: "residential/tertiary" is the description of SNAP sector 2 here, but "small combustion plants" is in Table S1.

ANSWER : The SNAP 2 "small combustion plants" refers to "non-industrial combustion plants" (e.g. fireplaces, stoves). This sector is often referred as the "residential/tertiary" sector since most the residential/tertiary emissions come from these small combustion plants.

4) P. 29250, L. 25: "Dudhia, 1993", not "Dudhia et al., 1993".

ANSWER : The correction is applied.

5) P. 29254, L. 15-16 and Fig. 6 right panel: Please clarify how to compute the diurnal profiles of BC, NOx and BC/NOx ratio. Are they the averages for the flight dates or for the all July dates?

ANSWER : This section has been changed (see answers to referee 2), including the following sentence (p29254/L14-16): "Urban background BC and NOx concentrations, their ratio and their diurnal profiles are presented in Fig. 6, considering only flight days."

6) P. 29257, L. 24-25: If the vertical measurements were conducted in the pollution plumes, I think it would be better to show the vertical profiles. Do those vertical profiles convince us of the well-mixed condition in the boundary layer?

ANSWER : See answer to the general comment.

7) P. 29258, L. 3: The value of 30 percentile is used for the background determination in this study. Does the value of the percentile affect the plume integration?

ANSWER : Several tests were performed with other values for the background determination, without strong influence on average emission error factors.

8) P. 29262, L. 8-10: These diurnal variations mentioned here with the lowest value in early morning can be also seen in Fig. 6. I think Fig. S7 in the Supplement is not needed. The lowest value of BC/NOx diurnal variation seems to lower than 0.05 micro-g m-3 ppb-1, close to 0.03 micro-g m-3 ppb-1.

ANSWER : The lowest value is indeed closer to 0.03 μg m-3 ppb-1 but the text refers

to the value over the morning rush hours (defined as 05:00-08:00 UTC), which gives a value actually close to 0.04 μg m-3 ppb-1 (and not 0.05 as in the text). Concerning Fig. S7, it is useful to show the day-to-day variability of the diurnal variations, which is not obvious in Fig. 6. Note that the paper has been rearranged following the recommendation of referee #2, and Fig. 6 and S7 become Fig. 3 and S11. The section now includes the following sentences : "Another possible source of variability in the BC/NOx emissions is related to the time window of emission sampling, as BC/NOx diurnal profiles at LHVP show much lower values during morning rush hours than in the end of the morning ($\sim$0.04 against $\sim$0.07 μg m-3 ppb-1; see Fig. 3), with a noticeable day-to-day variability (see Fig. S11 in Supplement).".

9) P. 29262, L. 22-24: The BC/NOx emission error factors for TNO inventories don't seem to be underestimated (see Fig. 12 and Table 6).

ANSWER : This was a mistake, this section has been changed (see answers to referee 2), including (p29262/L16-24) : "Results obtained at ground in Paris show an overestimation of the BC/NOx ratio in the TNO inventory and at a lesser extent in the EMEP one, while quite correct values are given by TNO-MP. This is not consistent with results obtained in the plume where the BC/NOx emission ratio appears highly underestimated in TNO-MP (while errors are lower for EMEP and TNO)."

10) P. 29279, L. 24-25: "Schmidt, H., Derognat, C., Vautard, R., and Beekmann, M.", not "Schmidt, H. and Derognat, C."

ANSWER : The correction is applied.

11) P. 29295, Fig. 6, caption: "BC, NOx and BC/NOx ratio concentration" should be "BC and NOx concentrations and BC/NOx ratio".

ANSWER : The correction is applied.

12) P. 29300, caption: "on the top right" should be changed to "on the right".

ANSWER : The correction is applied.

13) Fig. 11, Fig. 12, and Fig. 15: It would be better to add the labels of the x-axis, "July date (UTC)".

ANSWER : The correction is applied.

Additional modifications :

- (p29239/L5): "Paris plume" is changed to "Paris, France, plume"

- (p29239/L8): "error sources in the model" is changed to "error sources in the used model"

- (p29239/L13): "though" is changed to "through"

- (p29239/L17-19): "which additionally suggests potential error compensations in the BC emissions spatial distribution over the agglomeration." is changed to "which additionally suggests a spatially heterogeneous error in BC emissions over the agglomeration"

- (p29240/L13-15): "making the true forcing per unit emitted uncertain" is changed to "making the true forcing uncertain"

- (p29245/L18-19): "have been performed Paris at the LHVP (Laboratoire d'Hygiène de la Ville de Paris) station (48.829°N, 2.359°E) (urban background site in the center of Paris). " is changed to "have been performed at the LHVP (Laboratoire d'Hygiène de la Ville de Paris) station (48.829°N, 2.359°E), an urban background site in the center of Paris. EC "

- (p29256/L4-5): "when its photolytic conversion into HNO3 or HONO is not active" is changed to "when its photolytic conversion into HNO3 or HONO is less active"

- (p29256/L20-21): "Results reported in Table 5 show a high overestimation for the TNO inventory, around a factor of 4." is changed to "Simulated slopes of BC versus NOx reported in Table 2 show a high overestimation with respect to observed ones for the TNO inventory, around a factor of 4."

- (p29256/L26-27): "whose biases remain below +136% " is changed to "for the latter biases remain below +136%"

- (p29257/L9): "the TNO/MM5 case as well as two flights" is changed to "the TNO/MM5 case for two flights"

- (p29261/L17): "for which wind speed at higher levels is among the lowest" is changed to "for which observed wind speed at higher levels (110-210 m a.g.l.) is among the lowest "

- (p29264/L15-16): "The methodology does not evaluate emissions alone" is changed to "The methodology does not evaluate annual monthly emissions alone"

- (p29264/L19): "temporal emission gradients are important" is changed to "temporal emission gradients are strong"

- (p29265/L9): "shift that time window" is changed to "shift this time window"

- (p29264/L19): "This error source thus appears all the more important that the gradient in the diurnal proïňĄle sampled part is high." is changed to "This error source thus appears all the more important that the gradient in the diurnal emission profile in the sampled time window is high."

- (p29269/L5-8): "Considering the previous MAC estimations in the Paris region - 7.3 and 12.0 m2 g-1 by Sciare et al. (2011) and Liousse et al. (1993), respectively - the uncertainty associated to our MAC value (8.8 m2 g-1) is roughly estimated at 30%. "is changed to "Considering the previous MAC estimations in the Paris region - 7.3 and 12.0 by Sciare et al. (2011) and Liousse et al. (1993), respectively - the uncertainty associated to our MAC value is roughly estimated at 30%."

- (p29269/L28): "a combination of all the uncertainties" is changed to "a combination of all the systematic uncertainties"

- (p29270/L3): "Confidence intervals on average emission error biases" is changed to

"Confidence intervals (at a 95% confidence interval) on average emission error biases"

- (p29263/L2): "a simulation with traced emissions" is changed to "a simulation with spatially traced emissions"

- (p29263/L14): "the Paris ring" is changed to "the Paris ring road"

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