Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-923-AC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of the Paris NO_x Emissions from mobile MAX-DOAS observations and CHIMERE model simulations using the closed integral method" *by* Reza Shaiganfar et al.

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Received and published: 18 April 2017

Reply to reviewer #2

Before we respond to the individual comments of the reviewer we give a short overview about the most important changes compared to the previous version fo our manuscrip:

A) The diurnal cycle of emissions (Fig. 2) was corrected: local time => UTC. Accordingly, the upscaling to the daily average emissions was corrected and Figures 14 – 17, Fig. A6, Table 4, and the text were updated. The new upscaling caused slight changes compared to the previous version: => consisteny of Chimere emission in/out

is enhanced => overall most daily averaged values decreased

B) A discussion about 'special gaps' was added to section 4.2. Such gaps are characterised by large differences between the start and end points of a circle. An example for such a measurement (from 4 February 2010) was added to Fig. 4 (right). The following text was added at the end of section 4.2: 'In Fig. 4 (right) an example for measurements without an obvious gap is shown. However, on that day a large difference between the NO2 VCD between the start and end locations of the circle is found indicating that during the period of the measurements the NOx distribution around the location of the maximum outflux has changed significantly. Obviously, the NOx emissions derived from these measurements are subject to large uncertainties and are thus also skipped from the set of measurements considered for the comparison to the input emissions (section 6)'.

C) We added more discussion on the reasons for discrepancy between input emissios and car-MAX-DOAS results. Here two (related) aspects are important: -the rather high day to day variability of the car MAX-DOAS results -the enhanced seasonal cycle of the car MAX-DOAS results. We discuss both points in detail now in the conclusions. There the followoing text was added:

'Here it is interesting to note that a high day to day variability was also found by Petetin et al. (2015). For most of the measurement derived emission results, the day to day variability is within the range of the uncertainties, especially in summer. Thus we conclude that this variability simply reflects the uncertainty range of the measurements. However, for several days at the end of the winter measurement campaign on mid-February, significantly enhanced values were found compared to the other winter days. These days are also the reason for the rather high average values derived from the car MAX-DOAS measurements in winter. If these days are excluded, a similar ratio (1.4) of the NOx emissions derived from car MAX-DOAS or CHIMERE as in summer (1.5) is found. Interestingly, for these days the temperature was low (-4°C to –1°C) indicating that the high emissions might be related to these low temperatures (see Fig. 18) The

following effects might be responsible for enhanced NOx emissions on cold days: a) Residential heating According to Fig. 2 domestic heating contributes about 25% to the total NOx emissions in winter. If one assumes a factor of two variability between cold and warm (less cold) winter days (see e.g. Terrenoire et al., 2015), it becomes clear that the variability of the NOx emissions from residential combustion alone can only explain a part of the increase of about a factor of two found for the cold days. b) Temperature dependence of catalytic converters During winter time, NOx emissions from traffic contribute about a half to the total NOx emissions. Under cold conditions, three way catalytic converters for gasoline cars work less well, and they take longer time to reach to an optimized way of working for diesel cars (the cold start effect). It is probable that this effect leads to increased NOx emissions on cold days, but this additional emission is difficult to quantify. c) It is known that in the past during cold periods an older 250MW coal-fired power plant was temporarily restarted to meet the additional demand for electrical heating in the city. Several other fuel or gas driven combustion turbines can also be activated during periods of increased energy demand. On an annual basis such temporarily operating facilities would not add much to the annual total emissions but during episodes it could be important. Instead of being spread out over the year, the emissions would have to be allocated to a much smaller number of operation days causing the emissions during selective periods to be much higher than annual averaged, and on other moments to be zero. Unfortunately, we have no access to operation days for such facilities and cannot confirm that this contributed also during the February episode discussed in this paper.'

In this paper, Shaiganfar et al. Report on a series of car-based DOAS measurements of NO2 around Paris which they use to estimate NOx emissions of that city. The manuscript describes the measurements and approach to emission estimation and discusses the different contributions to the overall uncertainties of the derived NOx fluxes. It then applies the same flux estimation method to simulations of the CHIMERE model using the sampling of the measurements and compares the results to the integrated emission flux used in the model. Finally, emissions are derived from the measurements

on 18 days and compared to the emissions from the TNO / AirParif inventory. The paper is clearly structured, well written, reports on an interesting type of measurements and provides relevant emission estimates for Paris. The detailed error discussion provided is important for the application of similar measurements in other regions and will be useful for future measurements. My only major concern with this paper is that it mainly discusses the method used and its uncertainties and spends little time on the results and their implications. A journal such as AMT would therefore have been a better place for this manuscript. I therefore recommend publication of this paper only after strengthening the results and discussions part.

Author reply: We thank the reviewer for the positive assessment. We are aware of the fact that a large part of the paper describes technical aspects of the car-MAX-DOAS measurements. We therefore also asked ourselves whether submission to AMT would be more appropriate. However, in addition to the technical aspects, the paper provides the first detailed comparison of the experimentally derived NOx emissions from Paris to existing emission inventories and model simulations (during extended measurement campaigns). We regard the comparison results as important information for a wider community than only the measurement experts. Thus in our opinion publication in ACP is well justified. In the revised version we spend more emphasis on the comparison results and discuss in more detail possible reasons for the discrepancies between the experimental results and the existing emission inventaries (see point C above).

Major point

The paper reports measurements of NOx emission fluxes for Paris on 18 days and compares them to the TNO / AirParif emissions. The results as shown in Fig. 17 indicate good agreement between the two quantities on many days, but also large differences on other days. In particular in January / February, the car-DOAS based estimates show large day-to-day variations and much larger values than the emission inventory. This raises two questions:

1. Is it plausible that NOx emissions in Paris change by a factor of two between January 19 and February 11? The emission inventory suggests the same value for both days, and considering the fact that traffic is the dominant NOx source in Paris, what could be the origin of all the additional NOx? Or is this a problem of the measurements / method? The latter is not suggested by the results of the application of CIM to the model data, so this is a bit of a mystery.

Author reply: We investigated possible reasons for the high values at the end of February and added a detailed discussion to the conclusions section (see general point C above).

2. Is it realistic that the TNO / AirParif NOx emission inventory is off by a factor of three as it appears from the last 4 days of measurements shown? I think these two points deserve more discussion and analysis.

Author reply: As stated above, these high values occur only during the cold period in mid February. For other days, the differences are much smaller (10% to 50%). Thus our conclusion is that changes in emission sources due to the cold temperatures are the most probable reason for the discrepancies (see general point C above)

Minor points

âĂć Section 3, line 9: The Figure in the Appendix referenced to does not exist

Author reply: Many thanks for this hint. We removed this sentence from the text.

âĂć Section 4.1, line 19: Wrong Figure number in Appendix

Author reply: The number was changed to 'A2'

âĂć Section 4.1, line 25: Wrong Figure number in Appendix

Author reply: The number was changed to 'A1'.

âĂć Section 4.3, line 26: Is it expected that the emission flux depends on the largest

values? And isn't that maybe a problem indicating that the car DOAS measurements are affected by close-by local sources more than they should?

Author reply: It is true that close to emission sources high concentrations (and also VCDs occur. But this is not a problem, because the potemtially high concentrations (VCDs) are exactly balanced by their small spatial extent.

âĂć Section 4.3: Please mention and briefly discuss somewhere that you apply partitioning (and life time correction) to columns although strictly speaking this is something to be done on height levels.

Author reply: In section 4.3 (partitioning correction) we added the information that the partitioning ratio from the model was calculated from the respective VCDs. Thus it actually is representative for the VCD. In section 4.4 (lifetime correction), we changed the text to: 'Here it should be noted that these lifetimes are rough assumptions, and on individual days large deviations from the assumed values might occur. Moreover, in a strict sense separate lifetime corrections should be applied for individual height layers. But especially for wind speeds above about 2 m/s, the effect of the limited lifetime of NOx and thus of the uncertainties of the assumed lifetimes are small (the correction factor is close to unity)'.

âĂć Section 4.6.1., line 19: simply => simple

Author reply: corrected

âĂć Section 4.6.3., line 19: Last sentence of paragraph is unclear, please reformulate

Author reply: corrected (account => accounted)

âĂć Section 5: The definition of times for CIM application to the CHIMERE data is unclear to me – why did you not just use the times of the measurements? Using the time of the maximum measurement appears arbitrary to me but I may be missing the important point here. Please explain.

Author reply: We added the following information to the text: ‘By selecting this time period we take into account the average travel time of the pollued air masses until the location of the measurement.’

âĂć Section 5 last paragraph: I find the discussion of weekend effects confusing – in Fig. 17. We can clearly see the weekend effect In the emissions but not in the CHIMERE.CIM values. Therefore, this is not a result of changes in domestic heating but just random uncertainties introduced by the method and sampling. Please re-consider.

Author reply: We added the following information to the text: ‘. . ..not the lowest emissions are found, indicating that the variation of the NOx emissions derived from car MAX-DOAS is not dominated by the weekend effect.’ We deleted the statement about the domestic heating in this section. The possible influence of domestic heating is discussed in more detail in section 6.

âĂć Section 5, last lines: What is the logic of only showing data with small differences between TNO and CIM values here? I could understand if only data without obvious problems were used, but the other values should appear in this figure in my opinion. Please re-consider.

Author reply: We agree that in this figure this selection makes no sense. Therefore we included all data in the updated figure 15. inter-friendly versioniscussion paper âĂć Section 6, last paragraph: It is noted twice that a similar ratio is found between CHIMERE VCs and observed VCs on the one hand and the emissions on the other hand. I think this is to be expected considering the way the emissions are determined from the columns which assumes a nearly linear relationship (excluding life time and partitioning corrections).

Author reply: We deleted this staement at the end of section 6. However, we prefer to keep this statement in the conclusions, because this consistency between the comparison of the CIM results and the direct comparison of the NO2 VCDs serves as a

consistency check. In the conclusions we added the following information to the text: ‘. . .indicating that the differences between the measurements and the model simulations are not caused by the application of the CIM.’

âĂć Conclusions and perspectives: This section is mainly a summary and in parts identical to the abstract. As mentioned in the major comment, I think more focus should be on the results.

Author reply: We discuss the discrepancies between the experimental results and existing emission inventories in more detail. (see reply to general point above)

âĂć Conclusions and perspectives: I do not agree with the statement, that the large number of measurements was used to test the applicability of CIM under various atmospheric conditions. Actually, all the tests were performed on the model data which could have been done without measurements by just assuming certain measurement routes and patterns. The data themselves are only used for emission estimates which is of course very interesting.

Author reply: We agree that the important aspect here is that a large number of both car DOAS measurements and model simulations were available during the megapoli campaign. Thus we added ‘(together with the model results)’ after ‘the large number of measurements’.

âĂć Figure 2: I’m surprised that I cannot see the effect of daylight saving time in the diurnal emission pattern

Author reply: We thank the reviewer for this hint, which pointed our attention to a mistake we made (we mixed local time and universal time). The figure is corrected in the updated version of the manuscript (the time shift between summer and winter is now clearly visible).

âĂć Figure 2: Are the emission values in the map given per 3 x 3 km2 pixel? In the caption, it is said that they are averaged over this area but I assume they are summed

up?

Author reply: The text was changed to ‘summed up’.

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