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# *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.

**Reza Shaiganfar et al.**

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Reply to Reviewer #1

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



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Discussion paper

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.'

General Comments I believe that this paper is highly acceptable and appropriate for publishing in ACP. This paper substantially contributes to scientific progress in the new field of mobile-MAX-DOAS, a useful new tool for atmospheric science and air quality monitoring. The large dataset (relative to any other previous publication) and the thorough and comprehensive data analyses presented in this paper support substantial conclusions about innovations to and examinations of mobile-MAX-DOAS techniques. These include addressing scientific questions such what are the optimal meteorological and measurement conditions for use of this method, the factors contributing the most to total error, potential modifications to the CIM method, and how consistent the method was compared to modeling results. The authors contribute to the field by developing methods for error analysis and approaches to quality check data, essential for future utility of mobile-MAX-DOAS. The authors also successfully identify technique aspects requiring further exploration or improvement, such as the knowledge of the variability of the NO to NO2 partitioning ratio. This paper is of high scientific quality but may benefit from the authors adding some relatively minor clarifications to some of their methodology sections in order to optimize clarity and scientific reproducibility.

Author reply: We thank the reviewer for the positive assessment.

Scientific Comments

Section 3: In order to increase clarity (and reproducibility) when discussing methods of averaging or other manipulations of the hourly wind data obtained from the MM5 model, the authors could include more information such as: the maximum altitude range for the wind fields in the model, the size of the vertical altitude bins (e.g., what are the different heights referred to in section 4?) and the resolution of the model (i.e., is it also 3kmx3km?).

Author reply: In section 3 (and also Fig. 2), we replaced 'averaged' by 'summed up'. In section, 3, already the altitude range (up to 5 km) and the horizontal resolution (3x3km$^2$) of the model simulations were stated.

In section 4 we added the information that the wind data between the surface and 1000m are used (weighted by an exponetial profile with either 300m or 500m).

In the revised version we also mention the exact way in which the wind data are averaged for the individual circles: '...we calculated the average wind speed and direction for all individual locations and times of the car MAX-DOAS measurements along the circle'.

Section 4 Starting on line 5 the authors state that the wind speed and direction are averaged over the measurement area but it is not totally clear whether that refers to averaging in both the horizontal and the vertical or just the horizontal covering the measurement circle area. Author reply: In the text in section 4 describing the averaging in time and location we added the following hint '(for the vertical averaging, see below)' to the end of section 4, where the vertical averaging is described.

In general, it would help to improve clarity by specifying what is meant by "average wind" (temporally over a particular period and/or spatially over a specific vertical or horizontal distance) if the term means different things in different sections.

Author reply: We believe that with the changes described above, in the revised version of the manuscript all necessary information about the averaging process of the wind data is provided (see above).

It would be potentially helpful to specify more why the wind data are weighted by exponentially decreasing profiles and what actual form of equation was used (e.g., does the equation have any coefficients or variables other than scale height and altitude?).

Author reply: To make the motivation for our procedure more clear, we added the following information to section 4: 'Both wind speed and direction vary systematically with altitude (see e.g. Fig. 3). Thus a choice has to be made, in which altitude range most NOx is probably situated, because the wind data for this altitude range determine the NOx flux. Since our measurements were performed close to the NOx emission sources, and since most NOx emission sources are located close to the surface, we assume expontentially decreasing NOx concentration profiles with scale heights of 300 m (winter) and 500 m (summer). We added an equation describing the vertical averaging (new equation 3) We also added the following information at the end of section 4.1: 'Here it should also be noted that the exact choice of the scale height is not critical: changes of the scale heights between 200 and 700m usually lead to differences of the wind speed <0.5m/s and wind direction <5°. The errors of the derived NOx emissions associated with uncertainties of the wind speed and direction are quantified in section 4.6.3'.

Also, it is unclear whether the MM5 vertical wind profiles are interpolated into a continuous profile from the MM5 altitude bins or averaged to a single value in the vertical before an exponential profile is applied.

Author reply: The exact procedure is now described in the new eq. 3.

The authors state that these exponential vertical profiles account for different vertical mixing conditions but it is unclear if this could have been applied to the VCD instead since the exponential profile appears to account for the fact that the NO2 is unlikely to be uniform in concentration with altitude in the boundary layer. This is related to wind but also to the NOx sources being predominantly (presumably) surface-based.

Author reply: We added more information to make the motivation for our approach more clear (see replies to comments above).

On Line 19 it is potentially unclear as to how Fig. A1 shows that the stated assumptions are not necessarily valid.

Author reply: Here reference to Fig. A2 should have been made. This is corrected now.

Section 4.2 When discussing the comparison of clock-wise vs. counter-clockwise calculations of emission flux, it may be helpful for the reader to be reminded that when there is a gap the last VCD in the direction of calculation is used as the correct VCD for the gap segment (from previously published papers) and that this can contribute to the difference between the two calculations (unless this method was not used and then specify the new method).

Author reply: We checked the text in section 4.2 and found that the provided description should be sufficiently clear: 'Since the values of the wind speed and direction in equation 2 are determined for the location of measurement i, but the distance $\Delta si$ is determined between measurement i and i+1, the direction for which the sum is calculated leads to a difference in the derived total NO2 flux.'

Section 4.5 In section 4.5 the authors could chose to briefly address how homogenous the NOx sources are across Paris (e.g., major point source locations vs. high concentration road traffic/highways).

Author reply: We added the locations and types of the strongest point sources in the new table 1. We added the following text at the end of section 4.5: 'The location and type of the strongest point sources is presented in Table 1.'.

Section 4.6 In 4.6 on page 7, line 32: it may be useful within the error contribution discussion for the authors to address whether diurnal trends could introduce significant error when the time difference between measurements of influx and outflux is large. For example, if measurements start during a diurnal NOx emission peak but the emission rates decrease significantly well before the circle is completed (or vice versa). If this is the case, how would the authors determine what time period the resulting calculated emission value are representative of?

Author reply: At the beginning of section 4.6 (directly before section 4.6.1) it is already stated: 'It has, however, to be taken into account that the derived NOx emissions are only representative for a specific time period of the day (mainly depending on wind speed and the diameter of the driving circle, see also section 5)'. In section 5, we added the following information: 'Here it is interesting to note that for the cases considerd here time variations of $\pm$1 h lead to changes of the respective input emissions of 2% to 15%.'.

Section 4.6.1 The methodology in this section may benefit from greater clarity with some additional information. Starting on line 22: does this "error of F" refer to an estimate of the standard deviation of all the VCDs from an entire, single circle or for a specified segment of the measurement circle?

Author reply: We checked the text in section 4.6.1 and found that the information should be sufficient and correct. In order to avoid confusion, we slightly modified the text: 'In order to estimate the error of F due to gaps, we use the following approach: First we estimate the uncertainty of VCD simply by the standard deviation of all measurements VCDi:'.

This section may benefit from the authors also defining what is meant by a "single summand" and the difference between _VCD and _VCDi. _VCDi is slightly confusing in the sense that is there not only one VCD derived for each measurement location i?

Author reply: We checked the text, and found that the information in section 4.6.1 is consistent with the definition of the respective quantities in equation 2.

Technical Corrections

Title: The authors may choose to add "during the MEGAPOLI campaign" to the title so that it most clearly reflects the paper contents. Adding something about "examination of error and optimal conditions" may also help since these are important contributions to the field.

Author reply: We added "during the MEGAPOLI campaign" to the title. However, we did not add more text to the title, because it is already quite long.

I suggest writing emissions "from" rather than emissions "for" and writing measurements were performed "in" large circles rather than "at" or "on" for maximum clarity and grammatical correctness throughout.

Author reply: corrected

In general, when explaining data analyses completed or methodology used, use of the past tense is most correct (e.g., measurements "were" rather than "are" performed).

Author reply: We changed to 'past tense' for such parts throughout the manuscript.

Abstract Line 15: add NO2 to influx into and outflux out of the encircled area for maximum clarity.

Author reply: The suggested text was added.

Line 16: “The difference of both fluxes represents the total emission” could be changed to “The difference between the influx and outflux represent the total emission” for increased clarity.

Author reply: The text was changed.

Line 22: It may be helpful to specify or give examples of “uncertainties” to minimize ambiguity.

Author reply: We added the information that ‘. . .., which typically ranges between 30% and 50%’

Line 25: There is an extra “p” in “developed”

Author reply: corrected

Section 3 Line 11/12: missing an “and” between by month and by source sector.

Author reply: ‘and’ was added

Line 18: (typo) pick “a” or “the” in the sentence starting with “In Figure A1

Author reply: corrected

Section 4 Page 4 line 4: “I” needs to be small-caps. There may be other instances of this in other parts of the paper.

Author reply: corrected

In this section the authors refer to A2 when referring to comparison of averaged wind speed and directions during periods of MAX-DOAS measurements yet the diagram shows NO2 profiles rather than wind data. I think this should be referring to figure A1.

Author reply: Yes, it should be Fig. A1. The text was corrected

Section 4.1 Page 4 line 10: missing a close bracket at the end of the sentence.

Author reply: A brackett was added.

Page 4 line 28/29: The NOx layer scale heights or layer heights?

Author reply: The text was changed to 'when weighted with the exponential NOx profiles with scale heights of 300 m and 500 m, respectively.'

Section 4.2 Page 5 line 5: Lowercase "l" needed.

Author reply: corrected

Section 4.4 Page 6 line 14: comma needs to be replaced with a period.

Author reply: corrected

Page 6 sentence starting line 27: it is unclear what the "difference" is; this sentence may benefit from rewording to improve clarify and (e.g., which VCDs are subtracted from which?)

Author reply: The text is changed to '. . .the lifetime correction was only applied to the enhancement of the NO2 VCDs over the minimum NO2 VCDs at the upwind side.'.

Section 4.5 Page 7 line 19: typo, second last word in sentence.

Author reply: corrected

Section 4.6.3 Line 19: typo "accounted"

Author reply: corrected

Line 28: specify the relative differences in "what" (e.g., NOx emissions)

Author reply: The text is changed to 'Fig. 12 displays the relative differences of the derived NOx emissions with either averaged or spatio-temporally varying wind fields for all days. . .'.

Section 5 Line 33: typo, need the word 'beginning'

Author reply: corrected

Figures and Tables Fig. 3 Add to the caption: during the time of measurement of the entire single circle.

Author reply: The information was added.

Table1 – For large wind variability: Does “relative deviation of wind speed >30%” refer to a standard deviation using all the wind speeds during the measurement period or between the smallest and largest wind speeds for individual pairs?

Author reply: We added ‘(vmax – vmin)’ to the table.

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