

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.

Anonymous Referee #1

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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

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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.

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?).

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. 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. 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?). 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. 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

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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 NO_x sources being predominantly (presumably) surface-based. On Line 19 it is potentially unclear as to how Fig. A1 shows that the stated assumptions are not necessarily valid.

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).

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

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 NO_x 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?

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

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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. 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. 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).

Abstract Line 15: add NO₂ to influx into and outflux out of the encircled area for maximum clarity. 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. Line 22: It may be helpful to specify or give examples of “uncertainties” to minimize ambiguity. Line 25: There is an extra “p” in “developed”
Section 3 Line 11/12: missing an “and” between by month and by source sector. Line 18: (typo) pick “a” or “the” in the sentence starting with “In Figure A1

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. 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.

Section 4.1 Page 4 line 10: missing a close bracket at the end of the sentence. Page 4 line 28/29: The NO_x layer scale heights or layer heights?

Section 4.2 Page 5 line 5: Lowercase “l” needed.

Section 4.4 Page 6 line 14: comma needs to be replaced with a period. Page 6 sentence starting line 27: it is unclear what the “difference” is; this sentence may benefit

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from rewording to improve clarify and (e.g., which VCDs are subtracted from which?)

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

Section 4.6.3 Line 19: typo “accounted” Line 28: specify the relative differences in “what” (e.g., NO_x emissions)

Section 5 Line 33: typo, need the word ‘beginning’

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

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?

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