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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Retrieving tropospheric NO_2 vertical column densities around the city of Beijing and estimating NO_x emissions based on carMAX-DOAS measurements" *by* Xinghong Cheng et al.

Anonymous Referee #2

Received and published: 17 July 2020

General Remarks:

The manuscript by Cheng et al. describes mobile-MAX-DOAS measurements of NO2 VCDs in the Beijing area. The study is based on 19 circuits around the 6th Ring Road in Beijing during two seasons in 2014. Emissions of NOx by Beijing were estimated using the MAX-DOAS VCDs as well as winds and NOx/NO2 ratios simulated by the LAPS-WRF-CMAQ model system. Simulated winds were validated using wind observations at multiple locations in the region. The seasonal differences between average VCDs and NOx emissions were compared and found to be greater during the heating periods.

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Discussion paper

The estimated NOx emissions were compared to a bottom-up emission inventory from 2012.

The manuscript illustrates how mobile-MAX-DOAS measurements in Beijing and other megacities could provide dynamic monitoring of NOx emissions and validation of satellite NO2 VCDs. My concerns are about the application and interpretation of statistics for the validation of modelled winds (Issue #1), and about the validity of the scientific methods and assumptions for some of the measurement periods and in the calculation of errors (Issue #2), and error calculation (Issue #3). The authors should address the following specific issues and minor corrections before the manuscript is published.

Specific Issues:

Issue #1) In general, be careful about application and interpretation of statistics for discussion of means and validation of wind data. Some descriptions of error calculations are not sufficiently complete to allow their reproduction.

-Page 2 line 32-33: please include an uncertainty value when stating mean values throughout and, preferably, indicated the statistical type of uncertainty (e.g., standard deviation of mean).

-Page 10 line 266-267: what about Fig. S2 shows that the temporal variation between the simulated and observed wind speed were consistent? An R of 0.47 is quite low given that the R2= 0.22, meaning only 22% of variation in the model is explained by variation in the observation. In terms of the simulated wind-speed being greater than the observations, if that conclusion is from the slope being >1, the slope should be assessed with an y-intercept forced to zero. In an ideal case the simulated and observed speeds would be the same (1:1 line, 0 intercept) unless a systematic error or offset is expected. Also, the slope is barely above zero. Using a linear fitting algorithm program that provides the expected error on the slope and intercept will indicate whether the slope was statistically greater than zero.

-Page 10 line 268-269: it is still unclear how the relative error was used to “correct” the simulated winds. Did you simply add it as an error bar?

-Page 10 Page 273-274 The wind rose plots (Fig. S3) give little indication that the wind-direction was consistent between the simulation and observations during you driving periods. Without this information, the claim that the modelled winds were accurate is unsupported. Either apply circular statistics to determine the R2 of simulated and observed wind-directions or provide time series plots of wind-direction (perhaps in the supplemental).

-Page 10 lines 283-284 The regression statistics should be determined using a linear correlation line with an intercept forced to zero (see Page 10 line 266-267 comment).

Issue #2) A major assumption of the mobile-MAX-DOAS method for estimating emissions using a flux integral is that winds stayed relatively constant during the measurement period. Therefore, this method is only valid to use on MAX-DOAS data with these wind conditions.

-Page 14 line 386, point (4): the driving routes should be screened for changing wind field since relatively constant wind fields are a fundamental assumption of Eq. (1) for mobile-MAX-DOAS and, if violated, lead to large, unquantified uncertainties. This point is stated in the manuscript Page 16 lines 444-446 but contrasts with the inclusion of emission estimates in your figures and comparisons where large wind fluctuations occurred It is not scientifically valid to use Eq. (1) under these conditions and the emission estimates from these routes should not be presented unless designated as having unknown error.

-Page 14 line 395: Therefore, exclude journeys of wind type “other”. See comment above.

Issues #3)

-Page 14 line 408-409 The total error values appear too small given all the potential

factors contributing to uncertainty. Shaiganfar et al. (2011) suggests that the error of using the VCDgeo calculation alone is up to 20% (https://doi.org/10.5194/acp-11-10871-2011). A VCD error 10% is likely too small if both the SCD retrieval error and the VCDgeo errors are combined. Was the error on the simulated wind speed the RMSE between the modelled and observed winds during the driving period? What about the contribution of wind-direction error? If the wind-direction varied a lot, the error contribution would likely dominate.

Minor Issues and Corrections:

Page 2 line 41: replace “consumption” of fossil fuels with “combustion”.

Page 2 line 47: remove the word “obviously” here and throughout as it is unnecessary.

Page 2 lines 54-56: since you begin with a discussion of PM, may want to explicitly describe that NOx is a precursor of nitric acid, which is a precursor of nitrate aerosols for maximum clarity.

Page 2 line 60: replace “high” with “large”.

Page 3 line 64: define “top-down constraint” or use “top-down” emission estimate

Page 3 line 71: there is a missing space between sentences.

Page 3 line 72: specify whether it is a large decrease in precision or accuracy?

Page 3 line 74-75: please add citation(s) for these uncertainty factors.

Page 3 line 80 MAX-DOAS has been around for nearly two decades (see below).

Honninger, G. and Platt, U.: Observations of BrO and its vertical distribution during surface ozone depletion at Alert, Atmos. Environ., 36, 2481–2489, https://doi.org/10.1016/S1352-2310(02)00104-8, 2002.

Hönninger, G., von Friedeburg, C., and Platt, U.: Multi axis differential optical absorption spectroscopy (MAX-DOAS), Atmos. Chem. Phys., 4, 231–254,

https://doi.org/10.5194/acp-4-231-2004, 2004.

Wagner, T., Dix, B., von Friedeburg, C., Friess, U., Sanghavi, S., Sinreich, R., and Platt, U.: MAX-DOAS O4 measurements: A new technique to derive information on atmospheric aerosols – Principles and information content, J. Geophys. Res.-Atmos.,109, D22205, https://doi.org/10.1029/2004JD004904, 2004.

Page 3 line 82: add “pointing” between “zenith” and “directions”

Page 3 line 82: add “vertical” before “profiles”

Page 3 lines 84-86: I suggest adding these citations of MAX-DOAS NO2 measurements (below)

Tan et al. (2018) doi:10.5194/acp-18-15387-2018

Wagner et al. (2011) doi:10.5194/amt-4-2685-2011

Page 4 line 91: add “horizontal” before “spatial distribution of pollutants” (since the stationary inverse modelling MAX-DOAS technique also gives spatial information but in the vertical).

Page 4 line 95: in general, you may want to mention that mobile-MAX-DOAS could be very useful for validating the NO2 VCDs and NOx emission estimates from the new, high pixel resolution measurements by the TROPOMI instrument on the Sentinel-5P satellite.

Page 4 line 105: Consider adding the study objectives or aims before listing the sections. It can help the reader quickly determine if the paper of interest and makes it easier to follow the major conclusions.

Page 4 line 115: Please explain why wind-speeds at 10 m above surface was used instead of averaged within the boundary layer height from the model output given can be well-mixed into the boundary layer (up to ∼600 m in your study) at the location point and, thus, the 10 m wind speed is a lower-bound.

Page 5 line 142: The word “mounted” is more appropriate than “settled”.

Page 5 line 143: Please provide information about the instrument specifications (e.g., spectrometer model, spectral resolution, cooling mechanism etc.)

Page 6 line: How many measured spectra were averaged to produce a single measurement spectrum?

Page 5 line 145: This MAX-DOAS instrument was also used more recently in the Davis et al. (2019) mobile-MAX-DOAS measurements of NOx emissions.

Page 6 line 155: Please specify whether the sequence measured multiple 30o spectra or if every 30o measurement was immediately followed by a 90o measurement.

Page 6 line 164: Please quantify “changed slightly”. Example, the wind changed by < X degrees and < Y m/s during the circle journey.

Page 6 line 175: add “absorption” in front of cross sections.

Page 6 line 176 typo: “dimmer” should be “dimer”

Page 7 line 185: Do you mean “for in-situ MAX-DOAS measurements”?

Page 7 line 187: Should this be “extending Eq. (6) to Eq. (7)” ?

Page 7 line 194: replace “site” typo with “in-situ”.

Page 7 line 196: check equation number.

Page 7 Line 197: add (SZA) to DSCD_offset.

Page 7 line 204: check the Eq. number.

Page 7 line 205: replace “geometry” with “geometric” and cite Brinksma et al, 2008 https://doi.org/10.1029/2007JD008808 and Wagner et al. 2010 https://doi.org/10.5194/amt-3-129-2010

Page 8 line 209-210: the meaning of this sentence is unclear.

Page 8 line 211: NO2 needs subscript.

Page 8 line 215: missing space after “September 23”.

Page 8 line 216: missing period at the end of this sentence.

Page 9 line 233: hyphen needed between “three” and “dimensional”

Page 9 line 251: delete the extra period.

Page 10 line 265: change “area” to “areas”

Page 11 line 294: rewrite the sentence to read “The highest values were between. . .”

Page 11 line 296: see comment for Page 2 line 32-33.

Page 11 line 300-301: what factors would increase emissions in January compared to October? More home heating?

Page 11 line 301-302: were lower PBL heights and smaller wind-speeds found in January compared to September/October?

Page 12 line 337: subscript needed on NO2 VCD.

Page 12 line 349: see comment for Page 2 line 32-33.

Page 13 line 371: considering replacing “residents” with “residential”

Page 13 lines 375-377: in what way does it indicate the applicability? Please explain/elaborate. Page 22 623: fix error in reference name.

Pages 28 and 29: Figures 5 and 6: correct the unit notation (x1016) of VCDs in the caption.

Page 31 Figure 8: the caption says “under three types of wind fields” but only south and north winds are shown.

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