

Interactive comment on “Auto MAX-DOAS measurements around entire cities: quantification of NO_x emissions from the cities of Mannheim and Ludwigshafen (Germany)” by O. Ibrahim et al.

Anonymous Referee #1

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GENERAL COMMENTS

This paper is an application of the method introduced by Johansson et al. (2008) to estimate area emissions of air pollutants by remote sensing measurements from a car driving around the area. An estimate is given for the NO_x emissions from Mannheim and Ludwigshafen. This in itself is already worth publishing, although the method is not very innovative.

The new aspect of this paper is that the measurements are performed under an angle of 45°, for which the authors claim they are more sensitive to tropospheric NO₂ than the zenith sky measurements. However, the authors do not illustrate or argue that this

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increased sensitivity would indeed give a more accurate estimate of the area emission. Furthermore the systematic offset the zenith sky measurements would introduce are not relevant for this method since the difference between the in-flux and the out-flux over an area is not changed by a systematic offset in the vertical columns. However, the authors briefly mention the importance of measuring absolute tropospheric columns when using the method for deriving absolute values of fluxes through transects. It is a pity that the method is not applied to such an example.

The innovative character of this paper should be enhanced by either concentrating on the benefits of multi-axis as compared to zenith looking, including a relevant application or example, or concentrating on the accurate estimation of emissions by improving the error assessment. Johansson et al. (2008) already tried to minimise the uncertainty in the wind speed and direction over the time period of a measurement loop by using a meteorological model. The authors should make an effort to improve this.

SPECIFIC COMMENTS

Abstract, line 13-16: "In such cases, ... determined total emissions." The validity of this statement is not shown in the paper. With this method a systematic offset in the vertical columns would not lead to an offset in the determination of the area emission.

Abstract, line 24: "... which is in surprisingly good agreement with existing emission estimates." This sentence seems to bring down the message in your paper. If your method is valid, why does it surprise you that it is consistent with other estimates? Or are you surprised that existing bottom-up estimates are 'correct'?

I would suggest changing through the whole paper the vector \vec{s} to the scalar s , denoting the position along the driving route, and changing $d\vec{s}$ to $\vec{n}ds$, where \vec{n} is a unity vector directed outward and normal to the driving route direction.

You do not need Eq. (1) if you have Eq. (2). Please compact page 472, line 18 to page 473, line 9.

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The first part of Eq. (2) is wrong, should be $\int_A \vec{\nabla} \cdot (\text{VCD} \vec{W}) dA$.

page 473, line 13: The word 'averages' is used incorrect. The emission estimates are not averages of individual emission estimates, but they are based on instantaneous measurements of the NO₂ column and an average wind field.

page 474, line 10: 'processes, which are slow ...': do you mean 'fast'? If not, please explain.

Section, 2.2: Specify how you monitored the location (GPS?).

page 478, line 1: '... typically below 15%.' This is not true for small relative azimuth angles. Add a sentence on this and quantify.

page 478, lines 8-9: The total error estimate should not be given as 'larger than', but rather as 'smaller than' or 'approximately'.

page 480, line 23, and Table 1: how are the standard deviations of the wind speed and direction calculated, in time, or in both space and time? Does it make a difference?

Section 4.1.1 and 4.1.2: These error estimates should be improved by using the individual measurements from the three wind stations to determine whether the average wind field is systematically changing during the measurement loop. I suggest that you exploit this information, and possibly additional information (other stations or a model) to improve your error estimates. This is already recognized by the authors in Section 4.1.3. Since you are circling the area almost four times you might want to consider taking only the measurements during the period for which the wind was most stable, or the total estimated error is smallest. After you have improved the error estimates the last sentence of section 4.1.1 will probably change. If not, there should at least be some explanation why the error can be estimated as 'half of the standard deviation'.

Section 4.3 and Section 5. Since you know the Leighton ratio, you can calculate the mass of the NO_x emission using both masses of NO and NO₂. For a c_L of 0.35 this would result in a NO_x emission closer to 0.5 kg/s. You compare to existing emission

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estimates for Ludwigshafen and Mannheim. However, it is not clear if these emission estimates are comparable. What is the ratio of NO to NO₂ assumed in these existing estimates?

Section 4.4 title: only one additional error source is dealt with in this subsection, so maybe you want to change the title accordingly, e.g. 'Effect of ozone depletion'.

page 483, line 17-18: '... is found to be higher than the NO_x concentration'. This is not true before 11:00.

Section 5 contains statements that are not dealt with in the rest of the paper. These statements should go to e.g. the introduction. These are lines 7-16: 'Moreover, MAX-DOAS observations ... for satellite validation.'

Figure 1 should be removed. It does not give much additional information. Alternatively, Figures 1 and 2 could be merged to a (3D) sketch of the area over which the integration is performed.

TECHNICAL CORRECTIONS

Abstract, line 6: change 'based on of zenith' to 'based on zenith'

Abstract, line 23: change '17350±4100t' to '17350±4100 t/yr'

page 476, line 11: change 'light weighted' to 'light-weight'

page 476, line 24: change 'via one USB cable' to 'via a USB cable'

page 477, Eq. (3): specify α

page 479, line 2: change 'typically expressed as Leighton ratio ($L=[\text{NO}]/[\text{NO}_2]$).' to 'a function of the Leighton ratio ($L=[\text{NO}]/[\text{NO}_2]$; $c_L = 1 + L$).'

page 479, line 5: change 'terms' to 'factors'

page 480, line 14: change 'south-west' to 'South-East'

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page 480, line 23: change 'time of the measurements' to 'time period of each measurement loop'

page 482, line 12: change 'the time the air' to 'the time for the air'

page 482, line 12-13: change 'Because most NO_x ... area, we assume an effective transport time of 1 h.' to 'Assuming most NO_x ... area, the effective transport time would be 1 h.'

page 482, line 23: change '... of 1.35 to obtain to determine' to '... of 1.35 to obtain'

page 484, line 4: change 'but instead measuring scattered' to 'but instead of measuring scattered'

page 484, line 5: change 'sun light under zenith sky' to 'sun light from zenith-sky'

page 484, line 5: change '... we use a MAX-DOAS instrument mounted on a car.' to '... we measure under a slant angle with a MAX-DOAS instrument mounted on a car.'

Figure 5. The letters and numbers along the axes and within the plots should be enhanced with at least a factor two.

Figure 6, caption: change 'the driving distance during one observation' to 'the driving distance between two consecutive observations'

Figure 7, caption: change 'The vertical lines indicate ...' to 'The dashed vertical lines indicate ...'

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