

Interactive comment on “DOAS-measurement of NO₂ formation rate from NO_x emissions in the atmosphere” by E. Frins et al.

Anonymous Referee #2

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In this paper, the authors describe a method to estimate NO₂ formation rates in a plume from a stack using passive MAX-DOAS observations without the need to estimate wind speeds and directions within the plume. The basic idea is to use SO₂ as a passive tracer and assuming constant SO₂ emissions and similar dilution and transport rates for both trace gases. The method is then applied to two days of measurements in Montevideo demonstrating that it can be used under realistic conditions.

The paper is well written, provides a clear explanation of the method and fits well into the scope of AMT. However, I have a number of questions and concerns with respect to the measurement results and their interpretation as outlined below, and in my opinion, the paper needs to be strengthened considerably with respect to the discussion of the

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results before it can be accepted for publication in AMT.

Major points:

- 1) My main concern is linked to the results shown in Fig. 3. The basic assumption of the method is that there is a plume and that NO₂ and SO₂ are mixed and transported within this plume in a similar way. However, when looking at the data presented, a SO₂ plume can only be discerned in the first two scans while an NO₂ plume of varying shapes is apparent in all measurements. This is odd and raises questions concerning the reliability of the SO₂ observations. This dataset would be much more convincing if a decrease of SO₂ to background values could be shown in any of the data. In my opinion, the full plume needs to be sampled by such measurements and the data as they are shown are questionable, at least for SO₂.
- 2) There is no discussion at all in the manuscript about the fact that the light paths in the spectral regions used for NO₂ and SO₂ retrievals are different and how this affects the method used. As radiation dilution effects for SO₂ are well known, this point needs to be addressed by radiative transfer calculations.
- 3) I'm confused by the point of how the FOV of the instrument might affect the measurements. Clearly, the integration over different viewing directions will only yield the correct result if there is no overlap between viewing directions which is not possible in practice, in particular if the plume is observed at different distances. Some of the effect might cancel by taking the ratio of the NO₂ and SO₂ observations, but then the FOV of the observations at 320 nm and at 450 nm will be different. This point needs to be clarified in the manuscript.
- 4) As I expect different NO₂ formation rates in different parts of the plume (mixing with surrounding air), the partial sampling of the plume introduces additional uncertainties. As stated above, I think that measuring the full plume cross-section is important for the method.

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5) When looking at the picture of the measurement area I wonder how the effect of other NO₂ (and possibly also SO₂) sources can be excluded. In fact, the absence of a clear plume in the data could be the result of NO₂ / SO₂ from other sources, either at the location or in front or behind the plume of interest. Please comment.

6) The NO₂ formation rate derived is stated but not discussed. I think what is needed here are

- an uncertainty for this number
- a comparison to literature values
- a comparison to the “classical” derivation using wind information for the flux through the individual cross-sections

7) Why do the authors assume a constant NO₂ production rate? Shouldn't that decrease as NO (and O₃) are consumed along the plume?

Minor points:

Abstract: emitted in the atmosphere => emitted into the atmosphere (also elsewhere)

Introduction: natural solar radiation => solar radiation

P5718, I23: and the references => and references

P5723, I19: on the north => in the north

P5724, I2: traversed distance by the plume => distance traversed by the plume

P5724, I29: just out of interest – what do you estimate the temperature in the plume is compared to that at your measurement site? Do you expect the plume to cool down along the path and if so, how large would the impact on the results be?

P5726, I9: wind condition exists => wind conditions exist

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P5272, I14: for the help achieved => for the help provided

Fig. 1: Explain all symbols in caption

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