

We like to thank the reviewers for their effort to evaluate our manuscript and for their valuable comments, which helped to improve the quality of our manuscript. In response to the reviewers' comments we rewrote several paragraphs of the manuscript. Our answer and the major changes performed in the manuscript are the following:

Comments/answers to Referee 2:

In the following we follow the numbering of the referee.

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.

In our work we assume that all gases are transported by wind at the same speed, but the relative concentration of NO₂ and SO₂ will not be constant. In other words, to deduce Eq. (8) we do not assume that NO₂ and SO₂ are mixed and transported inside the plume in a similar mode, because NO₂ is being continuously created inside the plume. That is, it is to be expected that the concentration of NO₂ increases (in absolute terms or relative to SO₂), while the concentration of SO₂ decreases mainly due to widening of the plume cross-section. Thus, it is reasonable to observe a decrease of SO₂ to background values with increasing distance to the stacks. We recognize that probably the range of elevation angles for the cross scans 4-6 (shown in Fig. 3) should have been larger than 10° in order to show that effectively SO₂ decreases to background values (revised manuscript, p. 8, line 15-18). This issue will be taken in account in future works.

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.

Radiation dilution effects are important when the distance to the plume is of the order of the mean free photon path length in the atmosphere (approximately 10 km at 320 nm due to Rayleigh scattering, about 4-5 km including Mie scattering), as discussed in the literature (see e.g., C. Kern et al. Bull Volcanol., in the revised manuscript). In our case the distance to the plume was of the order of 2 km, and thus, radiation dilution effects should be of minor importance. We add a comment in the revised manuscript on page 8, line 5-6.

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.

The field of view of the instrument (FOV) is 0.4⁰, while the scans were performed in steps of 0.5⁰-1⁰. Thus, there is no overlap between the viewing directions of consecutive observations. In the revised manuscript we added the details of the measurement routine (Sec. 3.1, page 6, line 17 and 19-23).

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.

We realize that the experimental data presented in this work are not exhaustive. Our purpose was to illustrate the application of the method by achieving several vertical scans at different distances from the stack in a very short time interval. (See Comments/answers to Referee 1, Specific Comments, 2nd and 3th paragraphs.)

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.

In principle, we can not completely exclude the possibility that other sources of NO₂ (and/or SO₂) may exist. But, to the best of our knowledge, we can not identify any industrial NO₂- or SO₂-sources in the neighbourhood of the refinery.

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

In the revised manuscript we give an estimation of the uncertainty in the NO₂ formation rate. A comparison to the “classical” derivation using wind information for the flux through the individual cross-sections was not possible because the wind was rapidly changing in speed and direction during the measurements.

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

We agree with the reviewer that the rate of NO₂ production should exponentially decrease as the initial NO is consumed. However, the rate of NO-NO₂ conversion depends on the ozone concentration inside the plume, which can be assumed to increase while the plume ages (initially all O₃ will just be titrated by NO, since $[NO] \gg [O_3]$, so overall the rate of increase of NO₂ is not entirely easy to predict and deviations from a linear increase may not immediately be noticeable. At present we can not definitively state whether the NO₂ formation rate is constant or not, because we would require extensive field measurements. We consider our result (the linear fit) as a first approximation. We agree with the referee that this issue requires a further investigation, and it will be the topic of future works.

We corrected all the minor points mentioned by the referee.