

## ***Interactive comment on “DOAS-measurement of NO<sub>2</sub> formation rate from NO<sub>x</sub> emissions in the atmosphere” by E. Frins et al.***

### **Anonymous Referee #1**

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#### General comments

This paper proposes a method for estimating the formation rate of NO<sub>2</sub> without knowledge of wind speed and direction in the plume originating from a localized source. Its theoretical description and results obtained from actual measurements with ground-based DOAS instrument at Montevideo, Uruguay in the period from December 2009 to March 2011 are given. I think that this methodology and measurements are unique and the subject of this paper is appropriate for AMT. There are, however, some points that require additional consideration. I recommend this manuscript will be published after adequately addressing my concerns, which are described in detail below.

#### Specific comments

It seems that the present study has completely ignored the multiple scattering in the atmosphere (including inside the targeted plume). When the multiple scattering occurs in the plume, the integration of trace gas concentration over  $r$  (the distance to the plume measured from the instrument location) does not match the slant column density (SCD) retrieved by DOAS, making the equation (3) invalid. Moreover, the difference can be dependent on the distance between the plume and the source, due to different aerosol (smoke) and solar illumination (i.e., solar azimuth angle) conditions. Thus, I strongly recommend interpreting the retrieved SCDs with a consideration of the effects of multiple scattering or recommend adding some statements justifying the omission of multiple scattering.

According to the equation (2), the flux (denoted as  $\phi$ ) should be a quantity covering the whole range of elevation angles in which SCDs are significantly high. In the equation (8), however, whereas  $\phi(\text{SO}_2)$  is based on the above definition,  $S$  values are not, but based on a limited range of elevation angles at which DOAS measurements were made. In particular, test measurements at Montevideo only cover part of the plume at elevation angles less than 10 degrees. Does this inconsistency impact the results of this study (e.g., the result that the  $\text{NO}_2$ -formation rate per unit distance has been estimated to be  $25 \text{ kg (h km)}^{-1}$ )? Perhaps in relation to this, on p.5726, line 1-8, the authors have shown results obtained from a limited set of SCDs within 3-degree elevation angles of the center of the plume. Similar to this sensitivity analysis, I recommend testing some more different ranges of elevation angles (e.g., 0-8 degrees, 0-5 degrees) and discussing the results in the paper.

Technical corrections

Abstract and p.5723, line 19: Please mention the latitude and longitude of the measurement location.

p.5721, line 8: I guess that the unit of  $\alpha$  is degree. To make it clear, please give a unit just after "1". At this sentence, the authors approximate the equation (1), since the

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difference between two consecutive elevation angles is much smaller than 1 degree. Is this approximation really valid? Please mention a difference between consecutive elevation angles for test measurements at Montevideo in section 3.1.

p.5723, line 12: It is unclear to me what the "significant values" mean. According to Figure 3, the NO<sub>2</sub> SCD is still high even at an elevation angle of 10 degrees. Does omitting the range of elevation angles beyond 10 degrees influence the estimate of NO<sub>2</sub> formation rate?

p.5724, line 5: Please give the original words for MAX-DOAS.

p.5724, line 12-14: The authors state that trace gas concentrations outside the plume were negligibly small. I think that it would be better to state this quantitatively, for example, in comparison to the detection limit and/or uncertainty of the measurements.

p.5724, line 24-26: Would it be reasonable to add HCHO in the SO<sub>2</sub> evaluation?

p.5724, line 27: "molecule" should be "molecules".

p.5724, line 28: What do "These figures" indicate?

Section 3.2: While the test measurements were made for a long time period from December 2009 to March 2011 (as mentioned in section 3.1), why does the section 3.2 focus on only two days of 29 March 2011 and 1 December 2009?

p.5725, line 8: It may be better to delete a comma just after "SCD".

p.5725, line 19: Why does S(SO<sub>2</sub>) decrease?

p.5726, line 15 and 16: "Figure" should be "Figures". "shows" should be "show".

p.5726, line 15-19: How much is the estimate of NO<sub>2</sub> formation rate for the case of 1 December 2009? What about its comparison with the estimate for 29 March 2011? Can the difference be explained by different conditions (e.g., season) between the two cases?

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