Atmos. Chem. Phys. Discuss., 6, S5152–S5157, 2006
www.atmos-chem-phys-discuss.net/6/S5152/2006/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Lightning-produced NO_2 observed by two ground-based UV-visible spectrometers at Vanscoy, Saskatchewan in August 2004" *by* A. Fraser et al.

Anonymous Referee #3

Received and published: 7 December 2006

Review of the paper

'Lightning-produced NO2 observed by two ground-based UV-visible spectrometers at Vanscoy, Saskatchewan in August 2004' by Fraser et al.

The paper describes the estimation of the NO2 production by lightning from ground based DOAS observations of NO2 during a heavy thunderstorm. From the measured enhancement of the NO2 absorptions they separate the contribution due to enhanced light paths and attribute the remaining increase to the NO2 production by lightning. From the comparison of the derived NO2 production with simultaneously measured lightning counts, the authors derive the NO2 production per flash.

In general, the paper presents a nice case study of the effect of lightning on ground based DOAS observations of NO2 (and O3 and O4). Also the ideas to separate the effects of enhanced light paths and the actual NO2 productions are promising and help to improve the quantitative interpretation of DOAS observations under cloudy sky conditions. Nevertheless, I feel that (besides some general errors of the approach, see below), the conclusions are a little bit too ambiguous and the presented accuracies are by far too optimistic. However, taking into account the very large uncertainties of current estimates of NOx production by lightning, the derived results are still valuable, and I think the paper is a good contribution to improve our understanding of the NOx production by lightning. After a few major comments and several minor comments are addressed, I recommend publication in ACP.

Major comments:

A) Several assumptions, on which the final conclusions are based, are very optimistic. For example, I have doubts that the derived NO2 production from the measurements is really representative for this (and for other) thunderstorms. A1) The observations cover only a small part of the thunderstorm and already from the temporal variation of the observed NO2 production, it can be concluded that the production is not homogeneous throughout the volume. A2) The separation of the light path effects from the total enhancement is based on (implicit) assumptions on the profile shapes of NO2, O3 and O4 as well as on the height distribution of the light path enhancement. Since for the latter and the concentrations of NO2 and O3 no height profiles are actually known, the estimation of the effect of the light path enhancement on the total enhancement of the NO2 absorption is actually very uncertain. Light path enhancement in different altitudes will have very different effects on all three absorbers. The authors should state which assumption on the profiles they have made. They should also give more details on the uncertainties. (it should be also taken into consideration that besides the uncertainties of the vertical profiles, also the horizontal distribution is not known, but could have a strong effect.) The authors should also state which profiles they have assumed for the

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AMF calculations. The ratio of the AMFs for NO2 and O3 will also strongly depend on the assumptions made for the tropospheric profiles. B) The authors integrate the measured NO2 enhancement over the time. Is this really justified? I guess that the NO2 produced at the beginning will be still present at the end of the period of enhanced NO2 absorptions. If one integrates over the time, is it counted several times? C) The derived lightning production of NO2 is presented as NO2 SCD. However, what is really needed to quantify the total amount of NO2, is the VCD (vertical column density). The authors would have to correct their SCDs by an appropriate AMF (for multiple scattering inside the clouds). Like for the above point, here assumptions on the profiles of NO2 and the light path enhancements have to be made and an error has to be estimated. In any case, assuming that effective light path enhancement has taken place, the NO2 VCD should be expected to be systematically smaller than the NO2 SCD. This would have direct consequences for the derived NO2 production rate. I also recommend to state clearly which quantities are presented and discussed. In Fig. 7b, e.g. 'residual NO2 SCD' should be written. Quantities like 'amount' or 'value' should be avoided and replaced by more precise expressions. D) To derive finally the NOx production by lightning one has to make some assumptions on the NO2/NOx ratio. This ratio depends in particular on the actinic flux and temperature, thus depending strongly on altitude. Again, assumptions on the NO2 profile become important. What are the assumptions the authors make?

Minor comments

e) page 10065, line 14: As far as I know the lightning production is given as NOx. The authors should clearly state this here. They should shortly discuss how the NO2 production is related to the NOx production.

f) page 100066, line 14: What is the NO2 region and the O3 region? Please indicate wavelength ranges. Which settings are used for the analysis of O4?

g) page 100066, line 15: How large is the sensitivity (quantum efficiency) of the old

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and new detector?

h) page 100066, line 27: For an uncooled detector I would expect that the influence of the dark current can become important, especially during the presence of a heavy thunderstorm when the measured intensity is low. Please comment on this.

i) page 100068, line 25: It might be interesting to mention here that this finding is consistent with those of Erle et al., 1995 and Wagner et al., 1998.

j) page 100069, line 9: It might be interesting to mention here that this finding is consistent with those of Wagner et al., 1998.

Erle F., K. Pfeilsticker, and U. Platt, On the influence of tropospheric clouds on zenith-scattered-light measurements of stratospheric species, Geophys. Res. Lett., 22, 2725-2728, 1995. Wagner, T., F. Erle, L. Marquard, C. Otten, K. Pfeilsticker, T. Senne, J: Stutz, and U. Platt, Cloudy sky optical paths as derived from differential optical absorption spectroscopy observations, J. Geophys. Res., 103, 25307-25321, 1998.

k) page 100069, line 22: It might be interesting to include the reference of Greenblatt et al., 1990. Greenblatt G. D., J.J. Orlando, J.B. Burkholder, and A.R. Ravishankara, Absorption measurements of oxygen between 330 and 1140 nm, J. Geophys. Res., 95, 18577-18582, 1990.

l) page 100070, line 9: The ratio will be strongly dependent on the vertical profile of NO2 and the light path enhancement. Please mention here.

m) page 100070, line 10: Is there an explanation for the exponential dependence? Or is this just a best fit?

n) equation 1: This assumption is only valid if the vertical profile of NO2 and the light path enhancement do not change.

o) page 10072, line 4: To my knowledge, the wavelength dependence of the AMF becomes very important only for large solar zenith angles. However, the AMF is much

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more dependent on the assumed profile, in particular in the troposphere. Which tropospheric profiles of NO2 and O3 were assumed for the AMF calculations?

p) page 10072, line 8: Especially this ratio will strongly depend on the assumed profiles.

q) page 10072, line 21: How was the increase determined? How strong is the increase?

r) page 10073, line 9: The curve in Fig. 7 shows the NO2 SCD. For the determination of the amount of lightning produced NO2, however, the VCD has to be used. In particular, one has to make some assumptions on the light path enhancement (depends on assumed profiles).

s) page 10073, line 9: Why do the authors integrate over time? The lifetime of NO2 is long enough to let the NO2 accumulate during the thunderstorm. What do the authors assume for the lifetime of NO2?

t) page 10073, lines 19-25: I think the true errors are by far larger than stated here. I think, however, that this is not a great pity, taking into account the high uncertainties of current estimates of lightning produced NOx.

u) Fig. 1: Please show also fit results for O4

v) Fig. 2: In which units is 'cloud opacity' given?

w) Fig. 5: It would be very interesting to show also the observed (average) intensity here. The largest optical thickness of the cloud (giving largest light path enhancement) should be related to the lowest intensity (see e.g. Wagner et al., 1998).

x) Fig. 5: How is the RCD for O4 derived?

y) Fig. 8: How realistic are the assumptions of the radiative transfer modelling? I am a little bit puzzled by the low upper edge of the cloud (only 5km). Again, here the measured intensity could help to justify the model assumptions.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 10063, 2006.

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Interactive Comment