Atmos. Chem. Phys. Discuss., 6, S4662–S4668, 2006
www.atmos-chem-phys-discuss.net/6/S4662/2006/

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 #1

Received and published: 17 November 2006

General Comments

This paper presents NO2 spectrometer measurements in Canada during the passage of a thunderstorm. The paper addresses scientific questions relevant to the scope of ACP. The measurements presented seem to be of good quality, however the determination of lightning-produced NO2 and the following discussion need some improvements. Novel ideas or concepts of the paper are not pointed out clearly by the authors. The paper gives a brief but precise description of the performed work and used methods. The paper is well organized, well-written and fluent to read. The paper is suitable for publication in ACP after a major revision.

Specific Comments

- It should be mentioned that descriptions of these kind of measurements are rather rare up to now in the literature. Why are they rare (difficult to perform)?

- It should be added that these measurements were performed in a remote region (?) with no local pollution (?), if this is true. This is an important statement otherwise the enhancement in calculated lightning-produced NO_2 could also be due to upward transport of anthropogenic NO_2.

- It is assumed that the observed O_3 SCD increase is only due to multiple scattering in the thick cloud. However, it is mentioned that O_3 transported upward from the boundary layer could also enhance or decrease O_3 SCD measured in the thunderstorm (also known from airborne in situ measurements and cloud model simulations). Is there any possibility to quantify the amount of O_3 transported? Is it correct to assume that this contribution is negligible in comparison to the contribution from multiple scattering? You argue that the assumption is justified by the behaviour of the ozone to O_4 ratio.

- A big challenge is to determine which flashes, of all flashes in the monitored area (Fig. 4), contributed to the measured lightning-NO_2. This should be pointed out in the paper with some discussion. Some uncertainty for the average flash rate of 2.87 flashes/min should be given (page 10068).

- For the estimate of the amount of excess NO_2 you integrate between 60° and 85° (3 hours?). You multiply this excess NO_2 with the area of the heavy-precipitation cell (30+-3 km^2). Is not the air mass area (with elevated NO_2) that passed over the instrument during 3 hours much larger than this area (radius ~3 km)? The wind speed (multiplied with 3 hours) could perhaps give some better information on the size of the enhanced NO_2 area. I think that the area you use is not representative for the integrated NO_2 you estimated for 3 hours. A thunderstorm also has a complicated vertical structure as illustrated schematically in the Langford et al. (2004) paper.

ACPD
6, S4662–S4668, 2006

- In the introduction some information on O_3 and NO_x is given, however it would also be useful to add something about O_4 (first mentioned on page 10068). Explain more in detail the reasons for using O_4 in the paper and add some references that introduced the O_4-method to verify the influence of multiple scattering.

- Page 10070, Line 21-24: “The difference between the observed NO_2 slant column and the slant column calculated from the interpolated NO_2/O_4 ratio is the amount of NO_2 attributed to production by lightning.” In this case you assume that the same kind of thick cloud is present before and after the large increase in NO_2 SCD (thunderstorm passage). Is this a correct assumption (I would expect to have less thick clouds before and after)?

- For the AMF calculation with the radiative transfer model you assume “a thick cumulus cloud near the surface, of optical depth 70, extending between 1 and 5 km”. The cloud opacity in Fig. 2 indicates a cloud top of 9-10 km that I also would assume from the strong radar reflectivity in the radar image (Fig. 3) and for the presence of a thunderstorm with lightning (Fig. 4). Include some sensitivity tests where you change the cloud depth and optical depth in your model to see how the AMF changes. Give uncertainties.

Technical Corrections

Abstract:

Page 10064: Line 11: Change to “The enhanced NO_2 columns are partly attributed to”

Line 18: Change 6.58 to 6.18 (compare to values in the conclusions).

Line 19-21: It is not common to give references in the abstract. Shorten last sentence to “These results are within the range of previous estimates.”

1. Introduction:

Since the paper has the focus on tropospheric and not stratospheric measurements,

ACPD

6, S4662–S4668, 2006

these stratospheric parts can be cut in the introduction (page 10064: line 25-26, page 10065: line 1-2, line 4-5, and line 19-22).

Page 10065, Line 2-4: High NOx causes ozone production, low NOx causes O3 destruction. This statement is not completely correct, since if little sunlight is present in regions of high pollution (in winter or inside thick clouds), O3 is destroyed (titrated by NO).

Line 13-15: Add some more recent estimates (year $>$2000), e.g. some of [Huntrieser et al., 2002; Martin et al., 2002; Tie et al., 2002; Ridley et al., 2004; Boersma et al., 2005; Beirle et al., 2006] Check order of the listed references (chronological).

2. Instruments:

Page 10067, Line 4: Write out DOAS.

3. Thunderstorm observations:

Page 10068, Line 8: Change “Figure 3 shows the precipitation occurring” to “Figure 3 shows the radar reflectivity”. The precipitation (rain rate) is calculated from the measured reflectivity and not measured.

Line 9: Change “A cell of heavy rain” to “A cell of heavy rain and probably also hail”. (The elevated values of the radar reflectivity indicate that hail is probably also prominent.)

4. Slant column measurements:

Page 10069, Line 12-14: Change to “The observed enhancements in ozone and partly in NO2 are caused by increased path length through the atmosphere. In the case of NO2, the increase is partly also due to lightning-produced NOx.”

5. Derivation of lightning-produced NO2:

Page 10070, Line 3-5: Add some references for these two used methods.

Page 10071, Line 18-21: Why is the average of all ozonesondes during the campaign used and not a single ozone profile for the selected day used (would be more representative)?

6. NO2 flash production rate:

Page 10074, Line 11: In the original paper NOx/flash is stated and not NO2/flash.

Line 16-18: A more recent reference [Ridley et al., 2005] indicates that cloud-to-ground and intra-cloud flashes produce a similar amount of NO.

7. Conclusions:

Page 10074, Line 22-24: Add “ground-based UV-visible spectrometers”.

Page 10075, Line 4: “the range of (6.18-7.45)Ě)”, this is not the entire range of the estimates, just the range of the best estimates.

Acknowledgments:

Page 10075, Line 13: Change “Institude” to Institute.

References:

Page 10075, Line 27: Change to “Boccippio”.

Figures:

Fig. 1-8: Check that for all figures “day, time and location” are included in the text.

Fig. 2: What does first/second/third cloud height mean? Perhaps add that it is the cloud base height of different cloud layers.

Fig. 3 and 4: Add some information on longitude and latitude.

Fig. 3: Precipitation rate is not observed (only calculated from observed radar reflectivity).

Fig. 4: Add that the lightning flash data is superimposed on a GOES image (time?). Add “cloud-to-ground” lightning flash.

Fig. 5: Add a), b) and c).

References

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