Atmos. Chem. Phys. Discuss., 6, S6264–S6275, 2007
www.atmos-chem-phys-discuss.net/6/S6264/2007/

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.

A. Fraser et al.

Received and published: 15 January 2007

We thank the reviewer for their helpful comments on our paper. In the following, the reviewer's comments are repeated in italics, followed by our responses.

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)?

The measurements are rare due to the nature of thunderstorms and lightning-produced NOx and the measurement technique. Zenith-sky measurements are most sensitive at twilight and so are often not recorded at high frequency when the Sun is high in the sky (as was the case with the SAOZ during this study). Since most thunderstorms occur in the afternoon, they are not often captured. Furthermore, the NO2 enhancements are not always as dramatic as those seen during the this particular storm. Daily measurements made with the UT-GBS in Toronto over the summers of 2005 and 2006 have not seen the same dramatic increases in NO2 despite the occurrence of a number of thunderstorms during this period. We have added a comment on the rarity of these sorts of measurements in the Introduction.

"Measurements of lightning produced NO2 from ground-based UV-Vis instruments are rare. Zenith-sky measurements are most sensitive at twilight, and so are often not recorded at high frequency during high Sun conditions, which is when most thunderstorms occur. In addition, the NO2 enhancements are not always well-defined."

- 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 NO2 could also be due to upward transport of anthropogenic NO2.

Vanscoy was located upwind of Saskatoon (the nearest large city) during the storm, as shown by the radar images. We have added a more complete discussion of the storm in Section 3.

"Further examination of the radar imagery, available at 10-minute intervals, shows a series of thunderstorm cells forming to the west of Vanscoy, near the Alberta-Saskatchewan border, and traveling to the east, eventually dissipating to the

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east of the measurement site. In total, three cells or remnants of cells passed over Vanscoy. The maximum area of the three storm cells is (61 $\pm$ 10) km2. Vanscoy was upwind of Saskatoon for the duration of the storm, making it unlikely that NO2 enhancements are due to the upward transport of anthropogenic NO2"

- It is assumed that the observed O3 SCD increase is only due to multiple scattering in the thick cloud. However, it is mentioned that O3 transported upward from the boundary layer could also enhance or decrease O3 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 O3 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 O4 ratio.

Ozonesondes flown the day before and the day after the storm show no significant changes in the O3 profile. No sondes were flown during the storm. As stated in Section 4, line 9, the increase in ozone SCDs is of comparable magnitude to that observed by Pfielsticker et al. (1999), Wagner et al. (1998), and Erle et al. (1995) (new references) during cloudy-sky events with no thunderstorm activity. Together with the O3/O4 ratio, we maintain that our assumption of the ozone increase being mainly due to increased path length is justified.

- A big challenge is to determine which flashes, of all flashes in the monitored area (Fig. 4), contributed to the measured lightning-NO2. 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).

We have added discussion on the calculation of the number of flashes in Sect. 3. We now count all the flashes that occur upwind of Vanscoy in the three thunderstorms that pass over Vanscoy (discussed below).

"The total number of flashes observed by the CLDN during the five-hour total lifetime of the thunderstorms that eventually pass over Vanscoy is (524 $\pm$ 52)."

- For the estimate of the amount of excess NO2 you integrate between 60 and 85 (3 hours?). You multiply this excess NO2 with the area of the heavy-precipitation cell (30+- 3 km2). Is not the air mass area (with elevated NO2) 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 NO2 area. I think that the area you use is not representative for the integrated NO2 you estimated for 3 hours. A thunderstorm also has a complicated vertical structure as illustrated schematically in the Langford et al. (2004) paper.

We have recalculated the area of the thunderstorm by looking at the area of the thunderstorms upwind of Vanscoy. The remnants of two additional storms to the one shown in Fig. 3 pass over Vanscoy within a few hours of their formation. We now take the area to be the maximum area of the three storms from the CAPPI radar images available from 20:00 UT in 10 minute intervals. Using the wind speed calculates the entire air mass that passes over Vanscoy, and does not take into account that not all the air has been exposed to lightning activity.

- In the introduction some information on O3 and NOx is given, however it would also be useful to add something about O4 (first mentioned on page 10068). Explain more in detail the reasons for using O4 in the paper and add some references that

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introduced the O4-method to verify the influence of multiple scattering.

We have added the following discussion on O4 in Section 4.

"O4 concentrations are related to concentrations of oxygen, and, in the absence of an increase in the oxygen vertical column, are expected to be constant (e.g., Wagner et al., 2002). Hence a maximum in O4 such as the one observed in Fig. 5 is evidence of multiple scattering through the atmosphere, in this case due to the thick clouds associated with the thunderstorm. O4 SCD measurements are an established method of inferring the path length through the atmosphere in the presence of clouds (Erle et al., 1995; Wagner et al., 1998, 2002)."

- Page 10070, Line 21-24: "The difference between the observed NO2 slant column and the slant column calculated from the interpolated NO2/O4 ratio is the amount of NO2 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 NO2 SCD (thunderstorm passage). Is this a correct assumption (I would expect to have less thick clouds before and after)?

Figure 2 shows that the cloud opacity during and after the storm is approximately constant (ie. from 17:00 LT onwards). However, in Sect. 5.2 we find the maximum AMF of the clouds (Fig. 8 a and c). We have added a new section to address this comment (Sect. 5.3: Conversion to Vertical Column Densities) and a new Figure 9. We have scaled the residual SCDs derived using the ratio and AMF methods of calculating the path-enhancement using the enhanced AMF' calculated using the AMF method (using new Equation 6, derived from Equations 2 and 3). This has the effect of making the final VCDs attributed to lightning not entirely independent of one another.

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- 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.

We have re-calculated the AMF using a thick cloud extending between 1 and 10 km, to better agree with the observed maximum cloud thickness in Fig. 2 and from radar cloud tops (not shown). The AMF increases for all SZA. With other cloud thicknesses, the AMF lies between the thick cloud and no cloud calculations. Note that the derived AMF in Figs. 8(a) and (c) fall between the two. It should, however, be noted that the AMF calculated with a thick cloud is shown only for discussion, and is not used in any calculations.

Technical Corrections Abstract: Page 10064: Line 11: Change to "The enhanced NO2 columns are partly attributed to"

We have made this change.

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

Due to the recalculation of the flash production rate, this sentence now reads: "Using these two methods, the best estimate of the production rate is found to be

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(7.48+/-1.99)x10^{26} molecules NO2/flash from the UT-GBS and (6.47+/-1.72)x10^{26} molecules NO2/flash from SAOZ."

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."

We have made this change.

1. Introduction: Since the paper has the focus on tropospheric and not stratospheric measurements, 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).

We have removed pg. 10064 lines 25-26 and pg. 10065 lines 1-2 and 4-5. However, we leave pg. 10065 lines 19-22 to motivate the MANTRA campaigns.

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).

We have changed this sentence to read: "In the troposphere, NOx can act as both an ozone source and sink, depending on the concentrations of NOx and volatile organic compounds (VOCs), as well as the amount of available sunlight."

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.,

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2005; Beirle et al., 2006] Check order of the listed references (chronological).

We have changed these lines to read: 8220;Recent estimates of the global annual production rate due to lightning lie between 1 and 20 Tg N/year (Huntreiser et al., 2002;Tie et al., 2002; Ridley et al., 2004; Boersma et al., 2005; Beirle et al., 2006; Martin et al., 2006).

2. Instruments: Page 10067, Line 4: Write out DOAS.

We have made this change.

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.

We have made this change.

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.)

Hail was observed during the thunderstorm, and we have changed the phrase to "A cell of heavy rain and hail".

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.”*

We have made this change.

5. Derivation of lightning-produced NO2: Page 10070, Line 3-5: Add some references for these two used methods.

To the best of our knowledge, these methods are original to the paper. We have clarified this by changing the abstract, pg. 10064 lines 12 -13 to read: “Two new methods are used to separate the NO2 enhancements into contributions from the longer path length and production by lightning.” We have also changed pg. 10070 lines 3-5 to read: “Two new methods have been used to separate the observed enhancement of NO2 into a portion due to path-enhancement and a portion due to lightning production.”

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)?

The ozonesonde launched 28 August in the morning before the thunderstorm did not reach the stratosphere. An average of all the sondes was used since the ozone profile did not vary significantly during the campaign.

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

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In the original paper the value (5.8 +/- 2.9)x10^{26} molec/flash is given as both NO2 (pg. 13) and NOx (pg. 14).

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.

We have reworded this section to read: “Ridley et al. (2005) find that the production of NO from intra-cloud flashes is of the same order of magnitude as from cloud-to-ground flashes. No correction has been made to the flash frequency observed by the CLDN to account for intra-cloud flashes.”

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

We have made this change.

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

We have changed this sentence to reflect the new range of all of the estimates. (6.11 8211; 7.48).

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

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We have made this change.

References: Page 10075, Line 27: Change to "Boccippio".

We no longer cite this reference.

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

We have changed the captions of Figs. 1, 5, and 6 to include this information. We have also added SZA and time information when referring to Fig. 1 on pg. 10067, line 15.

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

The cloud height is the base height of the three cloud layers. We have added the following sentence to the figure caption: "First, second, and third heights are the base heights of the three layers of cloud."

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

We have added longitude and latitude markings on the edge of Fig. 3 and in Fig. 4.

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

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reflectivity).

We have changed the figure caption to “Precipitation rate at 17:00 LT (23:00 UTC, SZA 63) calculated from the Environment Canada radar measurements in Radisson, Saskatchewan on 28 August. Vanscoy is indicated by the red arrow.”

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

We have changed the figure caption to “Lighting flash data for the one-hour period beginning 28 August at 17:00 LT (23:00 UTC, SZA 63) in Vanscoy (X) from the Canadian Lightning Detection Network (CLDN) superimposed on a visible GOES image taken at 17:00 LT. Each plus (+) and minus (-) represents an individual lightning flash.”

Both cloud-to-ground and intra-cloud flashes are detected by the CLDN, as discussed in Sect. 3.

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

We have made this change.

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