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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.

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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.

The authors describe the use of ground-based zenith-sky differential absorption spectroscopy (DOAS) to measure the enhancement in the NO2 slant column during a thunderstorm, and have combined these results with additional information from nearby Doppler radar observations and the Canadian Lightning Detection Network (CLDN) to estimate the NOx production per lightning flash. The flash production rates so obtained are consistent with previous estimates. However, this agreement may be *coincidental since the authors make a number of simplifying assumptions in their analysis and the actual uncertainties in the derived rates are probably at least an order of magnitude larger than the stated uncertainty limits. Unless these uncertainties can be properly quantified and reduced, these results will be of limited interest to the scientific community.*

There are only a handful of papers that deal with measuring the NO2 or NOx production rate per flash from ground-based UV-Visible instruments. This paper builds on this previous work, demonstrating the ability of DOAS measurements to detect lightning-produced NO2, and combines these measurements with Doppler radar and CLDN data measurements to quantify NO2 production. While several assumptions were required, they have been described in the paper. These assumptions are discussed further below, along with the estimated uncertainties that result.

Specific Comments
The approach used to estimate the flash production rate in this paper consists of two parts: i) estimating the enhancement in the NO2 slant column that is due to lightning, and ii) identifying the number of lightning flashes that contribute to this enhancement.

The fundamental measurements of the NO2 slant column appear to be sound. However, the UT-GBS and SAOZ measurements are treated as if they are independent when in fact the SAOZ instrument was not operating when the NO2 maximum occurred and the missing peak was interpolated using the UT-GBS measurements. Thus, there was only one independent measurement of the NO2 slant column maximum and there can be only one independent estimate of the flash production rate. The SAOZ data can only be used to support the validity of the UT-GBS measurements through the good agreement before and after the NO2 maximum.

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The data from SAOZ and the UT-GBS were treated completely independently of one another. This is clarified in Sect. 5.1 by changing lines 11 to 14 to "Two independent exponential fits were made to the observed NO2/O4 ratios for the SAOZ and the UT-GBS, omitting the points between 65 and 82, which correspond to the beginning and end of the observed maximum in the ratio, These fits for both instruments are shown in Fig. 6b, along with the measured value of the ratio, which is used before and after the peak (before 65 and after 82)." The missing peak in the SAOZ data accounts for why the SAOZ values are slightly lower than those from the UT-GBS.

The authors use two approaches to separate the components of the observed slant column change that is due to an increase in the mean NO2 concentration along the light path from that simply due to photon path enhancement by multiple scattering within the cloud. This analysis seems reasonable. However, the flash production rate estimate is based on the erroneous assumption that the quantity derived from this analysis represents the vertical column of lightning-produced NO2. This is not the case. The derived column may be due to lightning, but it still lies along the mean path traversed by photons that have been multiply scattered within the cloud. The integrated NO2 column along this path may be many times larger than the vertical column.

We have added a new section (Sect. 5.3: Conversion to Vertical Column Densities) and new figure (Figure 9) to address this comment. 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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Also, it has not been demonstrated that the enhanced NO2 is exclusively due to lightning production. This is probably a reasonable assumption, but since Vanscoy is located within 20 km (albeit upwind) of a medium sized city (Saskatoon, pop. 200000) some of the NOx may have been entrained into the storm from the boundary layer. The authors should estimate this contribution. Were there any observations of NO2 enhancement in convective clouds that did not have lightning during the study period?

This was the only enhancement observed during the 43-day field campaign. As shown by the radar measurements, Vanscoy is upwind of Saskatoon, and pollution from the city is minimal. We have added a comment on this in Section 3: “Vanscoy is located upwind of Saskatoon for the duration of the storm, making it unlikely that NO2 enhancements are due to the upward transport of anthropogenic NO2.”

Neglecting for the moment that the derived vertical columns may be significantly overestimated, there are additional problems with the approach used to convert this quantity into a flash production rate. The authors calculate this rate using estimates for the storm cell area, and the number of lightning flashes that occurred over the nearly three hour interval for which the enhancement was observed. In other words, it appears that the storm cell was treated as a cylinder where all of the NO2 produced over the lifetime of the storm remained and was isotropically distributed.

As noted below, three storm cells passed over Vanscoy in the three hour period that thunderstorms were observed (shown in Fig. 3). We have recalculated the area of the storm to reflect the total area with lightning activity. (Explained in more detail below.) The thunderstorms passed over Vanscoy, and so NO2 was transported with the air masses. If the storm (and NO2) had been stationary, there would be no need to integrate over time (as in Sect. 6).

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A cylinder is a poor model for the thunderstorm structure since much of the NO2 will be in the anvil either as a result of transport or direct production by intracloud (IC) flashes. Even assuming that all of the lightning-produced NO2 was confined to the heavy precipitation cell for which the authors inferred an area of (30+/-3) km2 from the Doppler Radar plot (Figure 3) the uncertainties are much underestimated. This area corresponds to a cell radius of 3.09+/-0.22 km, but since this information is derived from a plot that has a resolution of 1 km/pixel, the cell radius cannot possibly be determined with a precision better than +/-1 km. A radius of 3+/-1 km corresponds to an area of 28+/-13 km2 increasing the uncertainty in the estimated area from 10 to 50%. A closer look at Figure 3 suggests that the cell radius really cannot be determined more accurately than about a factor of two. This is neglecting the uncertainties arising from the 90-minute time delay between when the radar scan was made and when the NO2 maximum was observed.

We have recalculated the area of the storm, by taking the maximum area of thunderstorm cells upwind from Vanscoy. The area is calculated by counting the individual pixels (ie. counting in km2) of the heavy rain areas in the CAPPI radar images, available in 10-minute intervals from 20:00 UT (which was start of the thunderstorm to the west of Vanscoy). Therefore the uncertainty is much less than if the radius of the storm was calculated from Fig. 3. This is now explained in more detail in Sect. 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 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."

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In addition, there are at least three reasons why the assumption that NO2 is isotropically distributed and conserved is invalid. First, convective cells are characterized by strong updrafts and downdrafts with air entrained from the boundary layer air and middle troposphere and vented into the upper troposphere from the anvils. The residence time of any NO2 produced by lightning will depend on many factors including the updraft velocity.

Due to these updrafts, NOx produced by cloud-to-ground flashes is transported up into the anvil of the storm cloud, as discussed in Sect. 5.3. NO2 is transported with the thunderstorms as they move over Vanscoy. Since the lifetime of NOx is on the order of several days, the decrease in the peak of the NO2 seen in Figs. 5, 7, and 9 is a result of transport.

Second, individual thunderstorm cells typically have lifetimes that are much shorter than the 3-hour period. Not all of the NO2 produced within one cell will be incorporated into the new cells of a multicellular storm. How did this particular storm evolve?

Radar imagery from Radisson, SK shows a series of storm cells forming to the west of Vanscoy at 20:00 UT, near the Alberta-Saskatchewan border, and traveling to the east, reaching maximum intensity and size over Vanscoy, and dissipating to the east of the city. Three thunderstorm cells passed over Vanscoy in total, with a maximum area of 61 km2. This is addressed in the comment above.

And third, lightning discharges do not produce NO2 directly and DOAS can only detect those molecules of NO that have been photochemically converted to NO2. The equilibration between these species is fast and the partitioning will vary with

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photolysis rates and hence altitude. Near the top of the storm much of the NOx will be in the form of NO and go undetected. Note that is really no such thing as an NO2 flash production rate because of this partitioning.

While it is true that lightning directly produces NO, NO2 is indirectly produced, and so an NO2 flash production rate can be calculated. Other papers that describe lightning-enhanced NO2 measurements use a variety of methods to derive NOx flash production rates. This introduces more uncertainty into the rate, so we chose to report only NO2 rates.

Finally, it is not obvious that the assumed value of the hourly lightning flash rate from the CLDN refers exclusively to the thunderstorm cell where the measurements were made.

We have recalculated the number of lightning flashes affecting the storm by counting the individual flashes detected by the CLDN upwind of Vanscoy. The volume of air that passes over the instruments experienced a total of 524 flashes over the lifetime of the storm. Section 3 contains a comment on the new method.

“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 ± 52).”

Also, Figure 4 shows a number of positive cloud-to-ground (CG) flashes to the west of the measurement site. These are typically of much higher energy than negative CG flashes and may have a much different NOx production rate. Conversely, IC flashes (which the CLDN is also somewhat sensitive to) probably have similar or lower production rates than negative CG flashes.

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The flash production rate we have calculated is an average over positive and negative CG and IC flashes. There is no way of determining which type of flash produced which NO2 molecule.

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