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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.

Anonymous Referee #2

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General Comments

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.

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.

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. 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

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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?

Neglecting for the moment that the derived vertical columns may be significantly over-estimated, 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.

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.

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

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troposphere from the anvils. The residence time of any NO2 produced by lightning will depend on many factors including the updraft velocity.

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?

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 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.

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. 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.

In summary, while the authors make a good case that they have observed an enhancement in column NO2 that is due to lightning, they have much work to do to quantify these results and derive a useful flash production rate from them.

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