# FLORIDA THUNDERSTORMS: A FAUCET OF REACTIVE NITROGEN TO THE UPPER TROPOSPHERE

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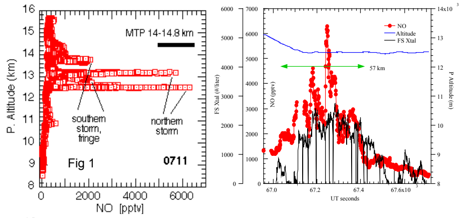
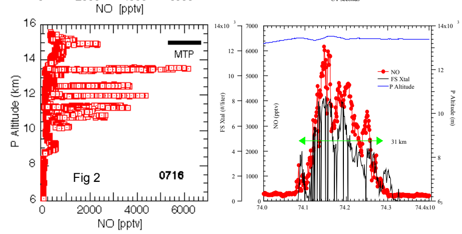
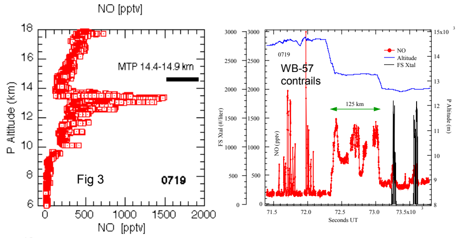
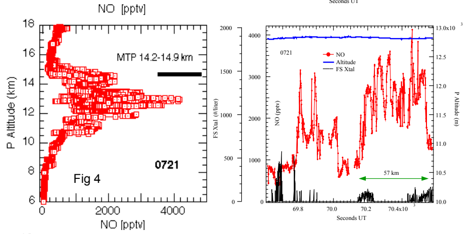
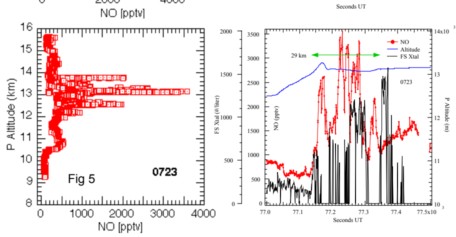
**CRYSTAL-FACE WB-57**

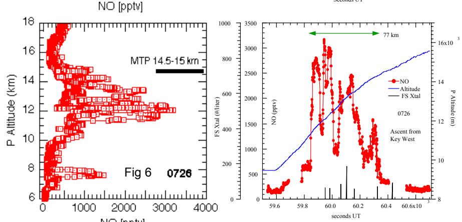


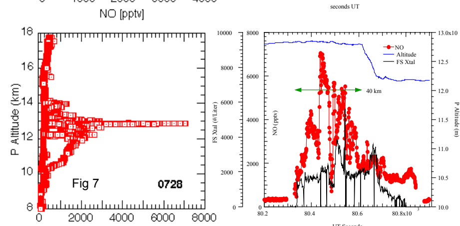



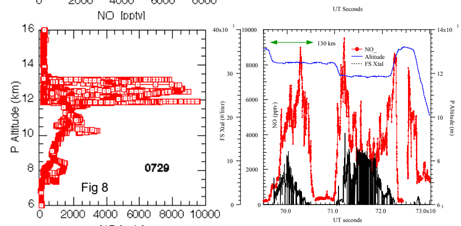



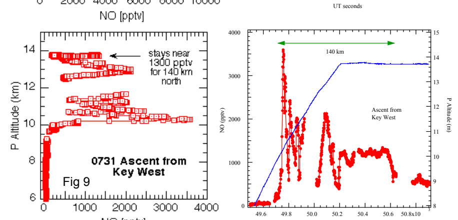




## INTRODUCTION

Global annual estimates of lightning production of $NO_x$ ($NO + NO_2$) are mostly in the range of 2-20 Tg(N)/yr. Recent estimates from first principles are 12-14 Tg(N)/yr [Price et al., 1997]. Most current global chemistry-transport models (CTMs) use a value of 5±2 Tg(N)/yr based on deriving general agreement between predictions and available air-borne measurements. In contrast to the budget estimate uncertainties, satellite sensors and extensive ground-based networks have provided a refined climatology of the global distribution of flash frequency (intra-cloud (IC) and cloud-to-ground (CG)) and their seasonal variations. As a result the global average flash frequency has decreased from earlier estimates of about 100 $sec^{-1}$ to 40-50 $sec^{-1}$.

From a chemistry perspective (e.g., tropospheric ozone) the altitude distribution of $NO_x$ generated by thunderstorms is as important as knowledge of the total production. For example, Tie et al. [1998] showed that their CTM gave equally good agreement with airborne observations in the middle and upper troposphere by using a source of 7 Tg(N)/yr distributed uniformly by mass over all altitudes of the storms or by using a source of one-half as much distributed over only the upper region of the storms. Previous studies (ELCHEM, STERAO, EULINOx) have demonstrated large increases of $NO_x$ in the anvil outflow of thunderstorms or above about 8 km as would be expected and these observations have impacted recommendations for sub-grid parameterization in CTMs [Pickering et al., 1998].

The lifetime of $NO_x$ in the UT is much longer than in the lower troposphere, several days to a week versus less than a 1/2 day, so $NO_x$ from lightning or that transported from near surface source regions through the storm into the UT can have chemical consequences for several lifetimes or more or over areas much broader than the scale of a typical storm. The integrated chemical impact of the storm after maturity and subsequent mixing/dilution requires detailed modeling because the chemistry can be highly non-linear. With the longer lifetime and because daytime partitioning of $NO_x$ strongly favors NO in the UT, NO is a reasonably good tracer of lightning production and the downwind impact.

## OBSERVATIONS

Figures 1-9 (left) present altitude distributions of NO from WB-57 flights in or near the anvil outflow regions. (Contrail intercepts have been eliminated.) Peak mixing ratios of NO can be enormous, 40-50 times typical background or extra-cloud mixing ratios of ~100-200 pptv for altitudes of 8-12 km. Mixing ratios of only 1000 pptv would be a strong perturbation to local rates of production of ozone. The apparent peaks of NO are the combined result of CG (usually known) and IC (unknown) activity and contributions from transport of $NO_x$ from the lower troposphere including the PBL (hereafter, LT/PBL). (See S. Wofsy/I. Xueref et al., Harvard group presentations that use $CO_2$ as a tracer of the LT/PBL influence.) LT/PBL $NO_x$ should be nearly conserved during the short time (~1/2 hr) of transport to the anvil region. The lower altitude input to the convective cell changes temporally, the cells also move with respect to source locations, and, from cloud resolving models, the input is not always from within the PBL but can be from near the top of the extra-cell PBL region. As well the PBL over south Florida can be very inhomogeneous vertically and horizontally depending on proximity to major urban areas and the strength of the convergent flows.

What appear to be sharp peaks or thin layers of NO in the altitude profiles are usually broad regions (many km) of enhancement depending on how the aircraft probed the anvils. An example from a transect for each flight is given in the figures to the right of the altitude profiles. Elevated NO is usually found within the "visible" anvil cloud indicated here by the FS-Xtal data (particles/ice larger than 50 microns). The variability along a pass within the anvil is large but not enormous and the maxima are usually found on any pass within the central "thicker" part of the anvil and usually within the upper half of the anvil thickness. Indeed, by comparison with the nadir view of the anvil provided by GOES, higher NO correlates well with the "brighter" more intense regions of the anvil and also increases as the core region is approached.

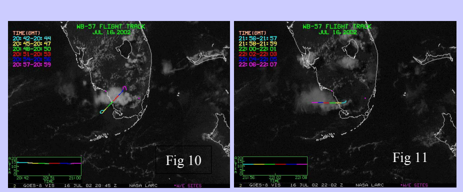
The anvil region acts as an integrator or accumulator of lightning production that occurs (and mixes) within the main and much more turbulent convective cell or from IC activity within that region or the anvil region. (There were only one or two reports of nearby lightning flashes by the flight crew during the program.)

Often the storms were not isolated but consisted of merged outflow from several or more cells. By comparison with the GOES images, many of the WB-57 flights investigated the peripheral regions (probably not so from the pilot's perspective!) of the anvils, near the top, along the edges, the downwind 2/3 etc. Nor did it investigate the lower regions of the anvil closer to the convective core but it is clear that large NO is found in the outflow between about 10 km to near the tropopause.

The 0721 flight is an example where a line of three storms was investigated over the west coast but not far downwind of a very large and very electrically active complex of storms. The high extra-cloud NO of order 800-1000 pptv likely results from the three anvils studied being imbedded in air already impacted by outflow from intense lightning in the large upwind complex.

## 0716 STORM:ESTIMATE OF TOTAL $NO_x$ VENTED TO THE UT

On 0716 the WB-57 extensively probed the anvil of an isolated storm at several altitudes (Fig. 2, left) that developed starting about 1915 UT (69300 sec) over central Florida west of Miami. The outflow had moved off the west coast by 2300 UT when the aircraft left the anvil to return to Key West. Four longitudinal passes and two and a half transverse passes were made at different altitudes. Examples of transverse (#5) and longitudinal (#10) passes are shown in Figs. 10 and 11.

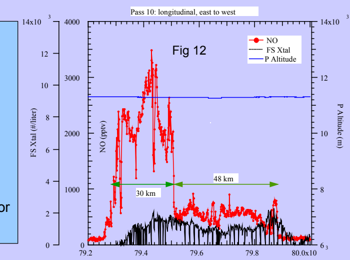



CG lightning started between 1900 and 1915 UT and essentially stopped by 2100 UT (Table 2). It is also evident from the GOES images, consistent with the drop in CG activity, that the anvil "disconnected" from the cell core around 2115-2130 UT. Thus it is likely that there was little further input of reactive nitrogen to the outflow after this time, but we have no record of possible IC activity. The disconnect time is also consistent with the nadir area of the anvil from GOES which remained relatively constant at 4200 ± 300 $km^2$ after pass # 5 and as the anvil moved west and until the aircraft left the storm.

Fig. 2 (left) shows the enhanced NO for pass 5. High NO was recorded over a distance of ~31 km. Relative to extra cloud air, the higher NO was accompanied by higher $O_3$ and CO but lower $CO_2$ although these trends were not always as clear on other passes. Within the more intense parts of the anvil, well away from the cloud boundaries, the $O_3$, CO and $CO_2$ mixing ratios should reflect the input to the storm from lower altitudes. Fig. 12 (below) shows data from a longitudinal pass, #10. As the aircraft entered cloud from east to west, higher NO occurred in the upwind region over a distance of ~30 km and then lower more uniform enhancement occurred over a distance of ~48 km until the aircraft exited the downwind anvil at 79880 sec. The downwind anvil reflects air exposed to less electrical activity or air that was transported vertically during the early stages of the storm development. These trends and average or median mixing ratios for individual passes, were relatively consistent in the other passes made at different altitudes. That is, the upwind anvil contained larger NO over a distance of 24-31 km; the downwind region lower NO over a distance of 46-57 km.

| Table 2: 0716 Activity | |
|---|---|
| Time (UT) | Accumulated CG Flashes |
| 1900-1930 | 16 |
| 1930-2000 | 142 |
| 2000-2030 | 244 |
| 2030-2100 | 269 |
| 2100-2200 | 277 |

(244 CG flashes occurred prior to pass # 5 near 2037 UT)




To arrive at an estimate of the amount of NO or $NO_x$ contained within the anvil after disconnect we approximated the anvil as an upwind circular region of higher NO of diameter 26 km, and a rectangular downwind region of lower NO of length 50 km and width 20 km, and each for the altitude bins shown in Table 2. The table also gives the total $NO_x$ estimate for altitude bins centered around the pass altitudes and for the entire volume of the anvil outflow from 10.5 km to near the tropopause. ($NO_2$ was determined from NO using clear sky J-values and the average $O_3$, T, P for each pass. J-values may be significantly perturbed by the anvil particles, but because the $NO/NO_2$ ratio is so large only relatively small errors result.) The sum of these areas (1531 $km^2$) is significantly smaller than the nadir view area (~4200 $km^2$) determined from the GOES images but the aim is to be conservative.

| Table 2 | 26 km diam High NO area | | 20 km x 50 km Low NO area | |
|---|---|---|---|---|
| Altitude bin km | median NO | molecs $NO_x$ | median NO | molecs $NO_x$ |
| 14-15 | 1000 pptv ? | $2.6 \times 10^{27}$ | ? | ? |
| 13-14 | 3317 | 10.3 | 1650 | $9.7 \times 10^{27}$ |
| 12-13 | 2497 | 9.1 | 952 (low?) | 6.5 |
| 11.5-12 | 2305 | 4.3 | 805 | 2.8 |
| 11-11.5 | 2096 | 4.8 | 587 | 2.5 |
| 10.5-11 | 1252 | 3.3 | 362 | 1.8 |
| Total | | $34.4 \times 10^{27}$ | | $23.3 \times 10^{27}$ |

The total estimate of $58 \times 10^{27}$ molecules is considered a lower limit. It is unlikely to be low by more than the total estimate for the low $NO_x$ downwind estimate.

Result: a range of 58-81 x $10^{27}$ molecules of $NO_x$

We stress that the total volume estimate represents the combined production from lightning within the anvil (IC), from lightning within the core that is transported to the anvil, and $NO_x$ transported from the PBL/LT, all of which is vented into the UT. It is not the total produced by the storm because we have no information on mixing ratios of reactive nitrogen at lower altitudes, from for example CG discharges external to the core, downdrafts, etc. To put the total into perspective, 9.6-13.4 x $10^4$ moles of NO (2.9-4.0 tonnes of NO) is roughly equivalent to the $NO_x$ emissions from a flight of a Boeing 757 aircraft if it could fly for 100-150 hours.

## Production of NO per Lightning Flash (0716 Storm)

If we assume that most of the lightning produced $NO_x$ appears in the anvil and that lightning accounts for 75% of the NO observed in the anvil (S. Wofsy, I. Xueref talk) then an estimate of the NO production per flash (P) appearing in the outflow can be made. We have to rely on the average IC/CG ratio of 2-2.5 for south Florida storms derived from climatology by Boccippio et al. [2001], giving ~277 x 2.25 = 623 IC flashes. (Applying an average ratio obtained over many storms to individual storms is a big assumption.)

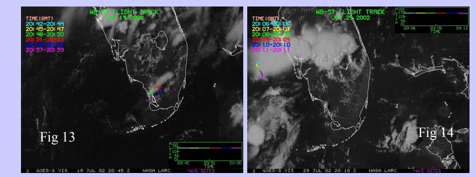
If P(IC) = P(CG), all flashes equivalent in production, then the production ranges from 0.49 to 0.68 x $10^{26}$ molecules of NO per flash. Using 45 $sec^{-1}$ as the average global flash rate, then extrapolating a single storm to represent the global amount would yield an estimate of lightning production to the upper troposphere of only 1.6-2.2 Tg(N)/yr.

If we make the common assumption that P(IC) =0.1 P(CG), then P(CG) ranges from 1.3-1.8 x $10^{26}$ molecules of NO per CG flash. (CG may produce more NO per flash but a smaller fraction of the production could be vented to the UT.)

Either estimate is significantly lower than P(CG) = 6.7 x $10^{26}$ molecules of NO per flash and P(IC)=0.1 P(CG) used commonly in current CTMs.

## BROAD COMPARISON OF THREE ISOLATED STORMS

Three flights investigated isolated storms and provide an interesting contrast between the maximum NO mixing ratios observed, the anvil/storm nadir GOES area, and the number of CG flashes derived from the ground network (Table 3). (Electrical activity is summarized in the poster by Lesley Ott et al., U. Maryland.) The 0719 storm was small, short-lived, and electrical activity (CG) very low. The 0716 storm was of moderate size and CG activity while that of 0729 was large and extremely active (Figs. 10 and 11, 13, 14). We do not have IC activity for any of the storms and again resort to the average IC/CG ratio of 2.0-2.5 for S. Florida. The total number of flashes could have been roughly 3-3.5 times the CG accumulation. However using either the CG flash or estimated total flash accumulation, the number varied by a factor of 400 to 500 while the maximum mixing ratios of NO varied by about a factor of 7 (~1300 to ~8800 pptv).



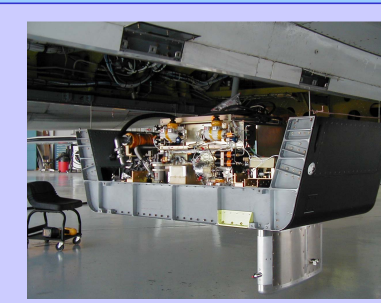

Under similar wind speeds at anvil altitude, similar anvil thickness (cf. left-most Figs. 2, 3 and 9), and assuming that the production of NO molecules per flash was roughly constant, then a very simple view is that larger anvil volumes are produced by larger or stronger updrafts and therefore accompanied by more frequent electrical activity. Thus the number of flashes divided by the anvil volume and the average mixing ratio might be roughly constant for storms with some electrical activity. Using the nadir GOES area as a proxy for the volume, and the range of maximum NO observed as a proxy for the anvil average mixing ratio results in the last two rows of Table 3 (below). The 0716 and 0729 scaling ratios are different by about a factor of two. That for the small 0719 storm is much lower but it is likely that the electrical activity was so low that a much larger fraction of the enhanced NO observed in the outflow was due to transport of $NO_x$ from the PBL/LT. Of course, the larger storm (0729) produced a much larger mass of NO to the UT than the other two, but the simple scaling suggests why the range of maximum mixing ratios is only a factor of 7 for all three storms or indeed much less (factor of ~1.6) if just the two more electrically active storms of 0716 and 0729 are considered.

Finally, since these three storms reached similar altitudes, it is clear that the flash rates do not scale as the 5th power of cloud top height [cf. Price et al., 1997].

Table 3: Comparison of 3 Isolated Storms

| | 0719 | 0716 | 0729 |
|---|---|---|---|
| CG flashes | 7 | 277 | 3400±200 |
| Calc. IC + CG flashes | 23 | 900 | 11050 |
| Max mr range (pptv) | 1000-1600 | 4500-6100 | 8000-9500 |
| GOES Area ($km^2$) | ~1200 | ~4200 | ~15300 |
| Wind speed (m/s) | 15-25 | 10-25 | 10-35 |
| Max altitude (km) | 14+ | 13.5-14 | 14+ |
| Duration* (hr) | ~1 | ~2 | ~3.5 |
| Flash rate (#/min) | <0.4 | ~8 | ~53 |
| CG/(area x mr) | $0.5 \times 10^{-5}$ | $1.2 \times 10^{-5}$ | $2.5 \times 10^{-5}$ |
| (IC+CG)/(area x mr) | $1.5 \times 10^{-5}$ | $4.0 \times 10^{-5}$ | $8.3 \times 10^{-5}$ |

*Duration of electrical activity or to time of leaving storm



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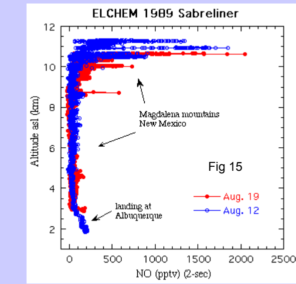
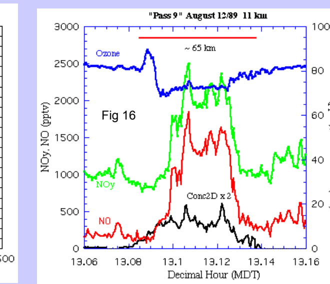
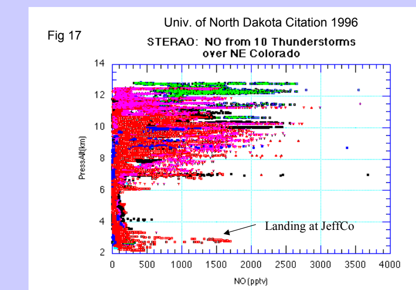
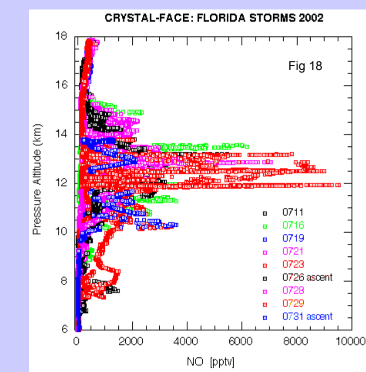
## COMPARISON WITH STORMS OVER NEW MEXICO AND COLORADO




Figs. 15, 17, and 18 give a comparison of NO mixing ratios from orographically generated storms over New Mexico (ELCHEM), from storm complexes over the north-eastern plains of Colorado (STERAO), and from Florida (CRYSTAL-FACE). The figures can be considered broad averages of the NO altitude distribution near the time of storm maturity. (Note that the altitude and mixing ratio scales are different in each figure.)

In the New Mexico study the Sabreliner investigated lower altitudes and beneath one of the storms. In the Colorado study the NOAA P3 studied the low altitude periphery of the storms. Elevated $NO_x$ attributable to lightning was not found in either experiment but the chances of seeing results from individual flashes are small. Unlike the anvil region the output from flashes external to the core(s) is not integrated by dynamics. Downdraft regions containing significantly enhanced NO were also not found. (The larger NO found at low altitudes in Figs. 15 and 17 are from pollution near the base airports.)

As in examples shown previously from CRYSTAL, the apparent spikes in the altitude profiles are broad regions of elevated NO determined by how the aircraft transected the outflow. An example of the NO, $NO_y$ (rear-facing inlet), $O_3$ and large particle behavior from one of the New Mexico anvil transects is shown in Fig. 16. (We do not imply that much larger NO enhancements than shown in the figures above are never observed. Large, short term, a few seconds, spikes of NO attributable to individual flashes have been observed but relatively rarely, e.g., in EULINOx up to ~30 ppbv and in STERAO up to ~19 ppbv.)

All three types of storms vent high mixing ratios of reactive nitrogen to the UT, above about 8-10 km to anvil top. In the Florida cases it is clear that the venting can occur up to very near the "local" tropopause, although larger mixing ratios of NO were usually seen more than a km below the tropopause. Indeed it seems that a large fraction of the lightning and PBL/LT transport is vented to the UT but quantifying "large" remains an issue. Thus all of these studies show that the direction of parameterization for models should be to vent most of the reactive nitrogen (and other LT/PBL constituents) to the UT.

There is a trend in the range of maximum NO mixing ratios for the three continental systems: New Mexico 1000-2000 pptv, Colorado 1500-3000 pptv, and Florida 4000-8000 pptv. This result does not mean that individual Florida storms produce a larger mass of reactive nitrogen compared to the plains storms. The latter are usually longer-lived than Florida storms due to regeneration of convective cells as the storms translate. On the other hand, given the complexity of storm formation, of dynamics and mixing, and of lightning physics and the temporal trends in all of these factors over the lifetime of deep convection, it might be surprising that the range of observed mixing ratios is less than an order of magnitude.

## Conclusions

Given the large number of thunderstorms that occur over Florida in the June through mid-August period, the region acts as a faucet of reactive nitrogen to the UT which will have a large impact on photochemical processing downwind and over several lifetimes of $NO_x$. These studies have shown that a large fraction of lightning production is vented to the UT and model parameterizatons should reflect these findings. It is more important to represent the distribution in the UT properly compared to lower altitudes because of the much longer lifetime of $NO_x$ at these altitudes.

A lot of analysis remains to be done!