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Lightning-produced NO_x during the Northern Australian monsoon; results from the ACTIVE campaign

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Abstract. Measurements of nitrogen oxides onboard a high altitude aircraft were carried out for the first time during the Northern Australian monsoon in the framework of the Aerosol and Chemical Transport in Tropical Convection (ACTIVE) campaign, in the area around Darwin, Australia. During one flight on 22 January 2006, average NO_x volume mixing ratios (vmr) of 984 and 723 parts per trillion (ppt) were recorded for both in and out of cloud conditions, respectively. The in-cloud measurements were made in the convective outflow region of a storm 56 km south-west of Darwin, whereas those out of cloud were made due south of Darwin and upwind from the storm sampled. This storm produced a total of only 8 lightning strokes, as detected by an in-situ lightning detection network, ruling out significant lightning-NO_x production. 5-day backward trajectories suggest that the sampled airmasses had travelled over convectively-active land in Northern Australia during that period. The low stroke count of the sampled storm, along with the high out-of-cloud NO_x concentration, suggest that, in the absence of other major NO_x sources during the monsoon season, a combination of processes including regional transport patterns, convective vertical transport and entrainment may lead to accumulation of lightning-produced NO_x, a situation that contrasts with the pre-monsoon period in Northern Australia, where the high NO_x values occur mainly in or in the vicinity of storms. These high NO_x concentrations may help start ozone photochemistry and OH radical production in an otherwise NO_x-limited environment.

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1 Introduction

Nitrogen oxides or NO_x ($NO_x = NO + NO_2$), constitute one of the primary sources of fixed nitrogen available in the atmosphere. Furthermore, NO_x plays a key role in the balance of the ozone budget by catalytically mediating the oxidation reactions that produce ozone (Crutzen, 1979; Chameides et al., 1973). Lightning-produced nitrogen oxides (LNO_x hereafter) play an important role in the chemistry of the troposphere, both by enhancing the potential of a given airmass to produce ozone catalytically and by affecting the concentration of the OH radical and thus the oxidizing efficiency of the atmosphere (Stockwell et al., 1999; Labrador et al., 2004). Produced in and around active thunderstorms, NO_x is readily transported by convection to the upper layers of the troposphere, where its lifetime increases to the order of days (Bond et al., 2001), and where it is subject to long-range transport and re-introduction to areas with low NO_x background levels, thereby increasing the potential for ozone production in NO_x-limited environments.

Of all the natural sources of nitrogen oxides, LNO_x remains the most uncertain; the violent nature of thunderstorms, along with the fast characteristic times of the lightning phenomenon, of the order of milliseconds, make it difficult to obtain precise measurements of its production. Measuring lightning-produced NO_x requires dedicated experiments and airborne platforms to sample the species, which usually involves flying in the convective outflow regions of thunderstorms. The difficulties and uncertainties associated with these tasks have resulted in the current LNO_x production estimates ranging from 1 to 20 Tg(N) yr^{-1}, with the most recent estimates settling around a 2–8 Tg(N) yr^{-1} range

(Schumann and Huntrieser, 2008). Given its role as an ozone precursor, the uncertainty in the production of LNO_x makes it difficult, in turn, to precisely estimate the global ozone budget.

Most LNO_x sampling experiments have been carried out in continental, isolated type convection (Ridley et al., 2004; Huntrieser et al., 2007; Huntrieser et al., 2002). Murphy et al. (1993) reported on airborne NO_y measurements over Northern Australia during January 1987, within the framework of the Stratosphere-Troposphere Exchange Project (STEP) (Russell et al., 1993) and established that the variability of both the NO_y mixing ratios and the NO_y/O_3 ratio indicated a source of NO_y in the troposphere, and that the most plausible source in the tropics is lightning production of NO_x. Similarly, Pickering et al. (1993) used photochemical models as well as data collected in cumulus clouds during the Stratosphere-Troposphere Exchange Project and the Equatorial Mesoscale Experiment STEP/EMEX and concluded that the most dramatic factor affecting ozone production in the upper troposphere during the period of those campaigns may have been lightning-produced NO_x; the authors estimated that production of ozone from 12–17 km in a region distant from active convection was 2–3 times higher than it would be without lightning, signifying a major role for lightning in upper tropospheric NO_x and ozone budgets in remote tropical regions. Although measurements of LNO_x and its impacts have been made for Northern Australia by Kondo et al. (2003), until recently, no measurements of lightning NO_x production in monsoon convection have been reported on for that region. In this work, we present the results of a flight carried out during the ACTIVE campaign, in January 2006, during the Northern Australian monsoon.

A description of the ACTIVE campaign, along with the main instruments used in deriving the data for this study will be given in Sect. 2. The general characteristics of the Northern Australian monsoon will be given in Sect. 3. This will be followed by a description of the meteorological conditions on the day of the flight, 22 January, in Sect. 4. Section 5 will offer a description of the Egrett's flight and its results. In Sect. 6 these results will be discussed and section 7 will offer the conclusions of this work.

2 Campaign description

The Aerosol and Chemical Transport in Tropical Convection field campaign (ACTIVE hereafter) (Vaughan et al., 2008) took place in Darwin, Northern Australia, between November 2005 and February 2006. The campaign, designed to study the impact of tropical convection on the composition of the tropical tropopause layer, was conducted during two distinct and markedly different regimes. The first phase, in November and December 2005, concentrated on deep, isolated pre-monsoon convection, particularly over the Tiwi islands, 100 km north of Darwin. The second phase, in January–February 2006, experienced a more organised and widespread monsoon convective environment (Keenan and Carbone, 2008). The campaign used a combination of airborne platforms and ground sensors to measure the composition of the atmosphere in the boundary layer, the free troposphere, the tropical tropopause layer (TTL) and lower stratosphere (Vaughan et al., 2008; May et al., 2008). The main high-altitude airborne platform used during ACTIVE was a Grob-520 Egrett, which sampled the anvil outflow region of storms at 12–14 km (Vaughan et al., 2008), as well as the surrounding background air. In eight of its campaign flights, two of which were during the active monsoon, the Egrett was fitted with a chemiluminescent NO-NO_2 sensor (Volz-Thomas et al., 2005). Of those, two flights, on 16 November and 3 December 2005, successfully sampled the outflow region of Hector storms during the first phase of the campaign; one flight, discussed in this work, measured NO_x in active monsoon conditions and another flight, on 10 February 2006, sampled a Hector storm during the monsoon break regime. On the ground, the dedicated Lightning Location Network (LINET hereafter) recorded the lightning's VLF transients, or strokes. LINET (Betz et al., 2004) was set up specifically for this experiment and comprised a ring of detection stations set up around the experiment area that permitted 3D spatial as well as temporal location of lightning strokes.

2.1 NO_x instrument

The NO-NO_2 instrument deployed onboard the Egrett was a modified version of the MOZAIC NOy instrument described in Volz-Thomas et al. (2005). For the instrument deployed in ACTIVE, the catalytic converter was replaced by a photolytic converter (Droplet Measurement Technology, Blue Light Converter, or BLC) for the specific conversion of NO_2 to NO. The BLC was modified by installing a quartz cell in order to prevent the memory effect encountered at reduced pressure during prestudies. The sensitivity of the chemiluminescence detector was enhanced to 0.7 cps/ppt by installing a second pump and a second ozone generator. The detection limit was 200 pptv at 10 Hz and 30 pptv for an integration time of 4 s. The conversion efficiency of the BLC was 70% on the ground and 50% at 150 hPa. The instrument was regularly calibrated before and after flight for both sensitivity for NO and conversion efficiency of NO_2.

There were two modes of operation during ACTIVE: Automatic operation, where the instrument alternated between NO and NO_x measurements every ten seconds, and manual operation by the flight crew. For the flight on 22 January, the instrument was switched manually between NO and NO_x measurement modes at approximately 10 min intervals. As the ambient NO/NO_2 ratio is not known during the NO_x measurement, an effective converter efficiency of 75% was employed for the calculation of NO_x at 150 hPa (increasing to 85% at 1000 hPa). The resulting uncertainty is +/−33% at

150 hPa (15% at 1000 hPa), due the fact that the efficiency would be 100% for pure NO and 50% for pure NO_2.

2.2 CO instrument

The CO detection instrument flown onboard the Egrett was based on the fast-response resonance fluorescence instrument for airborne measurements described in Gerbig et al. (1999). The instrument was regularly calibrated, both on ground and during flight and data were collected at a frequency of 1 Hz with a precision of ±2 ppbv (parts per billion volume) and an accuracy of ±3 ppbv (5% at mixing ratios >60 ppb).

2.3 Lightning detection network

A lightning stroke detection network comprising six detection stations was set up in an area of a radius of approximately 150 km centred on Darwin. LINET (Betz et al., 2004) is a VLF/LF detection network that utilizes a time-of-arrival technique for 3-D location of lightning strokes, which, along with a low detection threshold, allows for the discrimination of intra-cloud (IC) and cloud-to-ground (CG) discharges at distances of up to 100 km from a detection station. The accuracy of LINET degrades considerably after that distance as does its ability to distinguish between IC and CG discharges.

3 Meteorological conditions during the ACTIVE campaign

The onset of the wet season in Northern Australia typically occurs around October, with the pre-monsoon season. In a period marked by a strong diurnal cycle in convection, isolated deep storms over the Tiwi islands, 50 km north of Darwin, and squall lines over the continent develop on an almost daily basis. From December to April this regime alternates with phases of monsoon conditions, when a generally westerly flow predominates and there is widespread organized convection over land and ocean with no strong diurnal variation. During a monsoon the background atmosphere is characterized by widely varying amounts of stratiform rain, and a dense cirrus overcast, where the products of deep convective storms mix with aged cirrus from previous days' storms (May et al., 2008). In 2006 the monsoon's onset occurred on 13 January and produced three distinct phases; an active period, from 13 to 23 January, a suppressed monsoon period from the 26 to 2 February, and a monsoon break period from 3 February until the end of the experiment, when conditions were similar to the pre-monsoon regime.

Figure 1b shows a CO vertical profile composite plot using data from 7 Egrett flights between 20 January and 1 February 2006. Of note is the inverted vertical profile, wherein concentrations in the boundary layer were below 60 ppbv CO, whilst the maximum values, of between 70 and 75 ppbv CO, were measured between 7 and 9 km altitude. This contrasts

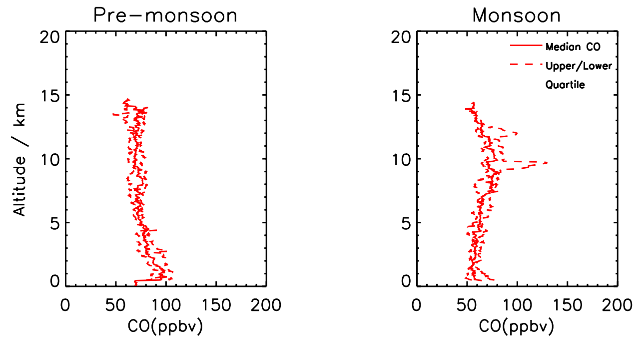

Fig. 1. Median CO vertical profile measured by the Egrett during **(a)** the pre-monsoon (left) and **(b)** monsoon (right) seasons. The dashed lines are the lower and upper quartiles of the measurements.

with the pre-monsoon phase (Fig. 1a), where the CO concentrations were highest (close to 100 ppbv) in the boundary layer and decreased with altitude to about 70 ppbv between 10 and 14 km. The low CO in the boundary layer during the monsoon shows that low-level air was free of anthropogenic pollution at this time.

During the active monsoon phase, the Darwin area experienced widespread convection with different levels of convective organization that included isolated storms as well as more organised structures (May et al., 2008). On 22 January, as the monsoon trough retreated north of Darwin, a large mesoscale convective system (MCS) developed in the experiment area that would dominate meteorological conditions for the next few days. This MCS appears to have been triggered by the convergence of three lines of thunderstorms to the southwest of Darwin that provided a strong initiation mechanism in an environment with high convective available potential energy (CAPE) and low convective inhibition, and abundant mid-level moisture and ice clouds from the earlier storms (P. May, personal communication, 2008). The system developed a distinct typhoon-like circulation and the area-averaged rainfall increased from 17 to approximately 55 mm day^{-1} with half of that rain being convective in origin. Similarly, the CAPE and relative humidity measurements registered the highest values of the active monsoon period (May et al., 2008). The system developed into a tropical low, with the atmospheric pressure dropping as low as 998 hPa on 31 January, and persisted as a stationary feature over the central part of Australia's Northern Territory until 2 February. Once stationary, the system's circulation had the effect of producing westerly winds of up to 20 m s^{-1} as well as advecting dry, continental air at midlevels to the experiment area giving rise to a suppressed monsoon period. Despite continuing high CAPE values, this resulted in a cap that limited convection to altitudes below 10 km which in turn led to rainfall rates averaging approximately 6 mm day^{-1} and very clean low-level air with CO vmr of less than 50 ppbv. This period lasted until the dissipation of the low pressure system on 2 February.

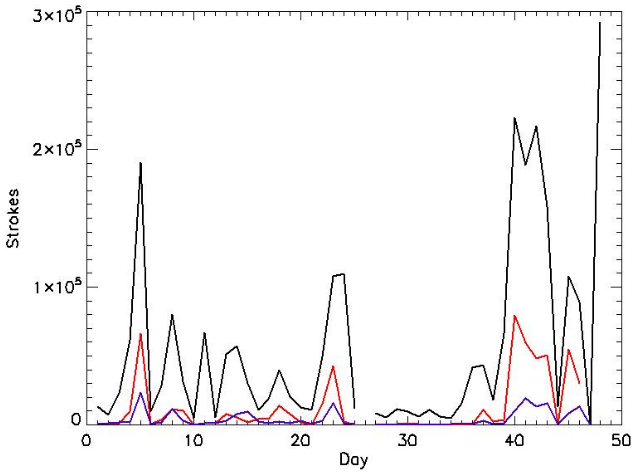

Fig. 2. LINET stroke count time series for the 1 January–12 February 2006, corresponding to the active monsoon in Northern Australia. Total strokes (black line), in-cloud-to-groundb strokes (red line) and cloud-ground strokes (blue line) are depicted. The rise in the count on 21 January corresponds to the onset of the mesoscale convective system. The gap in the data between 25–26 January corresponds to an inactive period of LINET.

Three days of clear skies followed before conditions similar to those of the pre-monsoon period were experienced over the experiment area, starting on 6 February. During this monsoon-break period the low level wind flow returned to easterlies. Rain rates, however, averaged approximately 8mm day (significantly less than the ~17 mm day^{-1} measured during the previous active monsoon period), indicative of lower average vertical motion. Similarly, convection recovered its continental pre-monsoon character and manifested itself in the form of intense afternoon isolated storms over the Tiwi islands as well as occasional squall lines over Darwin.

Figure 2 shows the LINET stroke time series for the 2006 monsoon in the ACTIVE experiment area. In the figure, the total stroke count does not equal the sum of cloud-to-ground and intra-cloud strokes; this is due to the fact that the LINET detection accuracy degrades at distances larger than 100 km. At such distances, if and when a stroke is detected by fewer than 4 LINET stations, then it is classified as undetermined. The trends in the stroke counts mirror the behaviour of convection throughout the three phases of the monsoon; starting on 5 January (day 5) there is a steady decrease in the average count as the monsoon regime is established. This decrease is only interrupted on 22 January (day 22), on the day of the formation of the mesoscale convective system, when there is an excursion from the trend and where over 40 000 strokes were recorded in the experiment area. During the next two days, at the peak period of influence of the MCS over the Darwin area, the count stayed at approximately 108 000 strokes day^{-1}, dropping back to 11 000 strokes on 24 January as the MCS's circulation advected dry continental air into the area thus suppressing convection. The decrease continues into the suppressed monsoon phase, with counts of between 5000 and 10 000 strokes day^{-1}. The stroke count rose to values typical of the pre-monsoon with over 220 000 on 9 February signalling the start of the monsoon break period; counts remained high until the end of the LINET recording period on 15 February. Table 3 shows the LINET stroke count for selected days of the different convection regimes sampled during ACTIVE. The first thing to notice is that the stroke activity during both the pre-monsoon and monsoon break periods is a factor of 5–10 higher than during the monsoon. The widespread, organized nature of the monsoon convection, along with the fact that unlike pre-monsoon and monsoon break convection, monsoon convection is a 24-hour phenomenon, however, resulted in more frequent storms covering a wider area than the episodic and oftentimes localized pre-monsoon and monsoon-break storms.

4 Conditions on the day of the flight

On 22 January deep convection ("system A" hereafter) started to organize at approximately 13:30 local time some 400 km south-east of Darwin, moving west. By 17:00 h LT, the convection had developed into a squall line and moved to approximately 200 km east south-east of Darwin. It eventually reached the city at about 21 hours LT and would later become part of the MCS described above. Along the coast SW of Darwin, another separate and much smaller storm system ("system B" hereafter) , made up of a series of storms aligned NE to SW in a 250 km-long line parallel to the coast and approximately 10 km inland, appeared at 14:23 LT in the satellite images. By 16:23 h LT this system, which preceded system A by at least 4 hours, had fully developed and started to move out to sea.

The LINET network registered system B's first 3 lightning strokes at 14:16 LT in its southernmost part. The period between 14:53 and 15:53 was the most electrically active, with 82 strokes, mostly on the southernmost part, where the storms were the most developed. Between 15:30 and 18:30 LINET recorded 27 strokes with the last 16 strokes being recorded between 18:30 and 19:30, mostly over land, along the system's trailing edge. By 19:23 h LT the system had moved entirely out to sea and was no longer electrically active. During the system's lifetime, from 13:30 to 19:30 LT, LINET detected a total of 128 strokes.

5 Egrett's flight on 22 January

The Egrett took off from Darwin at approximately 16:30 h LT to sample convection in system B's northernmost part. Figure 3 shows the Egrett's flight path on 22 January and the lightning activity detected by the LINET in the period from

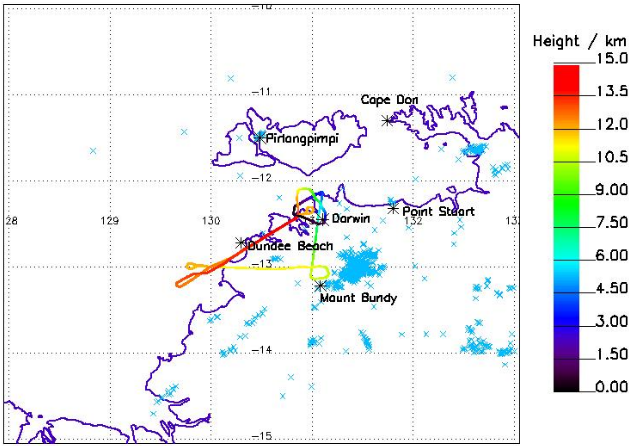

Fig. 3. Egrett's flight path on the 22 Jan flight, colour-coded to altitude. The black asterisks mark the position of the six LINET stations deployed during ACTIVE and the blue crosses are the LINET-detected strokes for the period 00:00–19:36 LT (i.e., from the start of the day to the end of the flight).

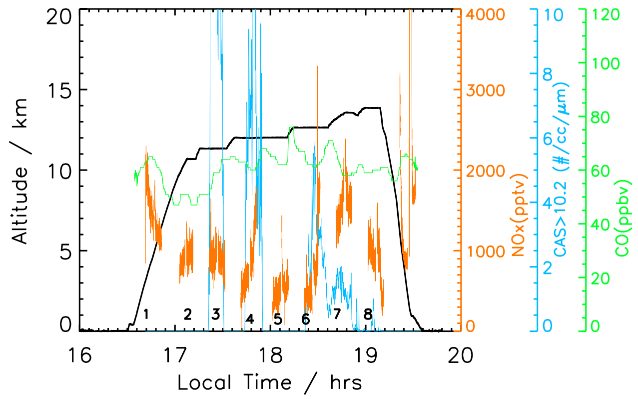

Fig. 4. Time series (in hours local time) of Egrett data on the 22 January flight: altitude (black line), CO (green), NO_x (orange) and particles larger than 10.2 μm (light blue line), used an indicator of in-cloud conditions, where N>1 indicates cloud. The numbers correspond to the different transects in the flight where NO_x was measured.

00:00 LT until the time the flight ended, at 19:36 LT. The aircraft first flew due south of Darwin to approximately 13° S in what was mostly cloud-free air, and then headed west towards the coast to sample system B's northernmost storm along the coast SW of Darwin. This storm produced only 8 strokes as detected by LINET, all between 16:49:47 and 16:49:48, and were registered as one cloud-to-ground and seven undetermined, all most likely part of a single lightning flash. Once over the coast, a series of SW-NE-SW transects were flown where the aircraft further probed the anvil outflow region of the storm. During the time of the flight the major part of lightning activity, as detected by the LINET, was concentrated some 400 km to the east of Darwin, in the region of the nascent system A (see also Fig. 5). The lightning activity in the figure, directly east of the Mount Boundy LINET station, and centred around 13° S, 131.5° E, started at approximately 18:05 LT and is as a result of the approaching system A to the east. Despite the proximity of the strokes to the Egrett's flight track, at that particular time the aircraft was flying along the coast and thus this activity had no impact on the NO_x measurements.

Figure 4 shows the NO_x time series for this flight. The NO-NO_x instrument was manually switched by the Egrett's crew between the NO and NO_x measuring modes at intervals of approximately ten minutes. Sections of this flight in and out of cloud were identified by using the Cloud and Aerosol Spectrometer (CAS), part of the Cloud Aerosol and Precipitation Spectrometer (CAPS) probe (Baumgardner et al., 2002), to measure particles larger than 10 microns; a value $>1\,cm^{-3}$ was taken to indicate in-cloud conditions. Once at altitude, the aircraft penetrated the outflow region of the storm in system B 5 times (transects 3, 4, 6, 7 and 8), which resulted in a series of large NO_x concentrations or

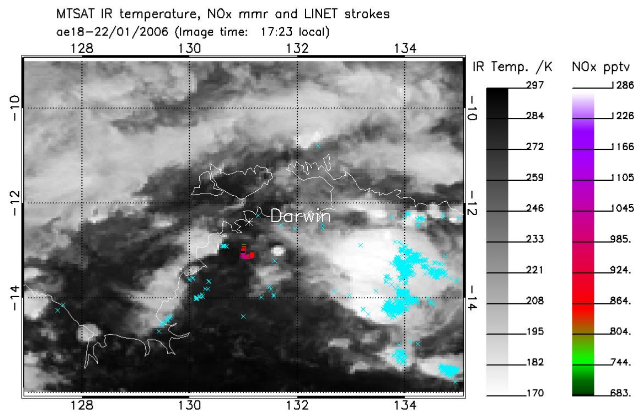

Fig. 5. Egrett's second cloud-free transect (coloured trace) colour-coded to NO_x and LINET strokes (blue crosses) between 14:05 and 17:11 LT (the sampled storm's electrically active period) on 22 January 2006.The transect occurred between 17:03 and 17:13 LT.

"spikes", with average mixing ratios of 901, 883, 741, 1570 and 870 pptv for each transect, respectively and a composite average of 984 pptv. Peak NO_x mixing ratios exceeded 3000 pptv in transect 6 (see Table 1). NO_x measurements in out-of-cloud conditions (spikes 1, 2 and 5 in Table 1 and Fig. 4) on the other hand, show average values of 1471, 920 and 525 pptv, respectively and a composite average of 723 ppt. The first two out-of-cloud transects occurred south of Darwin, the first during the climb to altitude from Darwin heading north, at altitudes between 3.1 and 6.4 km and distances of between 5 and 10 km from Darwin. However, due to the aircraft's proximity with Darwin and the low altitude of the sampling during the first transect, we have refrained from including this value in the out-of-cloud composite average mixing ratio. Of particular interest is transect 2 (see

Table 1. Egrett NO_x and CO transect parameters as measured by the Egrett on 22 January 2006.

Transect no.	Start (LT)	Dur. (sec)	Altitude (mts)	Avg. NO_x (pptv)	Peak NO_x (pptv)	Avge. CO (ppbv)	Remark
1	16:41:28	596	3300–6475	1471.3	2303	61.2	Out of cloud
2	17:02:56	496	10 658	920.3	1304	51.2	Out of cloud
3	17:21:27	595	11 330	901.6	1649	58.7	In cloud
4	17:41:28	594	12 005	833.9	1784	62.2	In cloud
5	18:01:39	578	12 018	525	1437	64.9	Out of cloud
6	18:21:29	592	12 600	741.3	3166	62.8	In cloud
7	18:41:35	586	13 420	1570.9	2390	59.3	In cloud
8	19:01:27	590	13 860–12 450	870.7	1550	56.6	In cloud

Fig. 5); the Egrett was at 10 km altitude, 50 km due south of Darwin and 19 km east and upwind of the storm in system B it would later sample. Likewise, the aircraft was approximately 300 km west of system A's centre of lightning activity. Between the start of system A's lightning activity period, at 14:05 LT and the end of this NO_x transect at 17:11 LT, a total of 1173 strokes had been detected by the LINET in the system A's area. The Egrett-measured winds at 200 hPa were from the east-northeast at 10.5 ms^{-1}. Clearly, at that distance, there would not have been sufficient time for in-situ produced LNO_x from system A to have reached the Egrett's flight path. Thus the measured values on this transect should not have been influenced by LNO_x from either system B to the west or system A to the east, and can be considered as background concentrations.

The out-of-cloud transect 5 (see Fig. 4 and Table 1) occurred during an inbound leg to Darwin, before turning around to further probe the anvil outflow region of system B's storm. During this leg, the Egrett was level at 12 km altitude, north of the storm and 15 km west of Darwin. As with the previous out-of-cloud transect, the MTSAT image confirms the absence of any discernable cloud. During transects 4, 6 and 7, the sampled NO_x is negatively correlated with CO. This can be explained by fast vertical transport of low-CO, clean planetary boundary layer air by the storm's convection (see Fig. 1a).

6 Discussion

Between 11:50 LT on 22 January and 07:50 LT on 23 January, the LINET network recorded a total of 49 899 lightning strokes, most of them due to the lightning activity in system A. Unlike the pre-monsoon season, when the lightning activity is concentrated in localized convective cells, lightning occurred over a much wider area and in a more organized and widespread convective environment. Given that the storm sampled produced only 8 strokes (most probably all part of the same lightning flash), it is unlikely that the NO_x concentrations sampled in-cloud were caused by that storm. Furthermore, the mean NO_x concentration of the out-of-cloud transects is 1100 pptv, whereas that of the in-cloud transect is 984 pptv. It may be argued that the first out-of-cloud transect might be contaminated with NO_x from anthropogenic sources as this transect occurred just 10 km north of Darwin and on ascent. However, even without this first transect, the mean of the two remaining out-of-cloud transects is 723 pptv, which is only 27% lower than the in-cloud concentrations. Assuming that the out-of-cloud transects represent concentrations characteristic of cloud-free airmasses, and that the in-cloud concentrations measured are not necessarily the result of in-situ production (since there was only 8 strokes associated with that storm) but of vertical transport and entrainment of NO_x-rich air, one can pose the question of where the measured NO_x comes from.

To answer this question, 5-day, European Centre for Medium-Range Weather Forecast (ECMWF)-calculated back-trajectories were used to trace the origin of the airmasses sampled on the different transects both in-cloud and out-of-cloud, where NO_x was measured on this flight. Heyes et al. (2009) used a similar approach to study the origin of airmasses composing the TTL over Darwin during ACTIVE. Figure 6a and b show European Centre for Medium-Range Weather Forecast (ECMWF)-calculated back-trajectories for two transects of the Egrett's flight on 22 January. Clusters of five back-trajectories were calculated for each airmass sampled, but only single trajectories are shown for clarity. Figure 6a shows the back-trajectory from the airmass sampled on transect 4. The airmass originated east of Darwin, on the Cape York Peninsula on the eastern edge of the Gulf of Carpentaria, and went on to traverse the gulf and the entire northern part of the Northern Territories on its way to the point of sampling. In the second case (Fig. 6b), the airmass on transect 8 originated less than 270 km south of the sampling point, close to the coast, and followed a curved path along the western coast of the Cape York peninsula to then turn west until reaching the sampling point. The two back-trajectories shown are characteristic of all the transects where NO_x was sampled: the airmasses sampled in transects 1–5, at between 5000 and 12 600 m altitude followed easterly

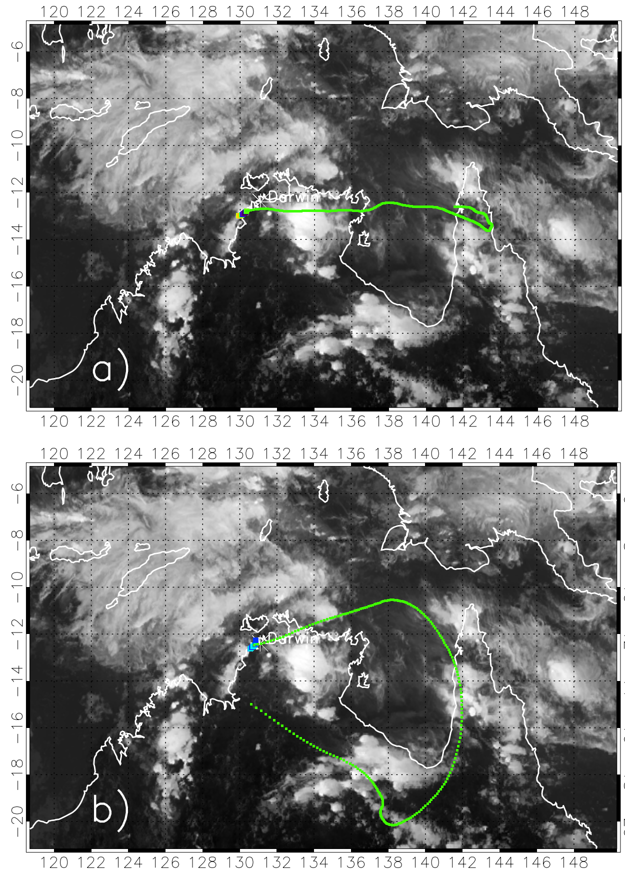

Fig. 6. 5-day backward trajectories from transects 4 **(a)** and 8 **(b)** overlaid on 07:30 UTC MTSAT IR satellite images of 22 January.

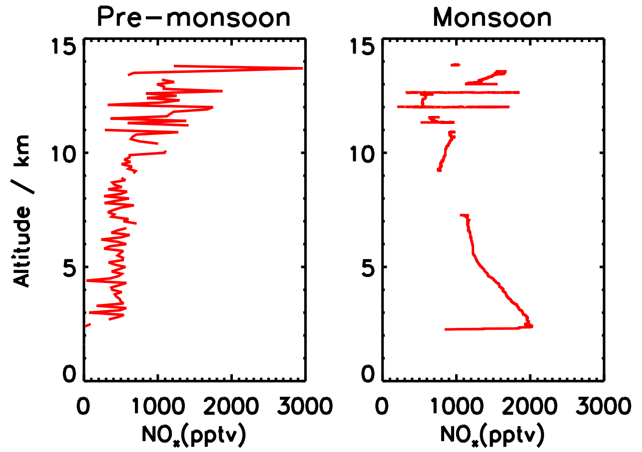

Fig. 7. Average NO_x vertical profile of two Egrett flights during the pre-monsoon period (left) and NO_x vertical profile for the flight on 22 January.

trajectories from the Gulf of Carpentaria whereas those in transects 6–8, sampled between 12 600 and 13 800 m, followed curved paths starting south of the sampling point. While backward trajectories in a highly convective environment are not an accurate representation of an airmass history, they do provide a broad indication of the airmasse's origin. In this case there seems to be consistency between groups of trajectories at different altitudes.

Figure 8a–e shows the location that the back-trajectory from transect 6 has reached for successive 24-h intervals, superimposed on the infra-red satellite image for that time. The figure also provides a good qualitative idea of the co-location of the convection and the stroke activity. In its journey, the airmass in question travelled over land that was convectively-active during the entire 5-day period, particularly over the northern end of the Northern Territory, where the airmass originated and where it was sampled five days later. This area coincides with the highest stroke activity as recorded by the LINET - partly to be expected, given that the detection efficiency is best within 300 km of the LINET stations, (Höller et al 2009), set up around Darwin. The LINET plots suggest that the airmass was exposed to the most intense lightning activity on 17 and 21 January (panels "e" and "a", respectively, in Fig. 8), not far from the place where it was sampled. The airmasses in transects 6–9 followed very similar trajectories, whereas those in transects 1–5 originated around the northern end of the Cape York peninsula and would have thus been exposed to the strongest lightning activity as they approached the sampling area, on 21 and 22 January.

The abundant convection may then have acted to transport any LNO_x produced to the upper troposphere, where the lifetime can increase to the order of days (Jaeglé et al., 1998).

In order to rule out possible biomass burning interference in the NO_x signal, data from the European Space Agency's Along-Track Scanning Radiometer (ATSR) Wild fire Atlas (WFA) (http://dup.esrin.esa.int/ionia/wfa) was used to locate wildfires that may have affected the airmasses sampled. A box was drawn around the area covered by the back-trajectories, along 130°–140° W, 10°–20° S and measuring 1530 by 1100 km and the occurrence of fires between 1 January and the day of the flight within that box was investigated. As per the level 3 ATSR WFA product, fires occurred in two locations within the box; on 4 January, a set of 6 episodes centred around 11° S, 145° W and on 7 January 5 episodes centred around 15° 40′ S, 144° W. Although these fires occurred in an area with the potential to affect the airmasses sampled, the fires occurred too early in the month to have any significant impact on the measurements on the day of the flight, if one assumes a one-week lifetime for NO_x in the upper troposphere. Furthermore, the CO values associated with the NO_x measured in the transects were in the 45–75 ppb range; in contrast, the CO values measured on the 16 November flight, where the troposphere was still influenced by a biomass burning signal, were in the 80-100 ppb range. We can thus conclude that no clear biomass-burning signal is present in the measurements made during the flight.

Table 2. Mean NO_x enhancements for selected field campaigns in the tropics.

Year	Project, region	Mean NO_x, pptv	Ref.
Jul 1985	GTE/ABLE 2A, Amazonia, Brazil	60	Torres et al. (1998)
Sep 1992	GTE/TRACE A, A. Cerrado, Brazil	300–900	Pickering et al. (1996)
Mar 2000	INCA, west coast S. America	40–800	Baehr et al. (2003)
Jul 2002	CRYSTAL FACE, Florida	1000–4000	Ridley et al. (2006)
Jan–Mar 2004	TROCCINOX, State of Sao Paulo, Brazil	500–1500	Huntrieser et al. (2007)
Jan 2006	ACTIVE, Northern Australia	500–1500	This work

Table 3. Stroke count for selected days of the ACTIVE campaign.

Day	Regime	Type of storm	Strokes, total	Strokes, CG	Strokes, IC
14 Nov 2005	Pre-monsoon	Sing. Cellular	89 562	16 609	35 810
16 Nov 2005	Pre-monsoon	Sing. Cellular	80 218	5639	27 701
17 Nov 2005	Pre-monsoon	Multi Cellular	19 1576	11 276	55 216
7 Dez 2005	Pre-monsoon	Multi Cellular	77 455	8014	23 339
20 Jan 2006	Monsoon	Monsoon	12 398	2736	2008
21 Jan 2006	Monsoon	Monsoon	10 836	397	130
22 Jan 2006	Monsoon	Monsoon	42 491	3266	15 288
9 Feb 2006	Monsoon break	Multi Cellular	223 080	10 061	79 370
10 Feb 2006	Monsoon break	Multi Cellular	188 309	19 436	59 784

The LINET stroke count over the previous 5 days shows an average of 20 380 strokes day^{-1}, with a pattern (not shown) not unlike that of 22 January, where the bulk of the convective (and lightning) activity started to the east of Darwin and moved west towards the coast south of Darwin, with storms still active after moving out to sea in the early hours of the morning. The area south of Darwin, where the airmasses sampled in transects 6–8 appear to originate, sustained the heaviest lightning activity; transect 7 (see Table 1), whose airmass appears to originate in that area, showed the highest mean NO_x concentration of the entire flight. Thus a pattern seems to emerge which, despite the relatively low stroke count, typical of monsoon storms, coupled with fast vertical transport and regional airmass transport patterns, results in significant accumulation of lightning-produced NO_x and high mixing ratios even in cloud-free air.

A comparison between the NO_x sampled by the Egrett during pre-monsoon flights and the flight discussed in this study shows an interesting contrast. Two NO_x flights during ACTIVE's first phase, on 16 November and 3 December 2005, respectively, successfully sampled Hector convection during the pre-monsoon season. Figure 7a and b show the average vertical profiles of the two NO_x pre-monsoon flights and the vertical profile of the flight discussed in this study, respectively. The first thing to notice is the significantly flatter profile in the free troposphere during the first phase's flights. Free tropospheric, out-of-cloud NO_x mixing ratios during the pre-monsoon phase ranged between 400 and 500 ppt up to 13 km (16 November) and 10 km (3 Decemebr). Upon probing the anvil outflow region of Hector storms, however, in-cloud NO_x mixing ratios of up to 4000 pptv were sampled during anvil penetrations. In contrast, the flight on 18 January exhibited a profile typical of a well mixed upper troposphere, where the maximum in-cloud NO_x mixing ratios reached 2000 ppt with out-of-cloud NO_x values only 27% lower than those measured in in-cloud conditions. This results in part from the more organized and widespread nature of monsoon-type convection, which may act to redistribute LNO_x vertically and over a much wider area much more efficiently than the episodic and highly localized pre-monsoon convection. The large values in the free troposphere at 2 km on the flight on 22 January (Fig. 7b) are from transect 1, on which the first large out-of-cloud values were registered.

Table 2 shows the mean NO_x enhancements measured in other field campaigns in the tropics, including ACTIVE. The monsoon NO_x enhancements measured during this flight show a range comparable to that measured during the continental-convection TROCCINOX field campaign and larger than that of any of the other listed continental tropical field campaigns except CRYSTAL-FACE, where deep convective, sea-breeze driven thunderstorms (similar to those sampled during the first part of the ACTIVE campaign and described in another manuscript currently in preparation) were sampled. It is important to notice, however, that the measurements made in the campaigns listed were made at point sources i.e., in the anvil outflow regions of

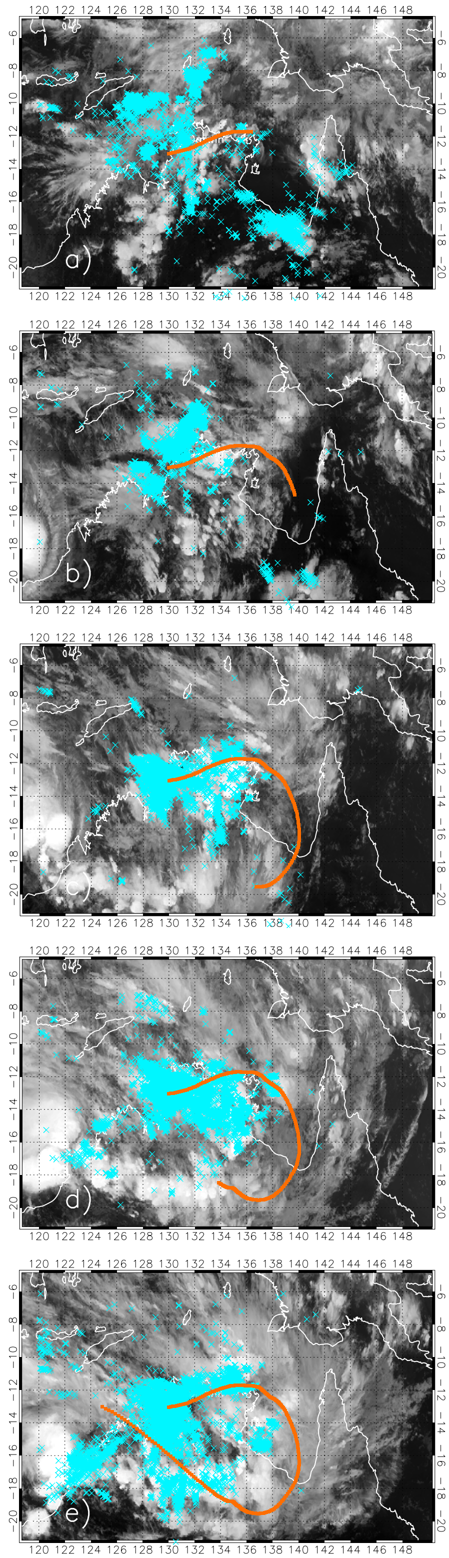

Fig. 8. Location that the back-trajectory from transect 6 has reached for successive 24-h intervals, starting on 22 January **(a)** through to 17 January **(e)**, with LINET strokes for the corresponding day overplotted on the 08:30 UTC satellite image for that day in each panel.

electrically-active storms. The process described here is most likely one of accumulation of NO_x through transport and convection.

The measurements presented here were made during active monsoon conditions and right at the onset of a large MCS with more abundant convection and whose lightning activity during the next 3 days, as recorded by the LINET, was at least a factor of 5 or larger than that on the 5 previous days. Unfortunately, no airborne NO_x measurements were made in the post-MCS environment, but it is reasonable to assume that LNO_x production should have been considerably larger under those conditions. While the MCS looks, at least from the lightning activity standpoint, as an outlier event and might thus be thought of as unrepresentative of the Northern Australian monsoon regime, MCSs are not an uncommon feature of that season. Between 4 and 5 such events can occur during each monsoon, although most will feature maritime-type convection and could thus be electrically weaker than the one encountered here (P. May, personal communication, 2008).

Mainly because they were recorded during one single flight, it is difficult to categorize the out-of-cloud NO_x concentrations as typical monsoon background values, yet they do provide an indication not only of the LNO_x production during previous days, but of the accumulation of NO_x in the background due to its longer lifetime in the upper troposphere, as well as of convective vertical transport and entrainment of this NO_x-rich air by active storms.

7 Conclusions

This study shows the result of one flight where airborne NO_x measurements were made during the 2006 Northern Australian monsoon, in the framework of the ACTIVE campaign. The average NO_x mixing ratios recorded were on a par with those measured during tropical continental field campaigns that targeted LNO_x. While the NO_x concentrations might equal those measured at point sources during those campaigns, the mechanism at work here is different; despite a markedly lower lightning activity compared to that during the previous (pre-monsoon) and succeeding (monsoon-break) periods, a combination of widespread, organized and electrically-active convection, combined with regional transport patterns and fast vertical transport by deep convection appears to have resulted in accumulation of lightning-produced NO_x in the upper-troposphere and tropical tropopause layer. The combination of these factors may have compensated for the lower lightning activity during this period to result in the high NO_x mixing ratios measured in the upper troposphere. During the monsoon season in Northern Australia, when biomass burning has abated, there is no other significant source of NO_x: an instrument flown on the low-level Dornier aircraft with detection limits of 230 and 450 pptv for NO and NO_2 respectively did not register any non-zero values during the monsoon phase, except at the airport. Hydrocarbon measurements on the Dornier showed median concentrations of a few to a few tens of pptv for a range of VOCs (Allen et al., 2008), so there is potential for ozone production in the TTL.

Despite the maritime nature of the convection in this regime, its organized, widespread nature, coupled with a constant lightning activity demonstrate that during this kind of regime the potential for lightning NO_x production is as large as that of continental convection regimes, with the attendant potential effects on ozone and OH radical production. Further study is warranted to determine whether the enhancements presented here are representative of monsoon regimes elsewhere.

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