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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 mixing ratios (mrs) of 723 and 984 parts per trillion volume (pptv) 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 accretion 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.

1 Introduction

Nitrogen oxides or NO_x ($\text{NO}_x = \text{NO} + \text{NO}_2$), 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 (LtNO_x hereafter) play an important role in the chemistry of the troposphere,

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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 in 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, LtNO_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. As a consequence, current LtNO_x production estimates range 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 LtNO_x makes it difficult, in turn, to precisely estimate the global ozone budget.

Measurements of LtNO_x are difficult, given the violent nature of the phenomena involved in its production, and require dedicated experiments and airborne platforms to sample, usually flying in the outflow region of thunderstorms. Most lightning LtNO_x sampling experiments have been carried out in continental, isolated type convection (Ridley et al., 2004; Huntrieser et al., 2007, 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) (Russel et al., 1993) and established that lightning is most likely a source of upper tropospheric NO_y at Darwin. Although measurements of LtNO_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 in during the ACTIVE campaign, in January 2006, during the Northern Australian monsoon.

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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 of 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 Sect. 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, encountered deep, isolated pre-monsoon convection, particularly over the Tiwi islands, 50 km north of Darwin. The second phase, in January–February 2006, experienced a more organised and widespread monsoon convection 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 (TLL) 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 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-NO₂ sensor (Volz-Thomas et al., 2005). 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 experimental area that permitted 3-D spatial as well as temporal location of lightning strokes.

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2.1 NO_x instrument

The NO-NO₂ instrument deployed onboard the Egrett was a modified version of the MOZAIC NO_y 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, BLC) for the specific conversion of NO₂ 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% at ground and 50% at 150 hPa. The instrument was regularly calibrated before and after flight for both, sensitivity for NO and conversion efficiency of NO₂.

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.

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).

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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 return 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 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 product 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

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altitude. This contrasts 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 (May, personal communications). 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 mrs of less than 50 ppbv. This period lasted until the dissipation of the low pressure system on 2 February.

Three days of clear skies followed before conditions similar to those of the pre-

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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 8 mm day (significantly less than the $\sim 17 \text{ mm day}^{-1}$ 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 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 recoded 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. 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, however,

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resulted in more frequent storms covering a wider area than the oftentimes localized pre-monsoon and monsoon-break storms.

4 Conditions on the day of the flight

On 22 January deep convection started to organize at approximately 13:30 local time some 400 km south-east of Darwin, moving west. By 17:00 LT, convection had developed into a squall line and moved to about 200 km east south-east of Darwin and would eventually reach the city at about 21:00 LT. These were the first manifestations of the mesoscale convective system described above. Along the coast SW of Darwin, a storm system, 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. This system preceded the MCS by at least 4 h. By 16:23 LT, the system had fully developed and started to move out to sea.

The LINET network registered the first 3 lightning strokes of this system 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 LT LINET recorded 27 strokes with the last 16 strokes being recorded between 18:30 and 19:30 LT, mostly over land, along the system’s trailing edge. By 19:23 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 LT to sample the anvil outflow of the northernmost storm of the line of storms along the coast SW of Darwin. Initially the aircraft headed due south of Darwin before heading west toward the coast and the

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storm. 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.

Figure 3 shows the Egrett's flight path on 22 January and the lightning activity detected by the LINET in the period from 00:00–19:36 LT, the time the flight ended. The aircraft first flew due south of Darwin to approximately 13° S in what mostly cloud-free air, and then headed west towards the coast to sample the convection there. Once over the coast, a series of SW-NE-SW transects were flown where the aircraft probed the anvil outflow region of a 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 nascent MCS's region (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 constitutes the first manifestation of the approaching MCS. Despite the apparent proximity of the strokes to the Egrett's flight track, at that 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 switched 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 5 times, which resulted in a series of large NO_x concentrations or “spikes”, with average mixing ratios of 901, 883, 741, 1570 and 870 pptv for each transect, respectively and a total 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 total average of 723 pptv. The first two out-of-cloud transects occurred south of Darwin, the first during

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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 total average mixing ratio.

5 Of particular interest is transect 2 (see Fig. 5); the Egrett was at 10 km altitude, 50 km due south of Darwin and 19 km east of the storm it would later sample. Likewise, the aircraft was approximately 300 km west of the nascent MCS's centre of lightning activity. Between the start of the lightning activity period of the MCS, at 14:05 LT and the end of this NO_x transect at 17:11 LT, a total of 1173 strokes had been detected
10 by the LINET in the MCS's area. The 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 LtNO_x from the MCS to have reached the Egrett's flight path. Thus the measured values should not have been influenced by LtNO_x from either the storm line to the west or by the MCS to the east, and can be considered as background
15 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. 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
20 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
25 recorded a total of 49 899 lightning strokes, most of them due to the MCS's activity. 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

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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 980 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 720 pptv, which is only 27% lower than the in-cloud concentrations. Assuming that the out-of-cloud transect 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 shows European Centre for Medium-Range Weather Forecast (ECMWF)-calculated back-trajectories for two transects of the Egrett's flight on 22 January. 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: airmasses sampled in transects 1–5, between 5000 and 12 600 m altitude followed easterly trajectories from the Gulf

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of Carpentaria whereas those in transects 6–8, sampled above 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 air mass's history, they do provide a broad indication of the air mass's origin. In this case there seems to be consistency between groups of trajectories at different altitudes.

The figures indicate that the sampled air masses all travelled mostly over land in the active monsoon region during the 5 days prior to the sampling. During that time, they transited over a highly-unstable and convective area with electrical activity every single day; thus it appears that the air masses accrued lightning-produced NO_x during those five days. The abundant convection transported the newly formed species to the upper troposphere, where the lifetime can increase to the order of days (Jeaglé et al., 1998).

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 air masses sampled in transects 6–8 appear to originate, sustained the heaviest lightning activity; transect 7 (see Table 1), whose air mass 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 air mass transport patterns, results in significant accretion of lightning-produced NO_x and high mixing ratios even in cloud-free air.

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.

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

5 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
10 that LtNO_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 (May, personal communications). 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 LtNO_x production during previous days, but of the accumulation of NO_x in the background due to its longer
15 lifetime in the upper troposphere, as well as of convective vertical transport and entrainment of this NO_x -rich air by active storms.
20

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
25 during tropical continental field campaigns that targeted LtNO_x . While the NO_x concentrations might equal those measured at point sources during those campaigns, the

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mechanism at work 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 accretion 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.

Acknowledgements. We thank the pilots and ground staff of the Egrett and Dornier aircraft for ensuring a successful campaign, and thank Peter May for particularly useful discussions and insight into the general meteorological condition over Northern Australia during ACTIVE, as well as the staff of the Bureau of Meteorology (BoM) Regional Centre in Darwin for their support in forecasting and logistics; also the Royal Australian Air Force for hosting the aircraft experiment, and Marcel Berg and Norbert Houben for maintenance of the CO and NO_x instruments. James Lee provided the Dornier NO_x estimates. Finally, we thank the UK Natural Environment Research Council (Grant NE/C512688/1) for supporting ACTIVE.

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- Baehr, J., Schlager H., Ziereis, H., Stock, P., van Velthoven, P., Busen, R., Ström, J., and Schumann, U.: Aircraft observations of NO, NO_y, CO and O₃ in the upper troposphere from 60° N to 60° S – Interhemispheric differences at midlatitudes, *Geophys. Res. Lett.*, 30, 52, doi:10.1029/2003gl1016935, 2003.
- Baumgardner, D., Jonsson, H., Dawson, W., Connor, D., and Newton, R.: The cloud aerosol and precipitation spectrometer (CAPS); a new instrument for cloud investigations, *Atmos. Res.*, 59–60, 251–264, 2002.
- Betz, H.-D., Schmidt, K., Oettinger, W. P., and Wirz, M.: Lightning Detection with 3-D-Discrimination of Intracloud and Cloud-to-Ground Discharges, *J. Geophys. Res. Lett.*, 31, L11108, doi:10.1029/2004GL019821, 2004.
- Bond, D. W., Steiger, S., Zhang, R., Tie, X., and Orville, R. E.: The importance of NO_x production by lightning in the tropics, *Atmos. Environ.*, 36, 1509–1519, 2002.
- Crutzen, P. J.: The role of NO and NO₂ in the chemistry of the stratosphere and troposphere, *Ann. Rev. Earth Planet. Sci.*, 7, 443–472, 1979.
- Chameides, W. L. and Walker, J. C. G.: A photochemical theory of ozone, *J. Geophys. Res.*, 78, 8751–8760, 1973.
- Gerbing, C., Schmitgen S., Kley, D., and Volz-Thomas, A.: An improved fast-response vacuum-UV resonance fluorescence CO instrument, *J. Geophys. Res.*, 104(D1) 1699–1704, 1999.
- Heyes, W. J., Vaughan, G., Allen, G., Volz-Thomas, A., Ptz, H.-W., and Busen, R.: Composition of the TTL over Darwin: local mixing or long-range transport?, *Atmos. Chem. Phys. Discuss.*, 9, 7299–7332, 2009, <http://www.atmos-chem-phys-discuss.net/9/7299/2009/>.
- Huntrieser, H., Feigl, C., Schlager, H., Schroeder F., Gerbig, C., van Velthoven, P., Flato, F., Thery, C., Petzold, A., Holler, H., and Schumann, U.: Airborne measurements of NO_x, trace species and small particles during the European lightning nitrogen oxides experiment, *J. Geophys. Res.*, 107, 4113, doi:10.1029/2000JD000209, 2002.
- Huntrieser, H., Schlager, H., Roiger, A., Lichtenstern, M., Schumann, U., Kurz, C., Brunner, D., Schwierz, C., Richter, A., and Stohl, A.: Lightning-produced NO_x over Brazil during TROCCINOX: airborne measurements in tropical and subtropical thunderstorms and the importance of mesoscale convective systems, *Atmos. Chem. Phys.*, 7, 2987–3013, 2007, <http://www.atmos-chem-phys.net/7/2987/2007/>.

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9, 10647–10673, 2009

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- Jaeglé, L., Jacob, D. J., Wang, Y., Weinheimer, A. J., Ridley, B. A., Campos, T. L., Sachse, G. W., and Hagen, D. E.: Sources and chemistry of NO_x in the upper troposphere over the United States, *Geophys. Res. Lett.*, 25(10), 1705–1708, 1998
- Keenan, T. D. and Carbone, R. E.: Propagation and diurnal evolution of warm season cloudiness in the Australian and maritime continent region, *Mon. Weather Rev.*, 136, 973–994, 2008.
- Kondo, Y., Koike, M., Kita, K., Ikeda, H., Tagekawa, N., Kawakami, S., Blake, D., Liu, S. C., Ko, M., Myazaky, Y., Irie, H., Higashi, Y., Liley, B., Nishi, N., Zhao, Y., and Ogawa, T.: Effects of biomass burning, lightning, and convection on O₃, CO, and NO_y over the tropical Pacific and Australia in August–October 1998 and 1999, *J. Geophys. Res.*, 108(D3), 8402, doi:10.1029/2001JD000820, 2003.
- Labrador, L. J., von Kuhlmann, R., and Lawrence, M. G.: Strong sensitivity of the global OH mean concentration and the tropospheric oxidizing efficiency to the source of NO_x from lightning, *Geophys. Res. Lett.*, 31, L06102, doi:10.1029/2003GL019229, 2004.
- May, P. T., Mather, J. H., Vaughan, G., Jakob, C., McFarquhar, G. M., Bower, K. N., and Mace, G. G.: The Tropical Warm Pool International Experiment (TWIPCE), *B. Am. Meteor. Soc.*, 89, 632–645, 2008.
- Murphy, D. M., Fahey, D. W., Proffitt, M. H., Liu, S. C., Chan, K. R., Eubank, C. S., Kawa, S. R., and Kelly, K. K.: Reactive nitrogen and its correlation with ozone in the lower stratosphere and upper stratosphere, *J. Geophys. Res.*, 98(D5), 8751–8773, 1993.
- Pickering, K. E., Thomson, A. M., Wang, Y., Tao, W.-K., McNamara, D. P., Kirchhoff, V. W. J. H., Heikes, B. G., Sachse, G. W., Bradshaw, J. D., Gregory, G. L., and Blake, D. R.: Convective transport of biomass burning emissions over Brazil during TRACE A, *J. Geophys. Res.*, 101, 23993–24102, doi:10.1029/96JD00346, 1996.
- Ridley, B., Ott, L., Pickering, K., Emmons, L., Monztkka, D., Wenhiemer, A., Knapp, D., Li, L., Heymsfield, G., McGill, M., Kucera, P., Mahoney, M. J., Baumgardner, D., Schultz, M., and Brasseur, G.: Florida thunderstorms: A faucet of reactive nitrogen to the upper troposphere, *J. Geophys. Res.*, 109, D17305, doi:10.1029/2004JD004769, 2004.
- Russell, P. B., Pfister, L., and Selkirk, H. B.: The tropical experiment of the Stratosphere-Troposphere Exchange project (STEP): science objectives, operations, and summary findings, *J. Geophys. Res.*, 98(D5), 8563–8589, 1993.
- Schumann, U. and Huntrieser, H.: The global lightning-induced nitrogen oxides source, *Atmos. Chem. Phys.*, 7, 3823–3907, 2007,

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<http://www.atmos-chem-phys.net/7/3823/2007/>.

- Stockwell, D. Z., Giannakopoulos, C., Plantevin, P.-H., Carver, G. D., Chipperfield, M. P., Law, K. S., Pyle, J. A., Shallcross, D. E., and Wang, K.-Y.: Modelling NO_x from lightning and its impact on global chemical fields, *Atmos. Environ.*, 33, 4477–4493, 1999.
- 5 Tie, X., Zhang, R., Brasseur, G., Emmons, L., and Wengfan, L.: Effects of lightning on reactive nitrogen and nitrogen reservoir species in the troposphere, *J. Geophys. Res.*, 16(D3), 3167–3718, 2002
- Torres, A. L., Buchan, H., and Tuck, A. F.: Tropospheric nitric oxide measurements over the Amazon basin, *J. Geophys. Res.*, 93, 1396–1406, 1988.
- 10 Vaughan, G., Schiller, C., MacKenzie, A. R., Bower, K., Peter, T., Schläger, H., Harris, N. R. P., and May, P. T.: SCOUT-03/ACTIVE High-altitude aircraft measurements around deep tropical convection, *B. Am. Meteor. Soc.*, 89, 647–662, 2008.
- Volz-Thomas, A., Berg, M., Heil, T., Houben, N., Lerner, A., Petrick, W., Raak, D., and Pätz, H.-W.: Measurements of total odd nitrogen (NO_y) aboard MOZAIC in-service aircraft: instrument
- 15 design, operation and performance, *Atmos. Chem. Phys.*, 5, 583–595, 2005, <http://www.atmos-chem-phys.net/5/583/2005/>.

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

Transect no.	Start (LT)	Dur. (s)	Altitude (mts)	Avg. NO _x (pptv)	Peak NO _x (pptv)	Avg. 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

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

Year	Project, region	Mean NO _x , pptv	Ref.
July 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)
March 2000	INCA, west coast S. America	40–800	Baehr et al. (2003)
July 2002	CRYSTAL FACE, Florida	1000–4000	Ridley et al. (2006)
Jan–March 2004	TROCCINOX, State of Sao Paulo, Brazil	500–1500	Huntrieser et al. (2007)
Jan 2006	ACTIVE, Northern Australia	500–1500	This work

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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	191 576	11 276	55 216
7 Dec 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

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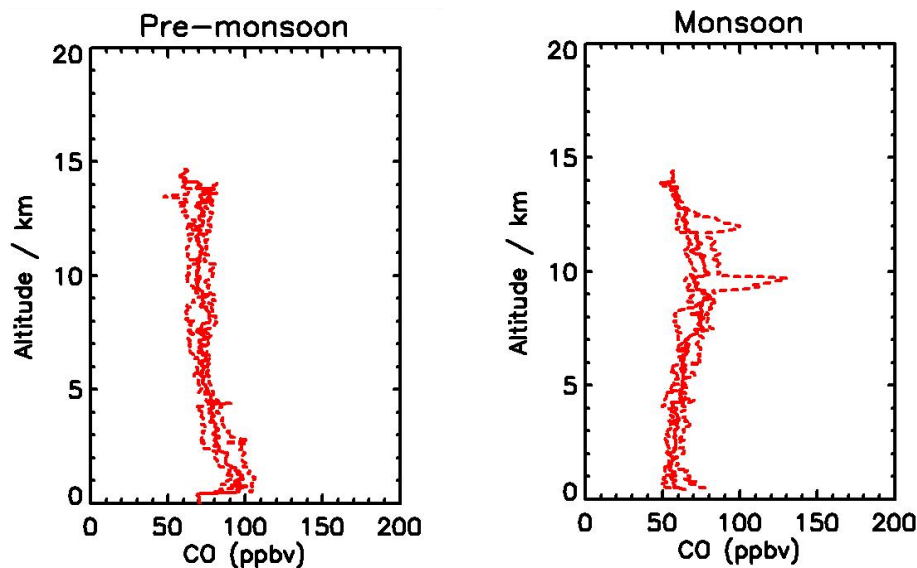


Fig. 1. Median CO vertical profile measured by the Egrett during **(a)** the pre-monsoon (left) and **(b)** monsoon (right) seasons. The dashed lines are the lower and upper quartiles of the measurements.

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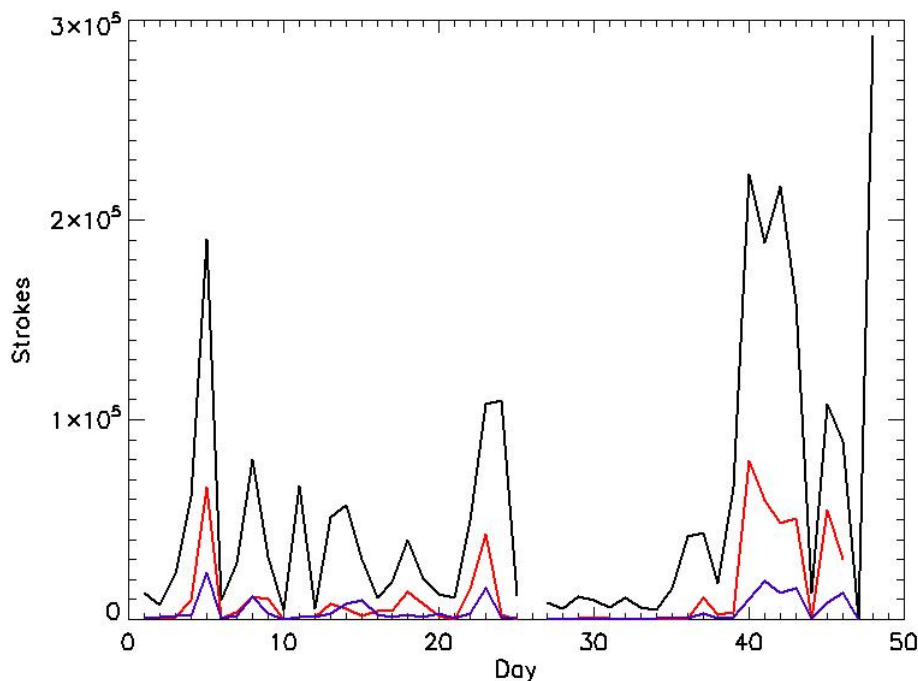


Fig. 2. LINET stroke count time series for the 1 January–12 February 2006, corresponding to the active monsoon in Northern Australia. Total strokes (black line), in-cloud strokes (red line) and cloud-ground strokes (blue line) are depicted. The rise in the count on 21 January corresponds to the onset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

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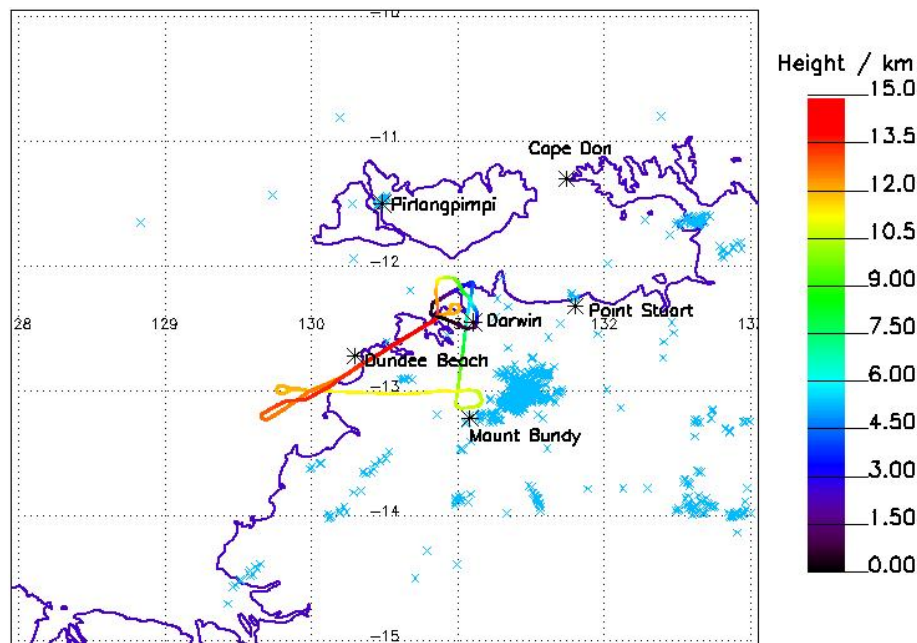


Fig. 3. Egrett's flight path on the 22 January flight, colour-coded to altitude. The black asterisks mark the position of the six LINET stations deployed during ACTIVE and the blue crosses are the LINET-detected strokes for the period 00:00–19:36 LT (i.e., from the start of the day to the end of the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

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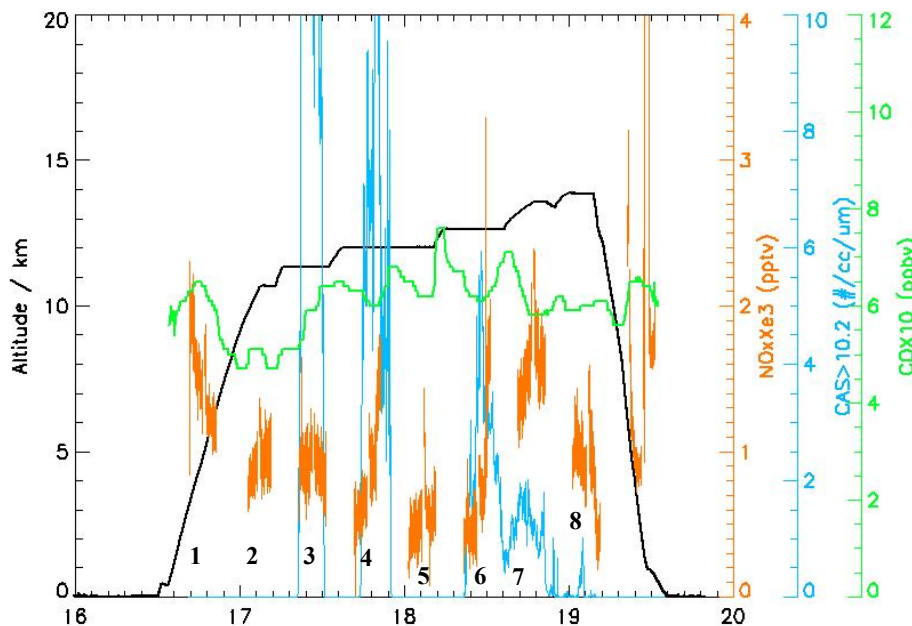


Fig. 4. Time series (in hours local time) of Egrett data on the 22 January flight: altitude (black line) CO (green), NO_x (orange) and particles larger than 10.2 μm (light blue line), used as an indicator of in-cloud conditions, where $N > 1$ indicates cloud. The numbers correspond to the different transects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

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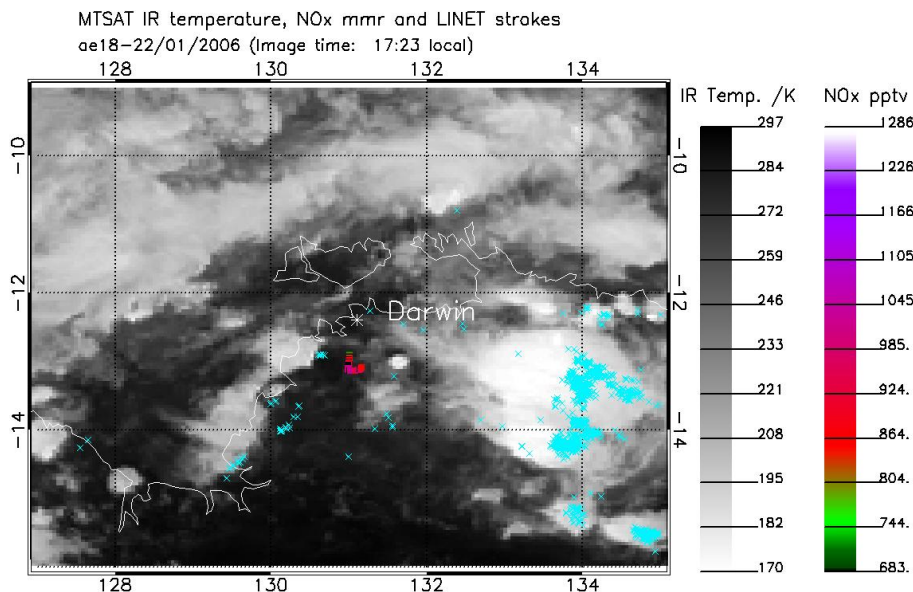


Fig. 5. Egrett's second cloud-free transect (coloured trace) colour-coded to NO_x and LINET strokes (blue crosses) between 14:05 and 17:11 LT (the sampled storm's electrically active period) on 22 January 2006. The transect occurred between 17:03 and 17:13 LT.

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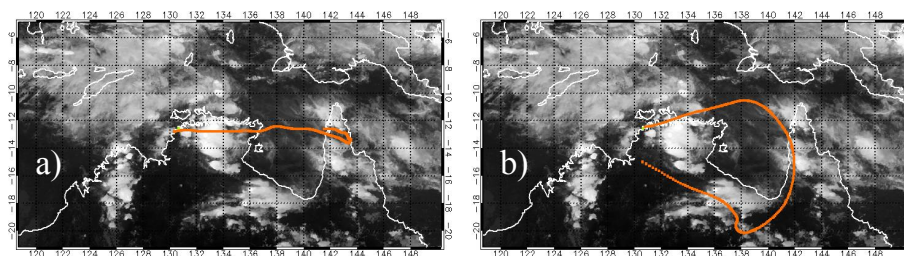


Fig. 6. 5-day backward trajectories from transects 4 (a) and 8 (b) overlaid on MTSAT IR satellite images of 22 January.

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