NITROGEN OXIDES FROM THUNDERSTORMS - RESULTS FROM EXPERIMENTS OVER EUROPE AND THE CONTINENTAL TROPICS

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1 INTRODUCTION

Deep convection causes thunderstorms with lightning. Within the hot lightning flash channels nitrogen and oxygen air molecules get dissociated and the products react to nitrogen oxides (NO_x = sum of NO and NO_2) at considerable amounts. Nitrogen oxides play a key role in the ozone (O_3) and hydroxyl radical (HO_x) budget (*Liu*, 1977). There are still large uncertainties in the knowledge of the magnitude of the different sources of reactive nitrogen (lightning, fossil fuel combustion, biomass burning, microbial activity, aircraft, transport from the stratosphere) for the troposphere. Most estimates of lightning-produced NO_x (LNOx, in mass units of nitrogen) are in the range 2-20 Tg(N) yr^{-1} (*Huntrieser et al.*, 2002). These values are considerably higher than NO_x emitted from aircraft (<1 Tg(N) yr^{-1}). The uncertainty in the LNOx source has large impact also on the best estimate for the life-time of methane in the atmosphere and hence on estimates of the greenhouse effect (*Labrador et al.*, 2004).

From the EULINOX experiment in 1998, and its predecessor LINOX of 1996 (*Höller et al.*, 1999), we concluded (*Huntrieser et al.*, 2002) that lightning production over Europe amounts to about ~0.03 Tg(N) yr^{-1}, which is less than the emissions from aircraft in the same region. The European findings were supported by more recent measurements during the experiment CONTRACE (*Huntrieser et al.*, 2004). Scaling the European measurements to the global scale implies a mean source of 3 Tg(N) yr^{-1}, far larger than the global contribution from aviation (0.6 to 1 Tg(N) yr^{-1}), but with an uncertainty of about one order of magnitude. As can be seen from satellite observations (OTD and LIS), lightning activity is highest over tropical continental areas. Hence, any reliable assessment on the global lightning NO_x source needs measurements in the continental tropics.

Previous projects which provided in-situ measurements of NO_x in the upper troposphere are rare in the continental tropics: STRATOZ III (Stratospheric Ozone Experiment) in June 1984 *(Drummond et al., 1988)*, TROPOZ II (Tropospheric Ozone Experiment) in Jan./Febr. 1991 *(Rohrer et al., 1997)*, and TRACE-A (The Transport and Chemistry near the Equator Experiment over the Atlantic) in Sept./Oct. 1992 *(Fishman et al., 1996; Pickering et al., 1996; Wang et al., 1996)*. For example TRACE-A results showed pollution from biomass burning in NE Brazil (5-15°S, 45-55°W) which was transported upward by deep convection (200-300 nmol mol^{-1} CO vs. 90 nmol mol^{-1} background, O_3 lower than background); and enhanced concentrations of O_3 precursors (carbon monoxide, hydrocarbons, and nitrogen oxides) were observed at cloud outflow levels (9-12 km). The concentration of NO_x in cloud outflow was about 1.3 nmol mol^{-1} compared to background of 0.2 nmol mol^{-1}. From observations of the NO_x/CO ratio it was concluded that the lightning-NO_x contribution in the outflow was of order 30-40%.

Airborne measurements have been performed by our team in the tropics over the continent of South America in March/April 2000 (EC-project "Interhemispheric Differences in Cirrus Properties from Anthropogenic Emissions," INCA) and in February/March 2004 (EC-project "Tropical Convection, Cirrus and Nitrogen Oxides Experiment," TROCCINOX).

During INCA, measurements of O_3, CO, NO and NO_y and aerosols have been performed onboard the DLR Falcon up to 12.5 km altitude in the tropics when passing along the west coast of South America from Panama over Lima to Santiago de Chile (March 2000) and over eastern Brazil from Foz do Iguacu to Recife (April 2000). The INCA measurements were interpreted using airmass back trajectories, GOES images, brightness temperature analysis, and OTD lightning flashes (*Baehr et al.*, 2003; *Schumann et al.*, 2004). The airmasses observed on the southward and northward flights passing the tropics over South

America during INCA were influenced by deep convection in the recent days before the measurements. Enhancements of NO and NO_y on the horizontal scale of up to 300 km have been observed which can be attributed to large convective systems with lightning. Enhanced CO mixing ratios of 120-180 nmol mol^{-1} in the upper troposphere (UT) occur in an extended region over tropical South America (30° latitude range), connected to the large-scale anticyclonic flow in the UT (Bolivian High). The enhanced CO in the UT originates mainly from the Amazon basin. Analysis of NO_y/CO correlations in the observed airmasses confirm that airmasses from the boundary layer in the Amazonian region were transported upwards into the mid troposphere by deep convection. The NO and NO_y concentrations decreases with the age after convection and amounts to typically 1 and 3 nmol mol^{-1}, respectively.

In the following we summarise the results obtained from the recent measurements over Brazil within the experiment TROCCINOX and compare the results with the results from EULINOX and its predecessors LINOX, and INCA.

2 The TROCCINOX Experiment

The TROCCINOX experiment in February and March 2004 was performed using the DLR research aircraft Falcon. Figure 1 shows the flight paths of three flights performed over South-East Brazil out of the airport Gaviao Peixoto (21.8°S 48.4°E) in the state of Sao Paulo. The flight path is projected onto Meteosat second generation (MSG) data in false colour. The aircraft was equipped with instruments to measure NO, NO_y, CO, O_3, and meteorological parameters. In addition the Falcon had instruments to measure aerosol and water vapour profiles above the aircraft with a Differential Absorption Lidar. The experiment included a total of 23 flights with 82 flight hours, of which 14 flights were performed measuring near deep convective events over South-East Brazil in the vicinity of the state of Sao Paulo. Three flights (14th, 28th February, and 3rd March) have been selected which are particularly suitable for further analysis concerning lightning-produced NO_x (see section 4). These flights were all local flights performed in a region with radar observations. Based on tropopause altitude and energy content of the airmasses, the first and third flights were performed in tropical airmasses, and the second flight in subtropical airmasses.

3 METHODS FOR ANALYSIS

The global lightning NO_x production rate, $P(NO_x)$ can be estimated in various ways. One way is to use estimates of the average volume mixing ratio of nitrogen oxides in the anvil of thunderstorms and to multiply these values with estimates of the air flux out of the thunderstorms (*Chameides et al.*, 1987),

$$P(NO_x) = [NO_x] F_C \, S \, C_1.$$

$[NO_x]$ is the average volume mixing ratio produced by lightning in the anvil, F_C is the average rate at which air is advected out of the anvil, S is the number of active cumulonimbus cells occurring at any instant globally (~2000), and C_1 is a conversion factor (1.5×10^7 g(N) $g(air)^{-1}$ s yr^{-1}). The flux F_C can be estimated from

$$F_C = (v_a - v_s)\, \rho_a \, \Delta x \, \Delta z.$$

Here v_a is the horizontal wind speed inside the anvil, v_s is the velocity of the storm system, ρ_a is the air density in the anvil, Δx is the width of the anvil, and Δz is the depth of the anvil.

It is obvious that this method suffers from important uncertainties, in particular in the definition and quantification of the number S and the flux F_C. It is difficult to provide accurate error estimates, and it appears that this method cannot significantly reduce the present uncertainty in our best estimate of the magnitude of LNOx. However, comparisons between various thunderstorm cases do not depend on the value of S.

Another way is to use chemical transport models which compute the concentration of NO and related trace gas components (including NO_x, NO_y, but also CO, O_3 etc.) for various assumed source distributions and source strengths and to identify the best estimate for the actual source strength by a least square fit between the observed and the modelled concentration results. Such comparisons between trace gas measurements from the Falcon aircraft and model simulations are under preparation and preliminary results will be presented at the Conference.

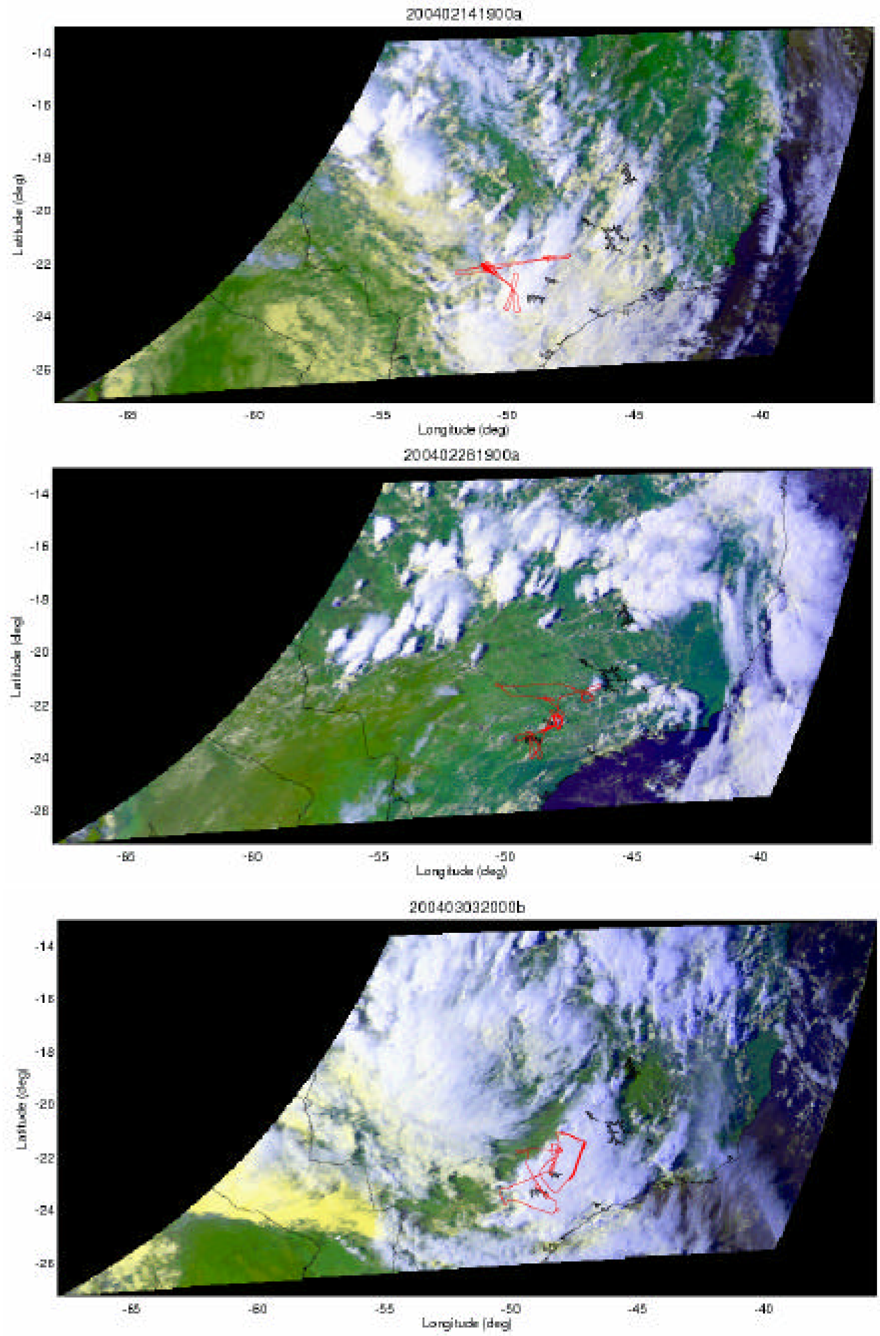

Figure 1. Flight paths of the Falcon (red curve) projected onto Meteosat second generation (MSG) red-blue-yellow false colour pictures from channels 1 (0.6 µm), 2 (0.8 µm) and 9 (10.8 µm) in an equi-latitude-longitude projection for 1900 UTC 14 February 2004 (top), 1900 UTC 28 February 2004 (middle) and 2000 UTC 03 March 2004 (bottom).

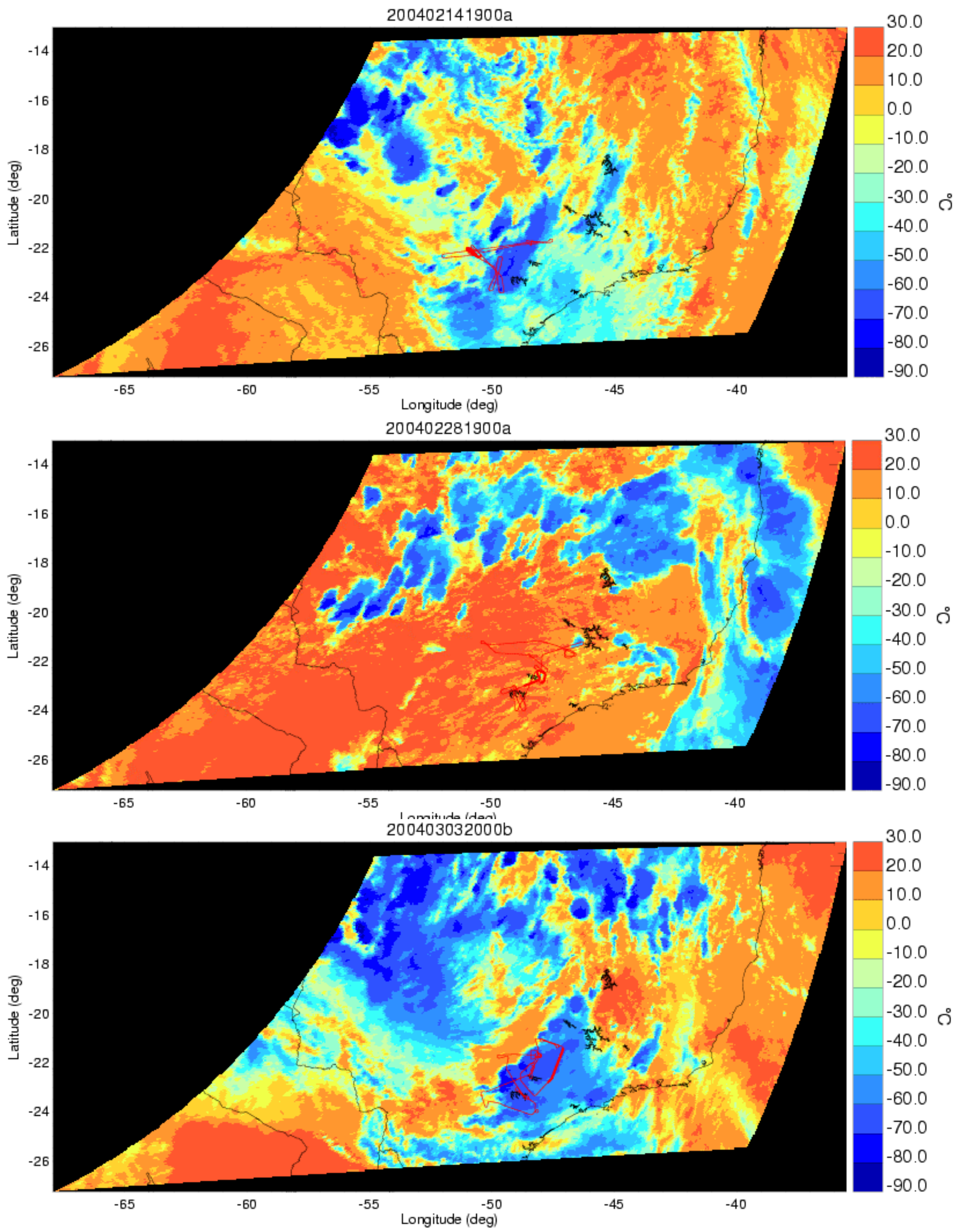

Figure 2. Same as Fig. 1, but MSG data only from channel 9 (10.8 μm). The minimum temperatures in the three cloud systems penetrated by the Falcon are approximately -70°C, -50°C, and -80°C from top to bottom, respectively (derived from the MSG data at the given time instants only). This corresponds to about 14.5, 11.5, and 16 km altitude for the three cases, respectively.

4 RESULTS

On 14th February a cluster of weak thunderstorm cells with heavy rain formed in a large scale convergence zone of moderate strength. Cloud tops reached typically up to about 13 km altitude, with a few towers reaching temporarily even higher.

The cluster of thunderstorms was penetrated by the Falcon aircraft four times at an almost constant flight level (~11 km) as shown in Figure 3. The high NO/NO_y ratio (0.6-0.8) in the anvil outflow indicates that NO was recently produced (mainly by lightning). In fact the NO_x/NO_y ratio is even closer to unity because of quick partial conversion of NO with O_3 to NO_2. The anvil outflow was about 40 km wide, and contained rather constant mean NO mixing ratios (0.4-0.5 nmol mol^{-1}) during each passage. In comparison, the background NO mixing ratio outside the clouds was ~0.1 nmol mol^{-1}. Furthermore, during the anvil passages CO increased to 100-105 nmol mol^{-1} and O_3 decreased down to 25-35 nmol mol^{-1} compared to background mixing ratios (80-90 and 35-45 nmol mol^{-1}, respectively). These observations confirm that anvil air partly originated from the boundary layer and was rapidly transported upwards by the strong updrafts in deep convection. In the boundary layer (measured during take-off and landing), CO and O_3 mixing ratios were in a similar range (110-120 and 20-30 nmol mol^{-1}, respectively), as in the anvil outflow. In this case both CO and O_3 were useful tracers for boundary layer air in the anvil outflow.

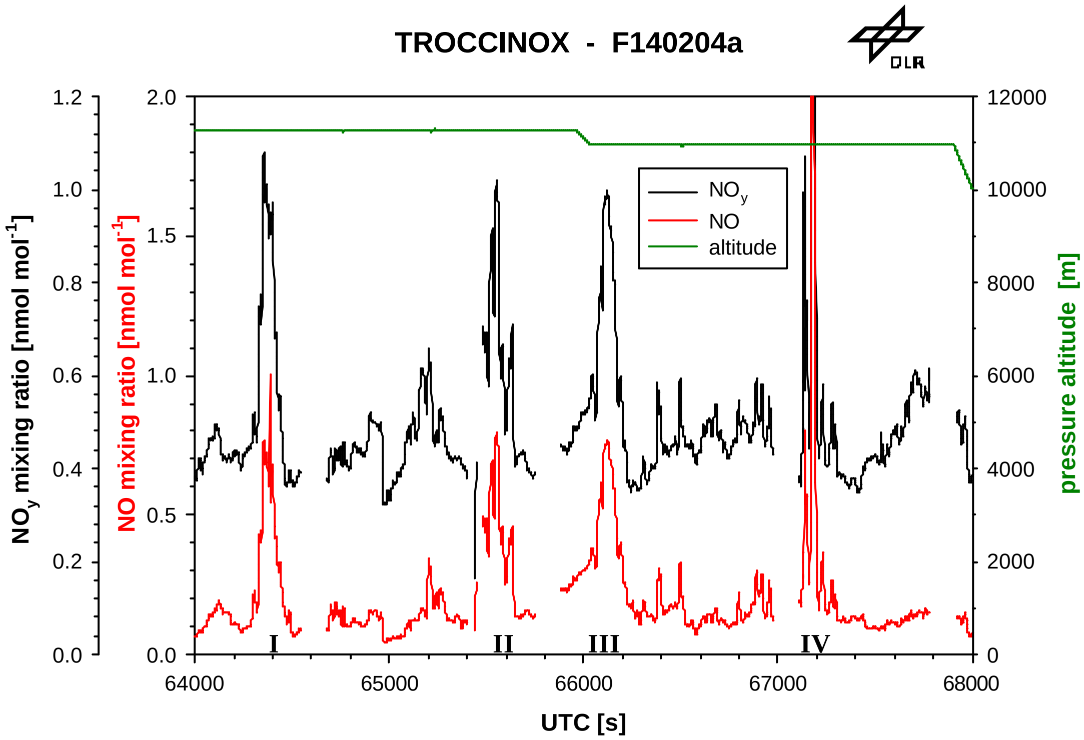

Figure 3. Time series (1 s resolution) of NO and NO_y mixing ratios on 14th February 2004. A thunderstorm cluster was penetrated four times (I-IV) at 11 km altitude, which is clearly seen in the enhanced NO and NO_y mixing ratios.

On 28th February an isolated thundercloud formed due to solar heating of the lower troposphere and weak convergence near the ground in an otherwise rather dry and stable airmass south of a weak cold front which reached north of Sao Paulo. The upper troposphere exhibited strong SW winds. Large anvils were observed during the daytime and the thunderstorms disappeared in the evening. The thunderstorm observed had a structure similar to isolated thunderstorms forming over Europe.

The aircraft penetrated the anvil outflow of the isolated thunderstorm at three different levels (10.7, 10.1, and 8.8 km) as shown in Figure 4. The mean NO mixing ratios measured during each passage were 1.2, 1.4, and 0.8 nmol mol^{-1}, respectively. The high NO/NO_y ratio (0.7-0.8) indicates that NO was recently produced. As observed in the previous case, CO mixing ratios in the anvil outflow were enhanced by 10-20 nmol mol^{-1} in comparison to the background. However, here the background CO (40-50 nmol mol^{-1}) was much lower in comparison to the previous case. In the boundary layer CO mixing ratios ~100 nmol mol^{-1} were measured. The measured O_3 mixing ratios were very different during each passage. During the first passage, O_3 was in the range of the mixing ratios measured in the background and in the boundary layer (40-50 nmol mol^{-1}). During the second and the third passage, mean O_3 mixing ratios were in the range of 140-150 nmol mol^{-1}. The cause of these high O_3 mixing ratios (real or artificial) is not yet known and work is ongoing to investigated this in more detail. Similar kind of signatures (tropospheric "ozone clouds") have been observed during CARIBIC flights as reported by *Zahn et al.* (2002).

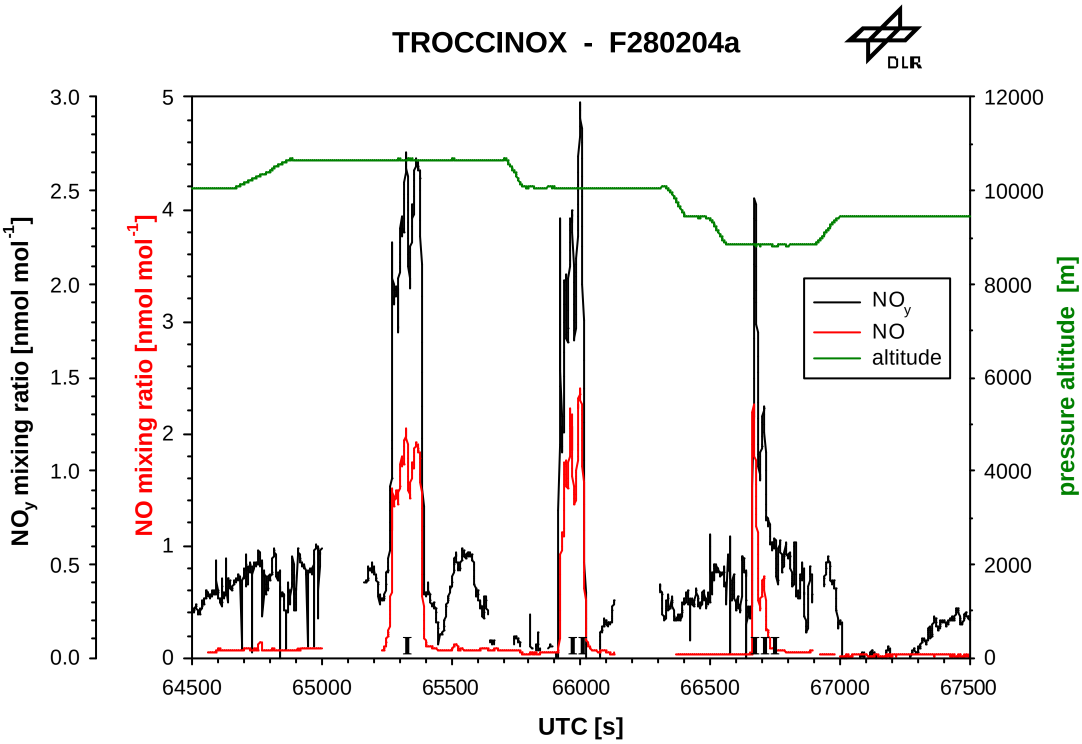

Figure 4. Time series (1 s resolution) of NO and NO_y mixing ratios on 28th February 2004. An isolated thunderstorm was penetrated three times (I-III) at different altitudes between 9 and 11 km altitude, which is clearly seen in the enhanced NO and NO_y mixing ratios.

For March 3rd, an upper ridge axis crossed southern Brazil, supporting deep convection. In the morning, clear skies dominated with strong solar heating of the lower troposphere. During noon, first convective cells developed just above the airport. During the afternoon, a large cluster of thunderstorms formed over the whole region, with widespread tropical convection and associated high flash rates. Two flights were scheduled, one in the morning to measure the airmass before convection set in, and one local convection flight in the afternoon. Here, we report on the results of the afternoon flight. On the next day, it was possible to measure the outflow plume of this convection.

During the afternoon flight on March 3rd, fresh outflow (NO/NO_y= 0.5-0.8) from embedded thunderstorm cells was investigated during five passages at flight levels between 9.1 and 11.3 km as shown in the Figures 5a-b. In comparison to the other two flights, the NO signatures in these thunderclouds were very spiky (NO up to 45 nmol mol^{-1}, NO_y and NO_x up to 60 nmol mol^{-1}). A high NO background mixing ratio in the range of 0.4 nmol mol^{-1} was measured, which indicated a widespread impact of lightning-produced NO. Mean NO mixing ratios, during each of the five selected passages (~30 km wide), varied between 1.4 and 1.8 nmol mol^{-1}. The CO mixing ratio in the upper troposphere was almost constant during the whole flight (100-110 nmol mol^{-1}), and during the five selected passages no clear increase in CO was observed as in the previous cases. CO in the boundary layer was only slightly enhanced (110-120 nmol mol^{-1}). In this case CO was not a useful tracer for boundary layer air in the thundercloud outflow region. The vertical CO profile showed a pronounced C-shape with high mixing ratios in the boundary layer and the upper troposphere, and mixing ratios down to 70 nmol mol^{-1} in 7 km. As described for the previous case, O_3 was strongly enhanced (even up to 170 nmol mol^{-1}) during the five selected passages, and was therefore also no useful tracer for boundary layer air.

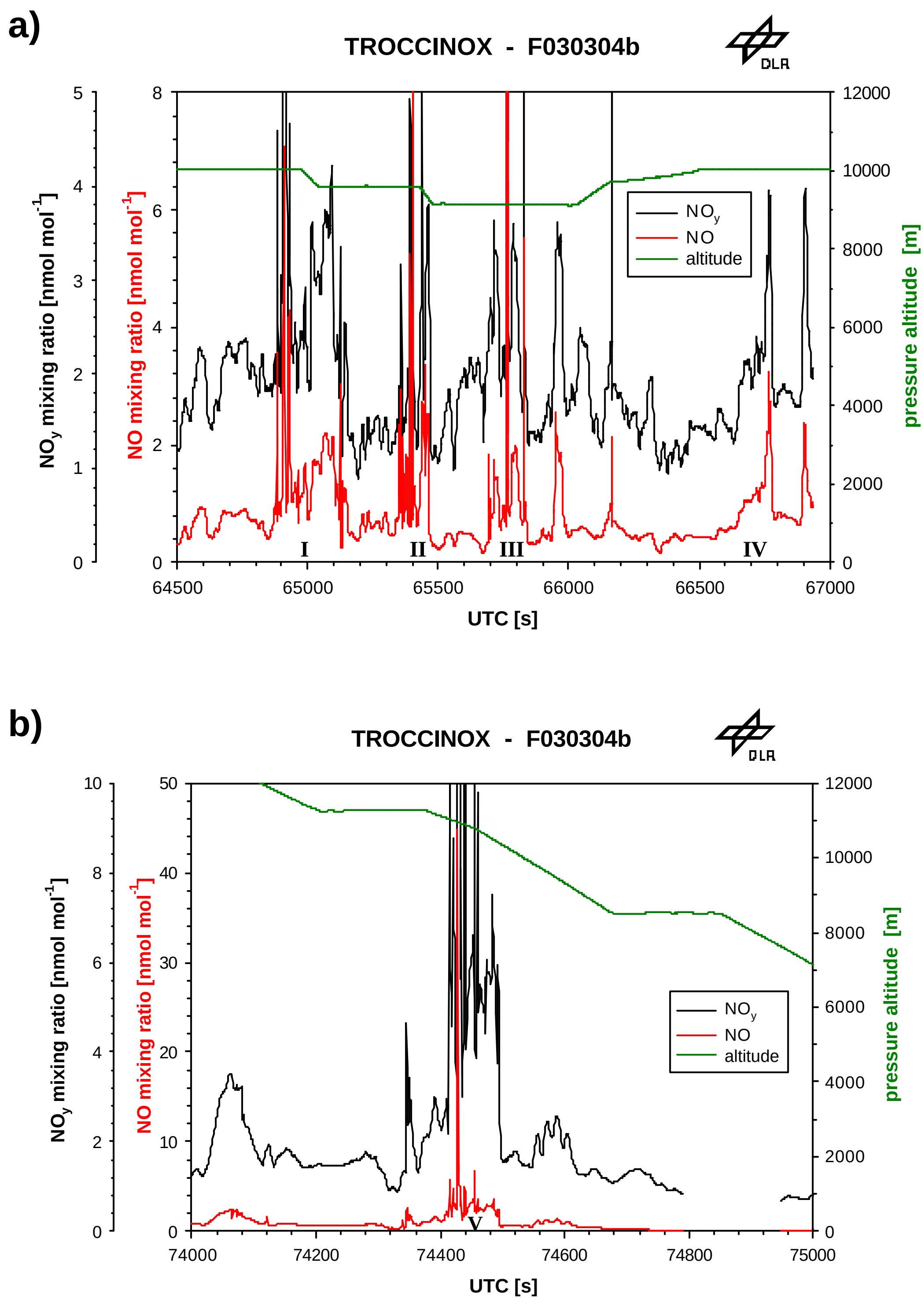

Figure 5. Time series (1 s resolution) of NO and NO_y mixing ratios on 3^{rd} March 2004. Large clusters of thunderstorm cells were penetrated four times (I-IV) at different altitudes between 9 and 10 km altitude, which is clearly seen in the enhanced NO and NO_y mixing ratios (a). In (b) enhanced NO and NO_y mixing ratios were observed during the descent in one of the thunderstorm cells from 11 to 10 km altitude. The 1 s maximum values reach 45 nmol mol^{-1} for NO and 60 nmol mol^{-1} for NOy.

	TROCCINOX			LINOX/EULINOX	
Case	**140204**	**280204**	**030304b**	**medium**	**large**
Cloud top, km	14.5	11.5	16		
Flight altitude, km	11-11.3	8.8-10.7	9.1-10		
NO_m	0.4	1.1	1.5	0.7	1.3
NO_{max}	3.2	2.4	5.0	2.6	3.8
NOx_m	0.5	1.3	1.9	1.3	2.2
NOx_{inflow}	<0.1	<0.2	<0.2	0.5	0.5
Δx, km	40	25	30	30	45
Δz, km	1	1.9	1	1	1
v_a-v_s, m s^{-1}	7	11	12	8	13
Fc, 10^8 kg s^{-1}	1.1	2.0	1.5	1.3	2.3
P(NOx), Tg(N) yr^{-1}	1.7*	7.8	8.6*	3.1 (2-4)	11.7 (10-13)
NO at 2 km	0.02	0.05	0.03		
CO_{anvil}	100-110	50-70	100-110		
$CO_{background}$	80-90	40-50	100-110		
CO_{BL}	100-110	100	110-120		
$O_{3,anvil}$	25-35	50 (150!)	50-60 (171!)		
$O_{3,background}$	35-45	40-50	50		
$O_{3,BL}$	20- 30	40-50	40		

Table 1. Parameters of observed convective events during TROCCINOX and comparison to European cases from LINOX (*Huntrieser et al.*, 1998); all mixing ratios in nmol mol^{-1}.
* Lower limit estimates, since only the lower part of the anvil outflow was investigated.

5 DISCUSSION

The main parameters of the three cases are listed in Table 1. The results can be compared to previous measurements of NO in anvil outflows during airborne field experiments in Europe (LINOX, EULINOX, EXPORT, and CONTRACE, *Huntrieser et al.*, 1998, 2002, 2004), in Florida (CRYSTAL-FACE, *Ridley et al.*, 2004) and in Brazil (TRACE-A, *Pickering et al.*, 1996). Furthermore, for the selected TROCCINOX thunderclouds the mass flux out of the anvils and the NO_x production rate by lightning, according to the methods introduced by *Chameides et al.* (1987), were estimated. In these (still preliminary) TROCCINOX estimates, see Table 1, it is assumed that the contribution to anvil-NO_x from the boundary layer was negligible. In previous measurements over Europe, this contribution had to be taken into account (about 30% of total anvil-NO_x). In most cases, a vertical anvil thickness of Δz = 1 km was assumed. This value may be adjusted to Lidar and Radar observations or detailed model results in the future.

Based on this preliminary analysis, the TROCCINOX nitrogen oxides measurements in the anvil outflow are in the same range as found during previous European experiments and the TRACE-A experiment in Brazil. However, during CRYSTAL-FACE in Florida, average NO mixing ratios in anvil outflow were higher (range 2-5 nmol mol^{-1}). These elevated NO mixing ratios were measured at higher flight levels, between 12 and 14 km, levels that were not reached by the Falcon aircraft during TROCCINOX and over Europe.

The first selected TROCCINOX case (14[th] February) was, concerning lightning-produced NO_x, similar to a medium European thunderstorm as defined by *Huntrieser et al.* (1998). A rough estimate of global lightning-produced NO_x, if we assume that the 14[th] February case was a typical global storm, gave ~2 Tg(N) yr^{-1}. The second TROCCINOX case (28[th] February) were comparable to a strong isolated

European thunderstorm, with about ~8 Tg(N) yr^{-1}. The third TROCCINOX case (3rd March) gave ~9 Tg(N) yr^{-1} , at least as high as in the European cases.

Especially for the TROCCINOX flight from 3rd March (in tropical airmass), our Lidar and the Meteosat observations show that the cloud tops reached up to 16 km, far higher than reached by the aircraft. In this and the first TROCCINOX cases only the lower part of the anvil outflow was probed and we have no information about NO mixing ratios higher up in the anvil. Therefore our estimates must be seen as lower limit estimates.

6 CONCLUSIONS

For the first time, a systematic study of continental thunderstorms in the tropics has been performed.

Both non-tropical and tropical thunderstorms were investigated during TROCCINOX.

The measured CO (and O_3) concentrations were sometimes useful tracers for transport of airmasses from the boundary layer or the lower troposphere into the anvil region. The data show larger variability in the convective transport of trace gases than found in LINOX/EULINOX thunderstorms.

In tropical airmasses, high NO and CO background mixing ratios were observed. In the anvil outflow, spiky NO structures above background were observed. Some of the spikes were notably wide (order several 10 km) indicating outflow from a thunderstorm anvil, others were narrow (order 200 m) clearly originating from fresh lightning events.

From the still preliminary analysis of the few cases it appears that the non-tropical and tropical TROCCINOX thunderstorms exhibit similarly enhanced nitrogen oxides concentrations (0.5-1.9 nmol mol^{-1}) and mass fluxes (1.1-2.0 x 10^8 kg s^{-1}) as found in the LINOX/EULINOX thunderstorms. The three TROCCINOX case studies indicate global lightning-NO_x production rates between 2 and 9 Tg(N) yr^{-1}.

However, for TROCCINOX we have to take into account that only the lower part of the anvil outflow was investigated. We have no information about NO mixing ratios in the upper part of the anvil. Moreover, we may have underestimated the thickness of the anvil outflow regions. Therefore, the estimates for the TROCCINOX cases are lower limit estimates.

Further studies are needed to get reliable estimates of the lightning-NO_x source rate. In cooperation with our Brazilian partners, analysis of the Radar and lightning observations are being prepared to assess the size and the electrical activity of the observed cases in comparison to long-term statistics of such observations. Further measurements are being prepared, and the comparisons between the measured data and the results from chemical transport models are underway. By best fitting global and mesoscale model results to the TROCCINOX observations we expect to be able to determine a better estimate for the lightning-NO_x source rate.

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