Impact of thunderstorms on the NO_x budget in the upper troposphere over Europe: Observations from airborne measurements during LINOX, EULINOX, EXPORT, and CONTRACE

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Abstract. Trace gas measurements in the convective outflow from European thunderstorms have been performed with the German research aircraft *Falcon* during the LINOX (1996), EULINOX (1998), EXPORT (2000), and recently CONTRACE (2003) field experiments. Here we compare the results from all four campaigns with focus on the contribution of thunderstorms to the NO_x (=NO+NO_2) budget in the upper troposphere. A rough estimate for the global amount of lightning-produced NO_x from CONTRACE resulted in ~4 TgN yr^{-1}. For Europe the amount of lightning-produced NO_x was estimated to 0.06 TgN yr^{-1}, which is less than the contribution from aircraft emissions (0.1 TgN yr^{-1}).

1. Introduction

Thunderstorms contribute to the upper tropospheric NO_x budget in two ways: (1) NO_x is produced by lightning, and (2) NO_x is uplifted from the boundary layer (BL) by the updrafts. The amounts of both of these NO_x sources are still very uncertain. For some remote regions the NO_x contribution from the BL can be neglected. However, for Europe, where our measurements were performed, the BL is frequently polluted which has to be considered. Most estimates of lightning-produced NO_x are in the range 2-20 TgN yr^{-1} [*Huntrieser et al.*, 2002]. These values are considerable higher than NO_x emitted from aircraft (<1 TgN yr^{-1}). NO_x emitted in the upper troposphere is of great scientific interest, since it has a strong impact on the ozone and HO_x budget [*Liu*, 1977].

2. Results from Field Experiments

Between 1996 and 2003 the DLR (Deutsches Zentrum für Luft- und Raumfahrt), located in Oberpfaffenhofen near Munich in southern Germany, coordinated four airborne field experiments with focus on the NO_x outflow from deep convection in the upper troposphere over western Europe. More than 100 penetrations of deep convective clouds with and without lightning have been performed up to now, mainly between 7-10 km altitude. In comparison, the average tropopause altitude during the campaigns was 11 km. The DLR research aircraft *Falcon* was equipped with instrumentation for the measurement of NO, NO_2, NO_y, O_3, CO, CO_2, and J(NO_2) [see Table 1 in *Huntrieser et al.*, 2002].

The NO_x content observed in thunderstorm anvils over Europe is a mixture of lightning-produced NO_x (LI-NO_x) and upward transport of boundary layer NO_x (BL-NO_x). To separate the contribution from LI-NO_x and BL-NO_x, tracers for boundary layer air like O_3 and CO have been used in earlier studies [*Dickerson et al.*, 1987]. For the analyses of our data we used CO_2-NO_x and CO-NO_x correlations to estimate the amount of BL-NO_x in the anvils [*Huntrieser et al.*, 1998, 2002]. Furthermore, these correlations agreed well with observations in deep convective clouds without lightning (only transported BL-NO_x).

During the LINOX experiment in July-August 1996 [*Huntrieser et al.*, 1998] the mean NO_x concentration in deep convective clouds without lightning was 0.5 ± 0.3 nmol mol^{-1}. The outflow from an average LINOX thunderstorm contained 1.3 ± 0.5 nmol mol^{-1} NO_x, which corresponds to 40% BL-NO_x and 60% LI-NO_x. The maximum NO mixing ratio measured was 3.8 nmol mol^{-1}. In comparison, during the EULINOX experiment in July 1998 [*Huntrieser et al.*, 2002], the maximum NO mixing ratio measured very close to the main lightning channel was 25 nmol mol^{-1}. The mean NO_x concentration in deep convective clouds without lightning was 0.4 ± 0.2 nmol mol^{-1}. The outflow from an average EULINOX thunderstorm contained 1.3 ± 0.7 nmol mol^{-1} NO_x, which corresponds to 30% BL-NO_x and 70% LI-NO_x.

NO_x mixing ratios in the same range were also measured in the outflow from thunderstorms observed during the EXPORT experiment in August 2000, except for one flight on 5th August. During more than 10 hours a stationary thunderstorm complex over the Marseille region (France) pumped polluted air masses into the upper troposphere. The flash rate was rather low with 15-20 flashes per hour. About 100 km downwind of the thunderstorm complex, between Torino (northwestern Italy) and Grenoble (southeastern France) at 9.4 km altitude, the aircraft penetrated the outflow (cloud free), which indicated a strong polluted air mass. Over a length of 100 km the average mixing ratios (in nmol mol^{-1}) were: NO = 4.0 (peak 7.7), NO_x = 5.8, NO_y = 5.8 (peak 9.9), CO = 119, and O_3 = 87. These are the highest average nitrogen oxides mixing ratios ever measured by our group in the outflow from thunderstorms. Before and after the outflow was penetrated, the aircraft was flying in stratospheric air masses with ozone and CO mixing ratios in the range of 200-300, and 40-60 nmol mol^{-1}, respectively. The thunderstorm outflow was in this case trapped by two tropopause foldings caused by the roll-up of a potential vorticity (PV) streamer. Between the two tropopause foldings, mixing with the surrounding air was inhibited, which may explain the high nitrogen oxides mixing ratios observed. On the return flight between Grenoble and Torino, at a slightly lower flight level at 9.0 km, the average mixing ratios (in nmol mol^{-1}) were distinctly lower: NO = 1.1, NO_x = 1.8, and NO_y = 2.3, and in the range of the mean values obtained from LINOX and EULINOX.

More recently, the CONTRACE field experiment was performed in July 2003. The mean BL-NO_x concentration in the anvil outflow was 0.5 ± 0.2 nmol mol^{-1} (estimated from the CO-NO_x correlation). The outflow from an average CONTRACE thunderstorm contained 1.2 ± 0.4 (range 0.7-1.7) nmol mol^{-1} NO_x, which corresponds to 40%

$BL-NO_x$ and 60% $LI-NO_x$. The maximum NO mixing ratio measured was 7.2 nmol mol^{-1}, and the mean NO_y mixing ratio was 2.1 ± 0.6 nmol mol^{-1}. Typical temporal variations in NO and NO_y along the flight track in an anvil outflow is shown in Figure 1. CO was used as tracer for BL air in the anvil outflow region as shown in Figure 2.

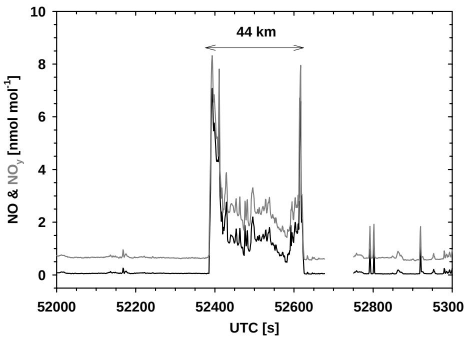

Figure 1. Penetration of anvil outflow (44 km wide at 9.1 km altitude) on 15th July 2003.

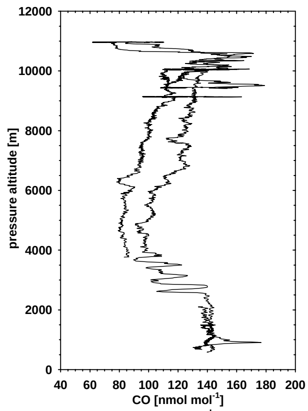

Figure 2. Vertical CO profile on 16th July 2003.

3. Estimate of lightning-produced NO_x

Our measurements in the anvil outflow over the past years show a rather constant amount of NO_x produced by lightning in an average European thunderstorm: LINOX: 0.8, EULINOX: 0.9, and CONTRACE: 0.7 nmol mol^{-1} NO_x. These mixing ratios can be used to make a rough estimate of the global lightning NO_x production rate, $P(NO_x)$ [*Chameides et al.*, 1987]

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

$[NO_x]$ is the average volume mixing ratio in the anvil produced by lightning, 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 x 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. The mean values of these parameters during CONTRACE are: $v_a - v_s = 10\ m\ s^{-1}$, $\rho_a = 0.4\ kg\ m^{-3}$, Δx = 77, Δz = 1 km, and $F_C = 3.1 \times 10^8\ kg\ s^{-1}$, which result in the average global lightning NO_x production rate, $P(NO_x)$ ~7 (range 4-10) TgN yr^{-1}. However, here we have to take into account that the major part of the thunderstorm systems investigated during CONTRACE were thunderstorm lines that consisted of several cumulonimbus cells. Δx was twice as high as found during LINOX and EULINOX (30 km). We therefore have to reduce the parameter S and then end up with ~4 TgN yr^{-1} for the global lightning NO_x production rate. This value is in accordance with our earlier estimates from LINOX (~4 TgN yr^{-1}) and EULINOX (~3 TgN yr^{-1}).

The final step is then to estimate the importance of European lightning NO_x emissions in comparison to other NO_x emissions. The average European flash density (2 flashes km^{-2} yr^{-1}) [*Simpson et al.*, 1999] is used to estimate the annual flash rate over Europe (0.7 flashes s^{-1}) and to compare it to the global flash rate (44 flashes s^{-1}) [*Christian et al.*, 2003]. If we assume that the NO_x production rate by lightning is proportional to the flash rate, then over Europe about 0.06 TgN yr^{-1} is produced by lightning. The amount of $BL-NO_x$ transported to the upper troposphere over Europe is estimated to 0.04 TgN yr^{-1}. If we consider the transport by larger mesoscale convective systems (MCS) and extratropical cyclones, which together transport about six times more air from the BL than single thunderstorms [*Cotton et al.*, 1995], we obtain almost 0.3 TgN yr^{-1} for the European BL transport to the upper troposphere. In comparison, the amount of NO_x emitted from aircraft in the upper troposphere over Europe is 0.1 TgN yr^{-1} [*D. Lee*, personal communication, 1999]. If we consider the total amount of NO_x emitted in the BL over Europe (7 TgN yr^{-1}) [*Simpson et al.*, 1999], then about 4% of the $BL-NO_x$ is transported upward by convection and frontal systems.

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