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# Estimating the $\text{NO}_x$ produced by lightning from GOME and NLDN data: a case study in the Gulf of Mexico

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**Abstract.** Nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) play an important role in tropospheric chemistry, in particular in catalytic ozone production. Lightning provides a natural source of nitrogen oxides, dominating the production in the tropical upper troposphere, with strong impact on tropospheric ozone and the atmosphere's oxidizing capacity. Recent estimates of lightning produced  $\text{NO}_x$  ( $\text{LNO}_x$ ) are of the order of 5 Tg [N] per year with still high uncertainties in the range of one order of magnitude.

The Global Ozone Monitoring Experiment (GOME) on board the ESA-satellite ERS-2 allows the retrieval of tropospheric column densities of  $\text{NO}_2$  on a global scale. Here we present the GOME  $\text{NO}_2$  measurement directly over a large convective system over the Gulf of Mexico. Simultaneously, cloud-to-ground (CG) flashes are counted by the U.S. National Lightning Detection Network (NLDN<sup>TM</sup>), and extrapolated to include intra-cloud (IC)+CG flashes based on a climatological IC:CG ratio derived from NASA's space-based lightning sensors. A series of 14 GOME pixels shows largely enhanced column densities over thick and high clouds, coinciding with strong lightning activity. The enhancements can not be explained by transport of anthropogenic  $\text{NO}_x$  and must be due to fresh production of  $\text{LNO}_x$ . A quantitative analysis, accounting in particular for the visibility of  $\text{LNO}_x$  from satellite, yields a  $\text{LNO}_x$  production of 90 (32–240) moles of  $\text{NO}_x$ , or 1.3 (0.4–3.4) kg [N], per flash. If simply extrapolated, this corresponds to a global  $\text{LNO}_x$  production of 1.7 (0.6–4.7) Tg [N]/yr.

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## 1 Introduction

Nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) play an important role in atmospheric chemistry. In the troposphere, they drive catalytic ozone production. Furthermore,  $\text{NO}_x$  controls OH concentration and thus the atmosphere's oxidizing capacity. In total, about 44 Tg [N] of nitrogen oxides are released annually, half of which are due to fossil fuel combustion (Lee et al., 1997). Further large sources are biomass burning ( $\approx 8$  Tg [N]/yr) and soil emissions ( $\approx 7$  Tg [N]/yr).

Lightning produced  $\text{NO}_x$  (hereafter denoted by  $\text{LNO}_x$ ) is estimated to contribute about 5 Tg [N]/yr (Lee et al., 1997). However, the best estimates of recently published studies still vary between 0.9 and 12.2 Tg [N]/yr (e.g. Nesbitt et al., 2000; Price et al., 1997), and the given uncertainties typically are one order of magnitude. Thus lightning is the least known important source of nitrogen oxides. Furthermore, in contrast to other sources,  $\text{LNO}_x$  is directly released also in the upper troposphere where background levels of  $\text{NO}_x$  are low and the lifetime of  $\text{NO}_x$  is of the order of a few days, i.e. several times longer than for the boundary layer ( $\approx$  hours). Hence both tropospheric ozone as well as OH concentrations are particularly sensitive to  $\text{LNO}_x$  (e.g. Stockwell et al., 1999; Labrador et al., 2004). For the correct assessment of  $\text{NO}_x$  inventories, a prerequisite for reliable model calculations of atmospheric chemistry, better knowledge on  $\text{LNO}_x$  is essential.

Over the last decades, several studies using different methods have been performed to estimate  $\text{LNO}_x$  production. A common bottom-up approach is to assess (a) the production of  $\text{NO}_x$  per energy unit, (b) the released energy per flash and

(c) the global frequency of flashes, and to estimate the global  $\text{LNO}_x$  production as the product of these quantities. Literature values range over some orders of magnitude, as a result of the many assumptions and necessary extrapolations of laboratory measurements involved (see Price et al., 1997 for an overview). Further complications arise from differences in cloud-to-ground (CG) and intra-cloud (IC) flashes. IC flashes are more frequent, but CG flashes are more energetic and hence produce more  $\text{LNO}_x$  per flash, concrete numbers still being under discussion (see e.g. Fehr et al., 2004 and references therein).

In-situ measurements of  $\text{LNO}_x$  have been performed in several aircraft campaigns, where global  $\text{LNO}_x$  estimates range from 0.9–220 Tg [N] per year (for an overview see Huntrieser et al., 1998). In their own study, Huntrieser et al. (1998) found annual  $\text{LNO}_x$  production to be 4 (0.3–22) Tg [N]. More recently, Huntrieser et al. (2002) analyzed European thunderstorms recorded during the EULINOX project in detail and estimated the annual  $\text{LNO}_x$  production at 3 Tg [N].

Also chemical transport models (CTMs) have been used to restrict the range of  $\text{LNO}_x$  production by comparing modeled  $\text{NO}_x$  concentrations for different  $\text{LNO}_x$  scenarios with local field measurements. The studies by Levy et al. (1996), Tie et al. (2002), and Jourdain and Hauglustaine (2001) find about 5 Tg [N], 2–6 Tg [N], and 3.5–7 Tg [N], respectively, as best estimates for yearly global  $\text{LNO}_x$  production.

The fact that several independent approaches result in a global  $\text{LNO}_x$  production of about 5 Tg [N] per year confirms that at least the order of magnitude can be expected to be correct. However, the uncertainties of the different methods are still quite high, indicating the need for further, independent information.

Satellite based measurements of atmospheric trace gases are a powerful addition to measurements from conventional platforms. They provide a global dataset with uniform instrumental features, and meanwhile span several years of measurements. The spectral data from the Global Ozone Monitoring Instrument GOME allow to determine column densities of various trace gases, in particular  $\text{NO}_2$  (e.g. Leue et al., 2001; Richter and Burrows, 2002; Martin et al., 2002). By estimating and subtracting the stratospheric column, and accounting for radiative transfer, tropospheric  $\text{NO}_2$  column densities can be derived from GOME data (e.g. Leue et al., 2001; Richter and Burrows, 2002; Beirle et al., 2003; Martin et al., 2003; Boersma et al., 2004).

The global view offered by satellite observations provides new insights on the spatial distribution of  $\text{NO}_x$  sources (e.g. Velders et al., 2001; Leue et al., 2001; Richter and Burrows, 2002; Martin et al., 2003; Beirle et al., 2004b, d). The analysis of characteristic temporal and spatial patterns has been used to identify and quantify the magnitude of different  $\text{NO}_x$  sources, e.g. continental anthropogenic emissions (Martin et al., 2003; Beirle et al., 2003), ship emissions (Beirle et al., 2004c; Richter et al., 2004), biomass burning (e.g. Richter

and Burrows, 2002; Spichtinger et al., 2004), or soil emissions (Jaeglé et al., 2004).

On account of this successful use of GOME  $\text{NO}_2$  data it is obvious to investigate  $\text{LNO}_x$  from GOME as well. However, while the signal of e.g. industrial sources is often unambiguous, the clear detection of  $\text{LNO}_x$  is more complex: In contrast to anthropogenic emissions, the occurrence of lightning is highly variable in space and time. Furthermore, lightning occurs predominantly in the late afternoon or evening, whereas GOME measurements take place before local noon. Due to the longer lifetime of  $\text{NO}_x$  of several days in the upper troposphere (Jaeglé et al., 1998),  $\text{LNO}_x$  can accumulate to detectable amounts, but these aged  $\text{LNO}_x$  plumes are diluted, and the spatial patterns are faint compared to the sharp  $\text{NO}_2$  maxima of boundary layer sources.

Moreover, thunderstorms are extreme weather events. As a consequence of deep convection and downdraft motions, the profile of lightning produced  $\text{NO}_x$  as well as  $\text{NO}_x$  from boundary layer sources is strongly modified. A large fraction of the produced  $\text{LNO}_x$  is uplifted in the anvil, resulting in a pronounced C-shaped profile of  $\text{LNO}_x$  (e.g. Pickering et al., 1998; Fehr et al., 2004). Furthermore thunderstorms are accompanied by high and thick clouds. Both factors strongly affect the visibility of  $\text{NO}_2$  from satellite.

Despite these difficulties, some studies report a correlation of lightning activity and increased  $\text{NO}_2$  column densities. Zhang et al. (2000) used  $\text{NO}_2$  data from the Upper Atmosphere Research Satellite (UARS) to substantiate a (rather qualitative) link between lightning activity and high levels of  $\text{NO}_2$  in the upper troposphere. Richter and Burrows (2002) report enhanced  $\text{NO}_2$  column densities above clouds due to lightning for Africa. Beirle et al. (2004a) analyzed correlations of monthly means of lightning activity and GOME  $\text{NO}_2$  column densities for Australia and estimated the global  $\text{LNO}_x$  production as 2.7 (0.8–14) Tg [N]/yr. Boersma et al. (2005) compared the 1997 GOME  $\text{NO}_2$  column densities to model output for different tropical regions and give a range for annual  $\text{LNO}_x$  production of 1.1–6.4 Tg [N]/yr.

Besides these statistical approaches, studies on particular lightning events using GOME data have also been reported. Hild et al. (2000) analyzed a lightning event south from Africa coinciding with enhanced  $\text{NO}_2$  column densities from GOME measurements nearby. Choi et al. (2005) found evidence for lightning enhancements of  $\text{NO}_2$  over North America and the western North Atlantic.

Here we present the direct GOME measurement of enhanced  $\text{NO}_2$  column densities over a large convective system in the Gulf of Mexico, while flashes are counted by the U.S. National Lightning Detection Network simultaneously.

## 2 Methods

### 2.1 NO<sub>2</sub> column densities from GOME

For this study we have used data from the Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999). GOME orbits the earth onboard the ERS-2 satellite, flying in a sun-synchronous nearly polar orbit and crossing the equator at 10:30 a.m. local time. The GOME instrument consists of four spectrometers measuring the radiation reflected by the earth in the UV/vis spectral range (240–790 nm) with a resolution of 0.2–0.4 nm. The extent of a GOME ground pixel is 320 km east-west and 40 km north-south (size and orientation of a GOME pixel are illustrated in Fig. 1). Within three days, global coverage is achieved at the equator.

Applying the established Differential Optical Absorption Spectroscopy (DOAS) (Platt, 1994), the GOME spectra are analyzed at 430–450 nm to derive slant column densities, i.e. integrated concentrations along the light path, of NO<sub>2</sub> (Wagner, 1999). The stratospheric fraction of the total NO<sub>2</sub> column at a given latitude is estimated over the remote Pacific at same latitude (e.g. Richter and Burrows, 2002) where the tropospheric pollution is negligible. Assuming the stratospheric NO<sub>2</sub> being independent on longitude, the stratospheric column can be subtracted from the total column, resulting in tropospheric (excess) slant column densities (abbreviated as TSCDs and denoted with *S* hereafter) of NO<sub>2</sub>.

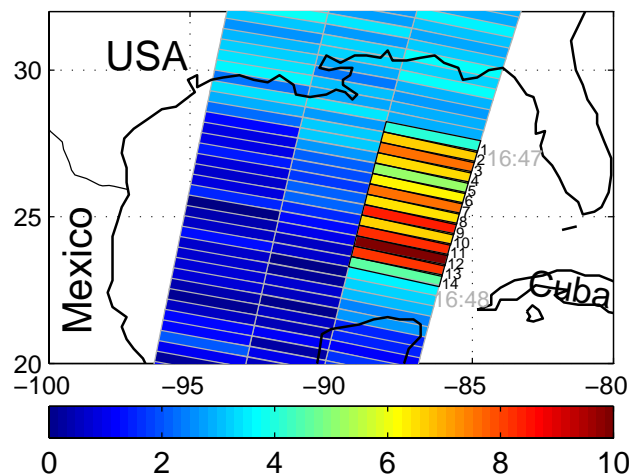
The slant column densities are commonly converted to vertical column densities (VCDs), i.e. vertically integrated concentrations, via the air mass factor (AMF) (Solomon et al., 1987). The AMF (*A*) is defined as the ratio of SCD (*S*) and VCD (*V*), hence VCDs are derived according to

$$V = S/A. \quad (1)$$

The stratospheric AMF depends mainly on the geometric light path, i.e. the solar zenith angle. In the troposphere, the effects of Rayleigh and Mie scattering become more important. Hence tropospheric AMFs depend on the trace gas profile, the ground albedo, the aerosol load and especially on clouds. Tropospheric AMFs are derived from radiative transfer modeling. According to Richter and Burrows (2002), Fig. 2, the tropospheric AMF at 437.5 nm is close to 1 for cloud free conditions, a homogeneous mixing in a boundary layer of 1.5 km height, maritime aerosols, and a surface albedo of 0.05. We apply the tropospheric AMF from Richter and Burrows (2002) for the cloud free pixels of our study.

Conditions for NO<sub>2</sub> from lightning, however, are quite different: deep convection causes high and thick clouds and leads to modified vertical NO<sub>x</sub> profiles. Both factors strongly affect the NO<sub>2</sub> visibility from satellite. The calculation of appropriate AMFs for NO<sub>2</sub> from lightning in the current study is described in Sect. 4.2 in detail.

After subtraction of the stratospheric column and AMF correction, the final data products are tropospheric VCDs that are abbreviated as TVCDs and denoted with *V* hereafter.



**Fig. 1.** GOME NO<sub>2</sub> TSCDs ( $10^{15}$  molec/cm<sup>2</sup>) on 30 August 2000 in the Gulf of Mexico. From 16:47–16:48, a series of 14 eastern pixels (marked in black and numbered) shows enhanced values above  $4 \times 10^{15}$  molec/cm<sup>2</sup>.

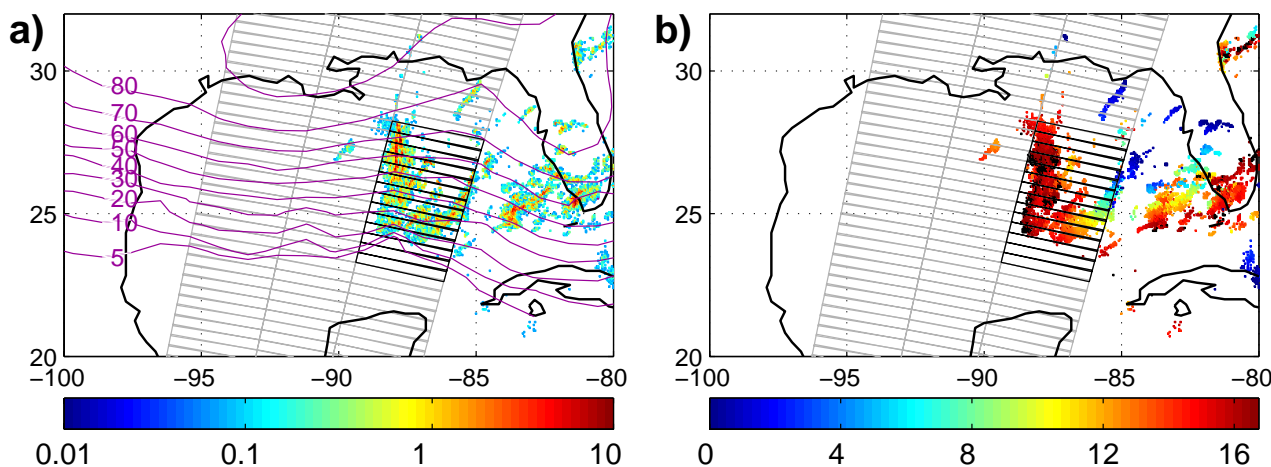
### 2.2 Cloud data

Cloud information is essential for the calculation of AMFs for given satellite measurements as they shield the boundary layer, but enhance the visibility for absorbers at the cloud top due to multiple scattering and above due to the high albedo. Cloud data are retrieved on global scale from various satellite borne VIS, IR and microwave sensors. Hourly infrared satellite images were available from the NOAA GOES-8 geostationary satellite. Channel 4 of the imager on board the satellite measures the intensity of the radiation emitted by the Earth between 10.2 and 11.2  $\mu$ m, providing the temperature of the Earth's surface and cloud tops at 4 km resolution.

In addition to data from such meteorological satellites, cloud information can be obtained from the GOME measurement itself. This has the advantage that the cloud data match the NO<sub>2</sub> observation exactly in space and time. At the IUP Heidelberg, cloud fractions are derived from intensity measurements of the polarization monitoring devices (PMDs) by the HICRU algorithm (Grzegorski et al., 2006). The spatial PMD resolution ( $20 \times 40$  km<sup>2</sup>) is 16 times higher than that of the GOME groundpixel. Thus HICRU provides also information on cloud heterogeneity across the GOME pixel.

### 2.3 Lightning detection: NLDN

The U.S. National Lightning Detection Network (NLDN<sup>TM</sup>) was the source of the archived CG lightning information. At the time of the case evaluated in this study (August 2000), the NLDN was comprised of 106 ground-based electromagnetic sensors operating in the VLF/LF frequency range. These sensors were configured to detect emissions produced by return strokes in CG lightning, and to locate these discharges using



**Fig. 2.** Lightning observations on 30 August 2000 from NLDN. **(a)** Number of detected flashes per  $\text{km}^2$  before the GOME measurement. Purple contour lines display the NLDN detection efficiency. The GOME pixel grid is added as a reference for both subplots. **(b)** Time (UTC) of the last flash detected by the NLDN previous to ERS-2 overpass (16:48). Black dots mark lightning from 16:40–16:48.

a combination of time-of-arrival and magnetic direction finding techniques (Cummins et al., 1998). The median location accuracy was 500 m, and the flash detection efficiency for events with peak current above 5 kA was estimated to be 80–90% over the continental U.S., falling off steadily out to about 500 km outside the network perimeter. Due to the fall-off of detection efficiency in the area of interest for this study, model-based corrections provided by Vaisala Thunderstorm (Tucson, Arizona, see <http://www.vaisala.com/>) were employed to correct for performance fall-off.

#### 2.4 Transport modeling: FLEXPART

GOME measurements at a given location are “snapshots” taken once every 3 days. For the clear identification of  $\text{NO}_x$  sources, effects of transport have to be taken into account. In this study, transport simulations of various  $\text{NO}_x$  tracers (i.e. anthropogenic  $\text{NO}_x$ , aged and fresh  $\text{LNO}_x$ , see Sect. 4.3) are performed with the Lagrangian particle dispersion model FLEXPART (version 6.2) (Stohl et al., 1998, 2005) (see also <http://zardozi.nilu.no/~andreas/flextra+flexpart.html>). FLEXPART simulates the transport and dispersion of non-reactive tracers by calculating the trajectories of a multitude of particles. It was validated with data of various large scale tracer experiments (Stohl et al., 1998) and the model results were compared to different kinds of satellite data. In detail, with respect to this paper, FLEXPART was already successfully used to study the advection of forest fire  $\text{NO}_x$  from Canada to Europe (Spichtinger et al., 2001), to simulate a power plant plume of  $\text{NO}_x$  traveling from South Africa towards Australia (Wenig et al., 2003), and to model the transport of anthropogenic  $\text{NO}_x$  from the US eastcoast towards Europe within a meteorological bomb (Stohl et al., 2003).

FLEXPART is driven by data from the European Centre for Medium-Range Weather Forecasts (ECMWF, 1995). The data set has a temporal resolution of 3 hours (analyses at 00:00, 06:00, 12:00, and 18:00 UTC; 3-h forecasts at 03:00, 09:00, 15:00, and 21:00 UTC), a horizontal resolution of  $1^\circ \times 1^\circ$  and 60 vertical levels. Although the ECMWF model reproduces the large-scale effects of convection, they do not resolve individual deep convective cells. In order to account for subgrid-scale convection, FLEXPART was recently equipped with a convective parameterization scheme (Emanuel and Zivkovic-Rothman, 1999; Emanuel, 1991).

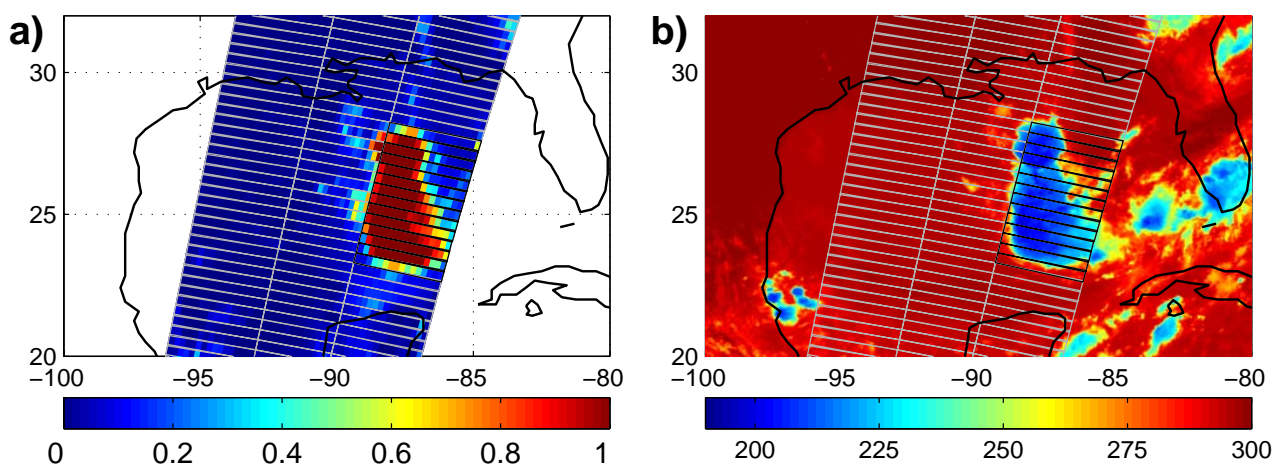
FLEXPART does not model chemical processes. The chemical decay of  $\text{NO}_x$  is considered by assigning the  $\text{NO}_x$  tracer with a constant e-folding lifetime. More information on the  $\text{NO}_x$  tracers simulated (anthropogenic  $\text{NO}_x$ , aged and fresh  $\text{LNO}_x$  and the respective e-folding lifetime) is given in Sect. 4.3.

### 3 Case study: a thunderstorm in the Gulf of Mexico

Lightning frequency is highest in late afternoon, while GOME measurements are taken around 10:30 a.m. local time. Nevertheless, the direct observation of lightning during an ERS-2 overpass occasionally occurs within the large amount of GOME data. An unique event of GOME capturing  $\text{LNO}_x$  just produced is found on 30 August 2000 over the Gulf of Mexico: a sequence of about 14 pixels, measured at 16:47–16:48 UTC, shows high  $\text{NO}_2$  TSCDs (Fig. 1) that by far exceed normal levels over ocean.

These high TSCDs coincide with a strong convective system causing high lightning activity. Figure 2a depicts the flashes detected by the NLDN on 30 August 2000 before the ERS-2 overpass at 16:48 UTC. For better comparison, the





**Fig. 3.** Cloud observations on 30 August 2000. **(a)** Cloud fraction derived from GOME PMD measurements (HICRU). **(b)** Cloud top temperature (K) from IR measurements (GOES) at 16:15 UTC. The temperatures  $<220$  (210) K correspond to cloud top heights  $>12.5$  (14) km. The GOME pixel grid is added as a reference.

GOME pixel grid is overlaid in all subplots. To illustrate the temporal coincidence of flashes and GOME measurement, Fig. 2b displays the time of the latest flash occurrence. More than half of the detected flashes occurred less than 3 h before ERS-2 overpass. Black dots indicate flashes with less than 8 min time difference to the GOME measurement.

Figure 3 shows cloud fractions (a) and cloud top temperatures (b) measured from HICRU and GOES, respectively. HICRU cloud fractions reveal a large cluster of totally clouded PMD pixels, covering an area of  $\approx 500$  km north-south and up to 300 km east-west. CTTs from GOES IR measurements at 16:15 UTC are about 200–220 K. This corresponds to cloud top heights of 12.5–16 km according to ECMWF temperature profiles.

This particular event is unprecedented as the lightning activity coincides perfectly with the GOME measurement both in space (the area affected by lightning fits in the eastern GOME pixels) and in time (most flashes have occurred during the last 3 hours). The lightning event took place over sea and remote from polluted regions. The enhanced  $\text{NO}_2$  TSCDs can not be explained with transport of anthropogenic emissions (see Sect. 4.3.1). The observed enhanced  $\text{NO}_2$  TSCDs are thus unambiguously due to  $\text{LNO}_x$ . As far as we know, such a clear and direct detection of  $\text{LNO}_x$  from satellite has never been reported before.

A closer look on the meteorological situation reveals that the convective complex originates from at least two systems of different history. This is illustrated in Fig. 4 that displays hourly GOES CTTs for 30 August. Figure 4a depicts a convective cell at  $\approx 25^\circ$  N,  $85^\circ$  W, moving WSW and growing during the next hours, becoming the southern part of the large complex detected at 16:15. In fact, this system has already existed several hours before 08:15 (see the blue/cyan dots in

Fig. 2b), and was even active on 29 August at the western coast of Florida.

The northern part of the 16:15 complex, however, is rather young. Tracking it back reveals that it stems from a small, singular cell emerging at about 10:15 and growing rapidly (see white marks in Figs. 4c, d, e), and some smaller systems in the north (gray marks in f, g).

For the quantitative estimation of  $\text{LNO}_x$  given in Sect. 4, we focus our analysis on the northern part of the convective complex, i.e. north from  $\approx 25^\circ$  N corresponding to the GOME pixels 1–9, since the northern convection cells are young and, thus, in contrast to the southern part, free from aged  $\text{LNO}_x$ . Furthermore, the detection efficiency (DE) by NLDN is above  $\approx 30\%$  in the northern part, while it decreases to zero further south (see DE contour lines in Fig. 2a). Nevertheless, we add a discussion of the southern part also in Sect. 4.5.

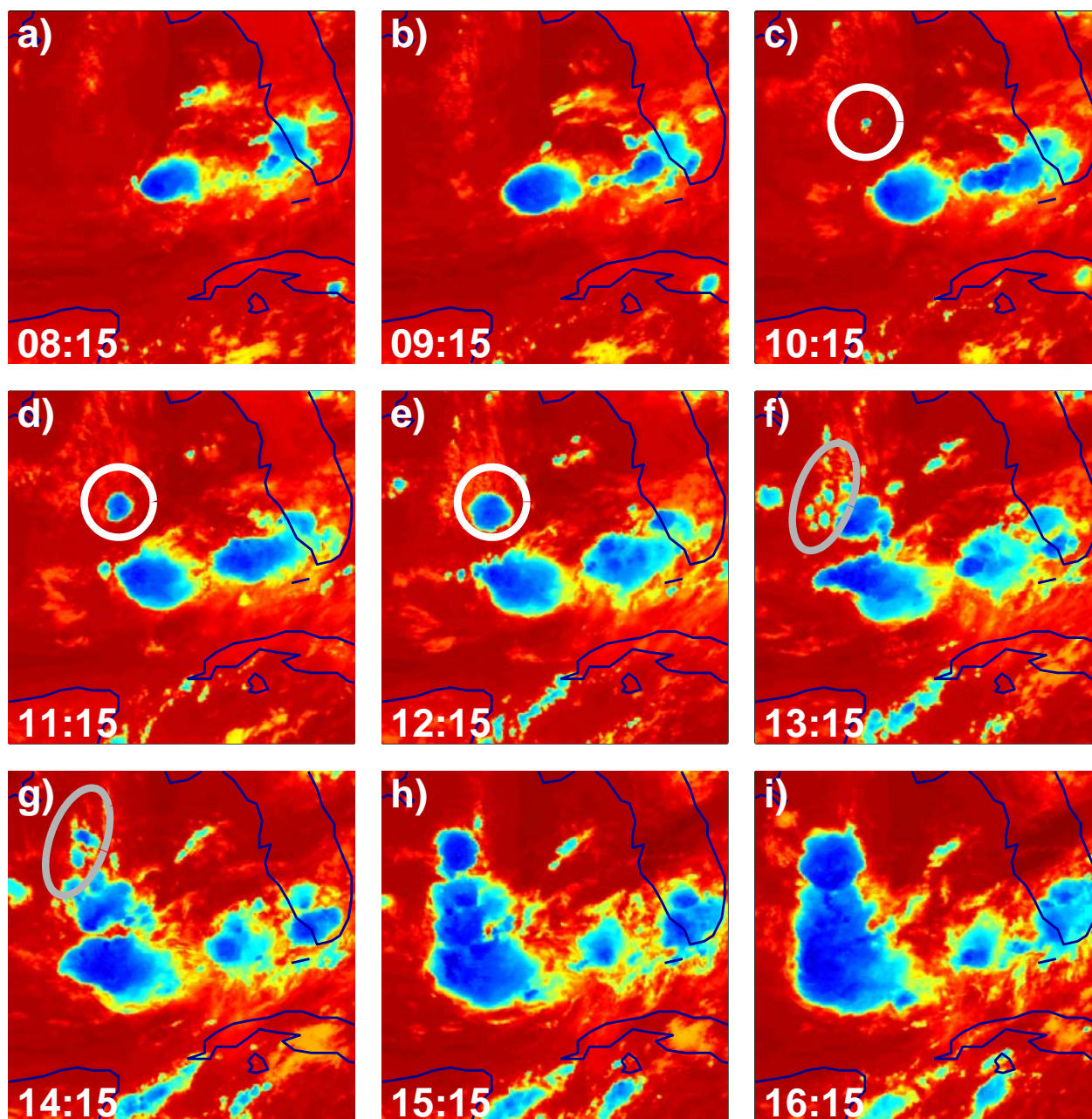
#### 4 Estimate of $\text{LNO}_x$ production

We use this particular lightning event to estimate the in-situ produced  $\text{LNO}_x$ . For this task, the totally produced  $\text{LNO}_x$  in the northern part of the convective complex is estimated and set in relation to the actual number of flashes.

For this quantitative analysis, the following aspects have to be discussed:

First, the actual number of flashes has to be estimated (Sect. 4.1). The flashes detected by NLDN have to be up-scaled since detection efficiency fades over sea and NLDN is insensitive for IC flashes.

Second, the sensitivity of GOME for  $\text{LNO}_x$ , i.e. the conversion of measured  $\text{NO}_2$  TSCDs into  $\text{NO}_x$  TVCDs, will be discussed in Sect. 4.2.



**Fig. 4.** Evolution of the convective complex as monitored by GOES CTT measurements (all times UTC). Color scale as in Fig. 3b. While the southern part (south from 25° N) has a history of several hours, the northern part just established around 10 a.m. (white marks in **c**, **d**, **e**) and after 1 p.m. (gray marks in **f**, **g**).

Third (Sect. 4.3), the role of transport processes has to be considered. NO<sub>x</sub> from anthropogenic sources in the USA, as well as aged LNO<sub>x</sub> from the previous days, may be transported into the considered region and interfere with the freshly produced LNO<sub>x</sub>. On the other hand, the fresh LNO<sub>x</sub> may be partly transported away.

Finally (Sect. 4.4), the total amount of LNO<sub>x</sub> produced will be determined and set in relation to the total number of

flashes: The resulting LNO<sub>x</sub> production (in moles per flash) is

$$E = \frac{\hat{V}^{\text{NO}_x} \cdot A}{N_A} \cdot \frac{1}{n} \quad (2)$$

with  $A$  being the area of the convective system (Sect. 4.4),  $N_A$  being Avogadro's number and  $n$ , the total number of flashes (Sect. 4.1).

The corrected NO<sub>x</sub> TVCD  $\hat{V}$  is calculated as

$$\hat{V}^{\text{NO}_x} = (c_{\text{cloud}} \cdot S_{\text{GOME}}^{\text{NO}_2} - S_{\text{anthr}}^{\text{NO}_2}) \cdot f \cdot c_{\text{aged}} \cdot c_{\text{out}} \quad (3)$$

The factor  $c_{\text{cloud}}$  accounts for the fact that the GOME pixels are only partly cloudy, i.e. the true TSCD for the cloudy part of the pixel is slightly higher than the measured TSCD (see Appendix A). The NO<sub>2</sub> TSCD due to anthropogenic emissions  $S_{\text{anthr}}^{\text{NO}_2}$  is estimated in Sect. 4.3.1. The conversion factor  $f$ , accounting for the NO<sub>2</sub> AMF and the NO<sub>x</sub> partitioning, is derived in Sect. 4.2. The factors  $c_{\text{aged}}$  and  $c_{\text{out}}$  account for the impact of aged LNO<sub>x</sub> (Sect. 4.3.2) and the loss of fresh LNO<sub>x</sub> due to transport (Sect. 4.3.3).

Our results will be compared to literature values and errors will be discussed in detail in Sect. 5.

#### 4.1 How many flashes occurred?

The NLDN detects CG flashes over the US with high detection efficiency (DE). But as the measurement stations are bound to the continent, DE decreases with distance from the shore (see Fig. 2a). In the southern part of the convective complex, the estimated DE is below 5%. The clouded area (Fig. 3) reaches further south than the detected cluster of flashes (Fig. 2a), and it is very likely that flashes at the southern end are not detected at all. For pixels 1–9, however, estimated NLDN DE is above 30%. The number of flashes detected in this area is  $4.3 \times 10^4$ . Scaling this number according to the respective DE results in  $9.4 \times 10^4$  flashes.

As NLDN did not report IC flashes in 2000, this number refers to CG flashes only. To derive the total number of flashes, the number of IC flashes has to be estimated. Information on the ratio of IC/CG flash frequencies can be derived from long-term comparisons of NLDN measurements with the satellite born instruments OTD (Optical Transient Detector) and LIS (Lightning Imaging Sensor) detecting both IC and CG flashes (Boccippio et al., 2000). According to the climatology for 16 July–14 October, the IC/CG ratio is 2.7 (1.8–4.0) in the considered region. I.e., the number of CG flashes has to be scaled by a factor of 3.7 (2.8–5.0). Hence, we estimate the total number of flashes on 30 August 2000 in the area covered by the GOME pixels 1–9 to be  $3.49 (2.64\text{--}4.72) \times 10^5$ .

#### 4.2 How to convert NO<sub>2</sub> TSCDs in NO<sub>x</sub> TVCDs?

For our study, the starting point are NO<sub>2</sub> TSCDs ( $S^{\text{NO}_2}$ ) derived from GOME spectra. For the quantification of LNO<sub>x</sub>, these have to be converted into NO<sub>x</sub> TVCDs ( $V^{\text{NO}_x}$ ). The conversion depends on (a) the profile of NO<sub>x</sub>, (b) the NO<sub>x</sub> partitioning  $L := [\text{NO}_2]/[\text{NO}_x]$  and (c) the AMF  $A$ . Both,  $A$  and  $L$  are functions of altitude. The overall (effective) AMF  $A_{\text{eff}}$  is the sum of box AMFs weighted by the NO<sub>2</sub> profile (Eq. B2). The effective partitioning  $L_{\text{eff}}$  can be calculated as a weighted sum of the height dependent NO<sub>x</sub> partitioning

(Eq. B8). These equations are derived in Appendix B. The final conversion is

$$V^{\text{NO}_x} = f \cdot S^{\text{NO}_2} \quad (4)$$

with the conversion factor

$$f = \frac{1}{A_{\text{eff}} \cdot L_{\text{eff}}} \quad (5)$$

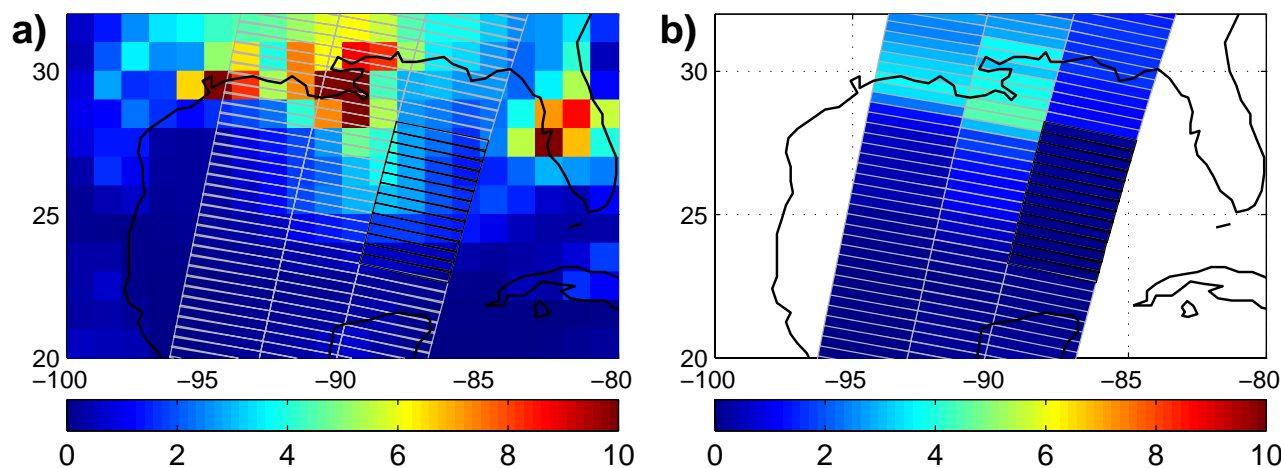
Information on NO<sub>x</sub> profile and partitioning and AMFs is gained from different measurements/model calculations performed for convective cloud conditions.

- (a) The profile of NO<sub>x</sub> is different for different source types. NO<sub>x</sub> from ground sources, here dominated by anthropogenic emissions in the U.S., is mainly located in the boundary layer. Parts of it are lifted due to deep convection, but a large fraction remains in the lowermost kilometers. For anthropogenic NO<sub>x</sub>, we apply the vertical profile as derived from FLEXPART simulations (see Sect. 4.3.1).

The situation is different for NO<sub>x</sub> from lightning: LNO<sub>x</sub> is directly released in the free troposphere, at the very places where updraft takes place. As a result, a large amount of LNO<sub>x</sub> is lifted up in the thunderstorm anvil, resulting in profiles often denoted as “C-shaped” (e.g. Pickering et al., 1998; Fehr et al., 2004). Pickering et al. (1998) used a cloud resolving model to construct vertical profiles of LNO<sub>x</sub> for use in chemical transport models. We apply the LNO<sub>x</sub> profile for tropical marine thunderstorms (Pickering et al., 1998, Table 2). This profile was computed as the mean of three independent case studies. We also apply these three profiles separately (Pickering et al., 1998, Fig. 7) as well as the LNO<sub>x</sub> profile published by Fehr et al. (2004) (modified to a cloud top height of 13 km) to investigate the sensitivity of the effective AMF and the correction factor  $f$  on the NO<sub>x</sub> profile shape.

- (b) The partitioning of NO<sub>x</sub> in NO and NO<sub>2</sub> depends on several parameters like temperature, O<sub>3</sub> concentration, and actinic flux and thus in particular on height. In the upper troposphere, most NO<sub>x</sub> is present as NO due to the low temperatures and high NO<sub>2</sub> photolysis rates. Direct measurements of the NO<sub>x</sub> partitioning for cumulonimbus cloud conditions have been performed by Ridley et al. (1994) during 12 measurement flights over New Mexico. Two of these measurements that took place during active thunderstorms are analyzed in detail by Ridley et al. (1996). Conditions for these thunderstorms are similar to the event on 30 August 2000, as they take place in August on similar latitude and close to local noon. We take the NO<sub>2</sub>/NO<sub>x</sub> ratios given by Ridley et al. (1996, Tables 2 and 4) for the upper core and anvil region and add the missing values below 8 km





**Fig. 5.** (a) Anthropogenic  $\text{NO}_x$  TVCD ( $10^{15}$  molec/ $\text{cm}^2$ ) as modelled with FLEXPART, assuming a  $\text{NO}_x$ -lifetime of 24 h and taking emissions from Frost et al. (2005). The GOME pixel grid is indicated as reference. (b) FLEXPART  $\text{NO}_x$  TVCDs converted to  $\text{NO}_2$  TSCDs (see Sect. 4.3.1) and regridded on GOME grid for direct comparison with Fig. 1.

from Ridley et al. (1994, Figs. 9 and 11). As we cannot exclude that conditions are different for the convective system under consideration, we allow a rather large range of uncertainty of 50% for the  $\text{NO}_2/\text{NO}_x$  ratio for the anvil region.

- (c) As mentioned in Sect. 2.1, the AMF for boundary layer  $\text{NO}_2$  under cloud free conditions is about 1 (Richter and Burrows, 2002). But the considered GOME pixels 1–9 are covered by high and thick thunderstorm clouds that strongly impact the visibility of  $\text{NO}_2$  from GOME. Clouds have two competing effects: On the one hand,  $\text{NO}_2$  below the cloud is shielded. Especially for thick thunderstorm clouds, the boundary layer is effectively invisible from GOME. On the other hand, multiple scattering at the cloud top leads to extended light paths. Thus absorbers at the bright cloud top show an increased visibility from satellite. For the clouded pixels we apply the box AMFs calculated for  $\text{NO}_2$  layers of 1 km thickness by Hild et al. (2002, Fig. 3). These box AMFs have been derived for cumulonimbus clouds and thus specifically match conditions for lightning events.

With these settings for the  $\text{NO}_x$  profile,  $\text{NO}_x$  partitioning (allowing the calculation of the  $\text{NO}_2$  profile) and box AMFs, we get a conversion factor of  $f=12.49$  for anthropogenic  $\text{NO}_x$  and  $f=4.02$  (2.12–7.14) for  $\text{LNO}_x$  for the convective system. The resulting effective AMFs are 0.11 and 0.89 (0.52–1.83), the respective effective  $\text{NO}_x$  partitionings are 0.72 and 0.28 (0.14–0.38).

#### 4.3 What is the impact of transport?

For a quantitative view, the role of transport of  $\text{NO}_x$  has to be considered. Anthropogenic  $\text{NO}_x$ , in particular from the

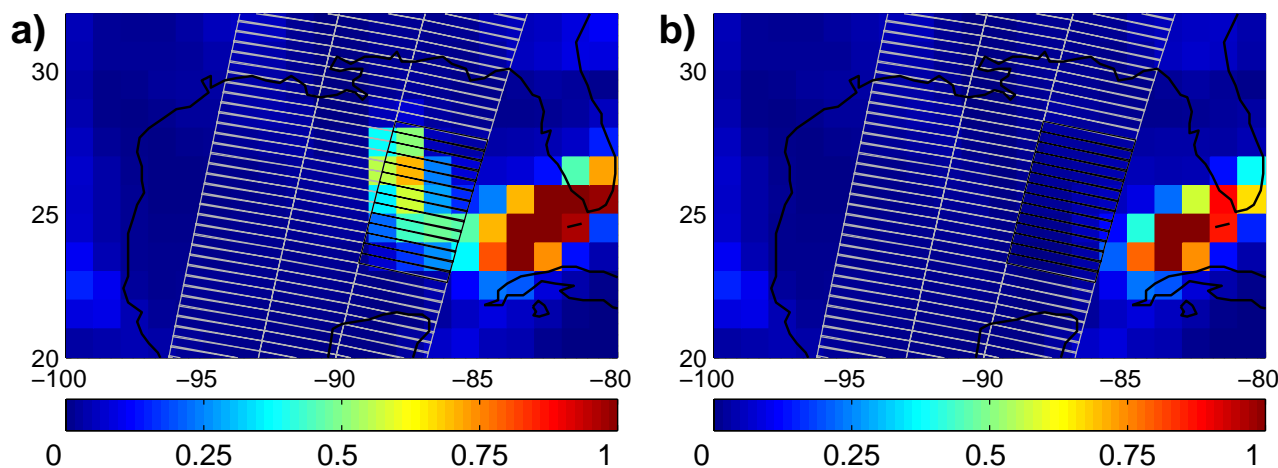
US, may have been transported in the considered region and uplifted (Sect. 4.3.1). Also aged  $\text{LNO}_x$  from lightning events of the previous days may in principle contribute to the detected  $\text{NO}_2$  plume, because of the  $\text{NO}_x$  lifetime of several days in the upper troposphere (Sect. 4.3.2). Finally, the produced  $\text{LNO}_x$  is partly transported outside the considered area (Sect. 4.3.3).

Transport is modelled with the Lagrangian tracer model FLEXPART (see Sect. 2.4). The capability of FLEXPART to track  $\text{NO}_x$  transport events has been demonstrated in several studies (Spichtinger et al., 2001; Wenig et al., 2003; Stohl et al., 2003).

##### 4.3.1 Anthropogenic emissions

The transport of  $\text{NO}_x$  from anthropogenic emissions is simulated with FLEXPART using the inventory for North America compiled by Frost et al. (2006) based on the U.S. EPA NEI-99 (National Emissions Inventory, base year 1999, version 3) (U.S. EPA, 2004a). This inventory was derived at 4-km horizontal resolution from spatial surrogates (U.S. EPA, 2004b) for each U.S. county and Canadian province, and average ozone season day (June through August) county level estimates of on-road, off-road, area, and point sources. The 4-km resolution emissions are also available through a graphics information system interface (Frost and McKeen, 2004). In FLEXPART, emissions were released on the original 4-km grid in regions with high emission densities and from the 300 largest point sources. Grid cells with low emission densities and small point sources were aggregated into lower-resolution grid cells by degrading the resolution in several steps.

The e-folding lifetime for  $\text{NO}_x$  was set to 24 h, which is a rather conservative assumption, as the lifetime of boundary



**Fig. 6.** FLEXPART simulations of  $\text{LNO}_x$  from 27 August on (artificial units). For every flash detected by NLDN, a fixed amount of  $\text{NO}_x$  was released. The  $\text{NO}_x$  lifetime was set to 4 days. Both runs are performed until 30 August 18:00. The GOME pixel grid is added as a reference for both subplots. **(a)** Aged plus fresh  $\text{LNO}_x$ : simulation accounts for all flashes until 30 August, 16:48. **(b)** Aged  $\text{LNO}_x$ : simulation accounts for flashes from 27–29 August only. For the considered GOME pixels 1–9, the fraction of aged  $\text{LNO}_x$  to total  $\text{LNO}_x$  is 11%.

layer  $\text{NO}_x$  is of the order of several hours (e.g. Martin et al., 2003; Beirle et al., 2003). The tracer (a total of 5.5 million particles) was permanently released in the box  $60^\circ\text{--}170^\circ\text{W}$  and  $25^\circ\text{--}75^\circ\text{N}$  over the time period of 25 to 30 August 2000, i.e. 5 days prior the strong lightning event. To consider the vertical transport within this convective system, FLEXPART was run with the implemented convection scheme.

Figure 5a displays the resulting distribution of anthropogenic  $\text{NO}_x$  TVCDs in ECMWF resolution of  $1^\circ \times 1^\circ$ . For direct comparison with GOME measurements (Fig. 1) we regrid the FLEXPART  $\text{NO}_x$  TVCDs to the GOME grid and calculate TSCDs of anthropogenic  $\text{NO}_2$  (Fig. 5b). For this purpose, an effective AMF of 1 (Richter and Burrows, 2002) and an effective  $\text{NO}_2/\text{NO}_x$  ratio of 0.5 were applied for the cloudfree pixels, while for the cloudy pixels the conversion factor  $f$  was taken as 12.49, corresponding to  $A_{\text{eff}}=0.11$  and  $L_{\text{eff}}=0.72$  (see Sect. 4.2).

The resulting  $\text{NO}_2$  TSCDs close to the source regions (mainly New Orleans and Houston) are apparently overestimated in the FLEXPART run. The main reason is probably that the assumed lifetime of 24 h is too long at ground (see above) what can easily explain a factor of 2.

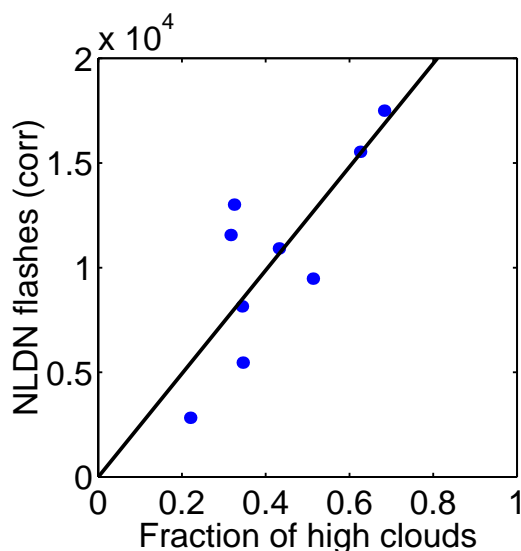
Over the convective complex, on the other hand, the modelled anthropogenic  $\text{NO}_x$  TVCDs are low ( $<2.5 \times 10^{15} \text{ molec/cm}^2$ ). The simulated anthropogenic  $\text{NO}_2$  TSCDs, however, seem to generally underestimate the measured  $\text{NO}_2$  TSCDs of the eastern GOME pixels, even north from  $30^\circ\text{N}$ . Possible reason might be that the emission inventory overestimates hot spots like New Orleans compared to the anthropogenic emissions smoothly distributed along the coast. We thus take the FLEXPART TSCD as lower limit for the actual TSCD due to anthropogenic emissions for pixels 1–9.

To derive an upper limit for the impact of anthropogenic  $\text{NO}_x$  we assume a maximum background level of anthropogenic  $\text{NO}_x$  TVCDs of  $5 \times 10^{15} \text{ molec/cm}^2$  over the considered area, i.e. twice as much as modelled by FLEXPART. This value corresponds to the  $\text{NO}_x$  TVSCD measured by GOME north from the convective complex for cloud free conditions, assuming an AMF of 1 and a  $\text{NO}_2/\text{NO}_x$  ratio of 0.5. Thereby, we ignore the probable decrease of anthropogenic  $\text{NO}_x$  levels southwards from the coast as detected for the western and middle GOME pixels. But even for an *actual* anthropogenic  $\text{NO}_x$  TVCD of  $5 \times 10^{15} \text{ molec/cm}^2$  at the eastern pixels, the expected *measured*  $\text{NO}_2$  TSCD would be only  $0.4 \times 10^{15} \text{ molec/cm}^2$ , since the strong shielding effect of the high and thick cloud cover leads to a conversion factor of 12.49 for anthropogenic  $\text{NO}_2$  (see Sect. 4.2). The upper limit of anthropogenic  $\text{NO}_2$  is thus below 5.8% of the actually detected TSCDs, while the lower limit, taking FLEXPART TSCDs, is half this value, i.e. 3.0%. We take the average, i.e. 4.4% as most probable value for the fraction of anthropogenic  $\text{NO}_x$ . The high  $\text{NO}_2$  TSCDs in Fig. 1 can thus by no means be explained with transport of anthropogenic  $\text{NO}_x$  alone.

#### 4.3.2 Aged $\text{LNO}_x$

Besides anthropogenic  $\text{NO}_x$ , aged  $\text{LNO}_x$  may contribute to the detected  $\text{NO}_2$  plume as well. In fact, NLDN detected several flashes on 29 August west from Florida. However, as discussed in Sect. 3, part of this  $\text{LNO}_x$  is only transported to the southern part of the large complex on 30 August, while  $\text{LNO}_x$  in the northern part is freshly released.

The possible impact of aged  $\text{LNO}_x$  was estimated with FLEXPART using the NLDN flash counts from 27 August



**Fig. 7.** Correlation of NLDN flash counts and the fraction of the GOME pixel covered with high clouds, i.e. having a CTT below 220 K, for pixels 1–9.

on. For every flash, a fixed amount of  $\text{NO}_2$  (artificial units) was released in the respective grid box according to the vertical profile given by Table 2 (tropical marine) in Pickering et al. (1998). The e-folding lifetime was set to 4 days (Jaegle et al., 1998). This run was performed twice: the first run involves all flashes detected till 30 August, 16:48 UTC and stopped at 18:00 UTC (Fig. 6a). The second run simulates the same time period, but stops the release of fresh  $\text{LNO}_x$  on 30 August 0:00 UTC (Fig. 6b). I.e. run 1 shows the combination of aged and fresh  $\text{LNO}_x$ , while run 2 only considers aged  $\text{LNO}_x$  (i.e.  $\text{LNO}_x$  prior to 30 August). The comparison of both runs, i.e. the ratio of run 2 and run 1, allows to assess the fraction of aged  $\text{LNO}_x$ . For the  $\text{LNO}_x$  in the area covered by the GOME pixels 1–9, the fraction of  $\text{LNO}_x$  that is aged is 11%. It has to be noticed that this relative number depends neither on the assumptions about the  $\text{LNO}_x$  released per flash nor on the detection efficiency of NLDN.

#### 4.3.3 Outflow of $\text{LNO}_x$

In the northern part of the convective complex, more than half of the flashes have occurred within 3 h before GOME measurement. But that also means that nearly half of the produced  $\text{LNO}_x$  is older than 3 h. Parts of the  $\text{LNO}_x$  produced in the area of GOME pixels 1–9 are thus transported outside. According to ECMWF windfields, main transport occurs in southerly direction. We estimate the amount of outflow using FLEXPART. Similar to the run performed in Sect. 4.3.2, the distribution of  $\text{LNO}_x$  was modelled by releasing an arbitrary fixed amount of  $\text{LNO}_x$  for every flash, starting from 30 August at 00:00 UTC with infinite lifetime. The resulting distribution

of  $\text{LNO}_x$  is compared to the hypothetical distribution of  $\text{LNO}_x$  in the absence of transport, i.e. the distribution of the flash locations itself. The comparison reveals that 80% of the released  $\text{LNO}_x$  remained inside the GOME pixels 1–9.

The (small) contribution of anthropogenic  $\text{NO}_2$  is subtracted from the measured  $\text{NO}_2$  TSCDs prior to conversion to  $\text{NO}_x$  TVCDs (see Eq. 3). The effects of aged  $\text{LNO}_x$  and outflow of fresh  $\text{LNO}_x$  are accounted for by applying the correction factors  $c_{\text{aged}}=0.89$  and  $c_{\text{out}}=1/0.8=1.25$  that partly balance each other out. The overall uncertainty due to the different effects of transport is taken as 10%.

#### 4.4 What is the total quantity of $\text{LNO}_x$ produced?

The detected  $\text{NO}_2$  plume can be directly assigned to the release of fresh  $\text{LNO}_x$ . Since the northern part of the convective complex is quite young (few hours), chemical decay of the produced  $\text{LNO}_x$  can be neglected due to the long lifetime of  $\text{NO}_x$  in the upper troposphere (Jaegle et al., 1998). Hence the detected  $\text{NO}_2$  TSCDs can be converted directly to the  $\text{LNO}_x$  produced. Using Eq. (3), we derive a mean corrected  $\text{NO}_x$  TVCD of  $3.0 (1.4\text{--}6.0) \times 10^{15} \text{ molec/cm}^2$ .

To derive the total  $\text{LNO}_x$  produced in the convective complex north of  $25^\circ \text{N}$ , these TVCDs have to be integrated across the corresponding area A (see Eq. 2). The area of the convective system for pixels 1–9 is determined by counting the respective PMD subpixels with a HICRU  $\text{CF} > 0.5$ . This results in 79 pixels of  $20 \times 40 \text{ km}^2$  each, i.e.  $63200 \text{ km}^2$  in total. Integration of the  $\text{NO}_x$  TVCDs over the clouded area results in  $3.1 \times 10^7$  moles of  $\text{NO}_x$ , corresponding to  $4.4 \times 10^5 \text{ kg [N]}$ , that are produced by this lightning event.

Combining both numbers, the total  $\text{LNO}_x$  release and the total number of flashes, results in a  $\text{LNO}_x$  production of 90 moles/flash, or 1.3 kg [N]/flash for this particular event. Errors are discussed in detail in Sect. 5.

#### 4.5 The southern part

Combining measured  $\text{NO}_2$  TSCDs and NLDN flash counts, we estimated the production of  $\text{LNO}_x$  for pixels 1–9. This approach is not feasible for pixels 10–14 due to low NLDN DE. Nevertheless, we give a rough estimate of  $\text{LNO}_x$  production for pixels 10–14 using the CTTs measured by GOES.

As lightning is caused by deep convection, flash rates have been found to be closely related to cloud top heights (Price and Rind, 1992). We use the fraction of the GOME pixels covered by clouds with a CTT below 220 K as proxy for high clouds. This temperature corresponds to a cloud top height above 12.5 km. Figure 7 displays the flashes detected by NLDN (scaled with respect to DE) relative to the fraction of high clouds for pixels 1–9. The correlation is  $R=0.79$ , and a linear fit (forced through zero) results in a slope of  $2.47 \times 10^4$ .

We use this relation for a simple estimate of the number of flashes for pixels 10–14, resulting in  $5.4 \times 10^4$  flashes. The inflow of LNO<sub>x</sub> from the northern part and the outflow of LNO<sub>x</sub> from the southern part are difficult to quantify and partly cancel each other out. Effects of transport are thus neglected for our rough quantification of the southern part. Taking the same factors for IC/CG ratio and NO<sub>x</sub> profile and partitioning as above, this results in a LNO<sub>x</sub> production of 143 moles/flash. This number is about 50% higher than that derived for pixels 1–9. The main reason is probably an underestimation of the number of flashes for pixels 10–14, where lightning has taken place for several hours before the ERS-2 overpass (see Fig. 4), while the relation shown in Fig. 7 has been derived for the relatively young convective system covered by pixels 1–9.

## 5 Discussion

In our estimate, resulting in a LNO<sub>x</sub> production of 90 moles/flash, several assumptions on different parameters are involved that are discussed in detail in the following.

The single errors/uncertainties are not gaussian and in particular not symmetric. We thus give a conservative error range for our estimation by considering the extreme values for the factors involved in Eqs. (2, 3), i.e. the number of flashes  $n$  (depending on the assumed IC/CG ratio), the estimated anthropogenic NO<sub>2</sub> SCD, the correction factors  $c_{\text{cloud}}$ ,  $c_{\text{aged}}$  and  $c_{\text{out}}$ , and the conversion factor  $f$ . This results in a range of 32–240 moles/flash, i.e. 0.4–3.4 kg [N]/flash, for LNO<sub>x</sub> production.

The climatological IC/CG ratio is a good first guess, but it has been reported in literature, that individual thunderstorms may show a very high IC/CG ratio of up to 100 (Dye et al., 2000). For such an extreme event, we would have underestimated the actual number of flashes drastically, thus overestimated the LNO<sub>x</sub> production. However, such a scenario is rather unlikely, since the LIS measurement on 30 August, overpassing the detected convective system at 14:07–14:09 UTC, shows no increased number of flashes compared to NLDN.

The transport and uplift of anthropogenic NO<sub>x</sub> has been simulated with FLEXPART using up to date emissions and an improved convection scheme. Nevertheless, we cannot rule out that the upward transport is underestimated by FLEXPART in particular for such a rapidly evolving system. If FLEXPART underestimates the amount of anthropogenic NO<sub>x</sub> uplifted in the anvil, we would also underestimate the AMF for anthropogenic NO<sub>x</sub>. But on the other hand, in this case the NO<sub>x</sub> would be shifted towards NO and the assumed NO/NO<sub>2</sub> ratio for anthropogenic NO<sub>x</sub> would have to be modified. For the extreme scenario of all anthropogenic NO<sub>x</sub> lifted up and mixed homogeneously between 7 and 13 km, leading to an AMF of 0.90, and a NO<sub>2</sub>/NO<sub>x</sub> ratio of 0.26,

anthropogenic NO<sub>x</sub> still could only explain less than 20% of the observed NO<sub>2</sub> TCVDs.

Correction for the partly cloudy GOME pixels and consideration of aged LNO<sub>x</sub> and outflow of fresh LNO<sub>x</sub> only lead to small modifications with negligible errors.

The largest source of uncertainty is the conversion factor  $f$ , depending on NO<sub>x</sub> profile/partitioning and box AMFs.

LNO<sub>x</sub> profiles are taken from different cloud resolving model studies. The ensemble of different profiles allows to study the impact of the LNO<sub>x</sub> profile on  $A_{\text{eff}}$ ,  $L_{\text{eff}}$  and  $f$ . While the different profiles lead to variations in  $A_{\text{eff}}$  up to a factor of nearly 2, the effect on  $L_{\text{eff}}$  is rather small (30%). The uncertainty in  $f$  due to profile variations alone is below 25%.

The NO<sub>x</sub> partitioning in thunderstorm clouds of geolocation, season, and local time similar to the lightning event under consideration was measured by Ridley et al. (1994, 1996). Since we cannot exclude that the actual NO<sub>x</sub> partitioning differs for our case study, a large uncertainty range of 50% for  $L$  in the anvil region is assumed in Sect. 4.2.

Our calculations are based on box AMFs calculated for cumulonimbus clouds by Hild et al. (2002) that are given without errors. Strong deviations from these box AMFs would lead to significant variations in our calculated conversion factor. Hence further effort has to be put on additional box AMF calculations for cumulonimbus conditions with independent models.

Simple extrapolation of our estimated LNO<sub>x</sub> production per flash, assuming a mean flash rate of 44 flashes per second globally (Christian et al., 2003), gives a global LNO<sub>x</sub> production of 1.7 (0.6–4.7) Tg [N]/yr. This number is in good agreement with current literature values, but lower than the often cited number of 5 Tg [N]/yr. However, this particular event is not necessarily representative for global lightning.

Also other LNO<sub>x</sub> estimates using GOME data result in rather low estimates (2.7 (0.8–14) Tg [N]/yr (Beirle et al., 2004c); 1.1–6.4 Tg [N]/yr (Boersma et al., 2004)). However, the remaining uncertainties are yet too high to state the annual LNO<sub>x</sub> production being significantly lower than 5 Tg [N]. Further case studies and statistical evaluations of LNO<sub>x</sub> from satellite measurements are necessary to reduce current uncertainties.

## 6 Conclusions and outlook

In the past, GOME NO<sub>2</sub> data has been used to estimate LNO<sub>x</sub> production by statistical approaches (Beirle et al., 2004a; Boersma et al., 2005). The direct observation of active convective systems, however, has several advantages: Due to deep convection, the NO<sub>x</sub> is lifted up to the cloud top, where satellite measurements are quite sensitive for it. The LNO<sub>x</sub> plume is not yet diluted, hence local NO<sub>x</sub> levels are high. Spatial patterns can be identified and compared to flash rate patterns. And shortly after the LNO<sub>x</sub> production, its



chemical loss is rather negligible, simplifying the calculation of the total NO<sub>x</sub> production.

Within this study, we could identify NO<sub>x</sub> from lightning with GOME data for a particular convective system, matching the GOME observation in space and time. The LNO<sub>x</sub> produced is estimated as 90 (32–240) moles of NO<sub>x</sub> per flash, corresponding to 1.7 (0.6–4.7) Tg [N]/yr globally. This case study impressively illustrates the fundamental feasibility of LNO<sub>x</sub> detection and quantification with satellite NO<sub>2</sub> measurements. Hence space borne spectrometers provide a new and independent approach for the estimation of LNO<sub>x</sub>.

The lightning data from NLDN is limited to the USA, where anthropogenic sources are often interfering with the quantification of LNO<sub>x</sub>. The recently established World Wide Lightning Location Network WWLLN (Lay et al., 2004), as well as the long-range lightning detection networks operated by Vaisala (Pessi et al., 2004; Demetriades et al., 2005), will allow similar case studies to be carried out on a global scale and hence to fully use the potential of satellite data. Since WWLLN is partly sensitive to IC flashes, the uncertainty arising from the IC/CG ratio used is also reduced.

In the future, similar case studies will be performed systematically using the improved spatial resolution of the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartography SCIAMACHY, the Ozone Monitoring Instrument OMI, and the GOME successor GOME-2. Of particular interest will be the analysis of LNO<sub>x</sub> plumes from strong convective systems that are subsequently overpassed by different satellite instruments, e.g. GOME-2 (9:30 a.m.), SCIAMACHY (10:00 a.m.) and OMI (1.45 p.m.). Such scenarios allow the study of LNO<sub>x</sub> plume evolution that holds – if combined with meteorological data – valuable information on LNO<sub>x</sub> profile, NO<sub>x</sub> lifetime, and the LNO<sub>x</sub> produced in thunderstorms.

For the reduction of errors, it would be a milestone to have simultaneous measurements from aircraft (providing NO<sub>x</sub> profile and NO<sub>x</sub> partitioning) and satellite (capturing the whole system at once) for a strong convective complex.

Further efforts will have to be assigned to the modeling of radiative transfer and the calculation of AMFs for fresh LNO<sub>x</sub> in thunderstorm clouds.

## Appendix A

### Correction for partly clouded pixels

The GOME NO<sub>2</sub> measurements are taken with a rather large footprint of 320×40 km<sup>2</sup>, hence they represent mean SCDs of generally inhomogeneous NO<sub>2</sub> distributions and cloud fractions.

To estimate the SCD above the convective complex, we assume the GOME pixels to be divided in a cloud free and a totally clouded part. Let  $f$  be the fraction of the pixel being clouded,  $S_0$  the true SCD for the cloud free part and  $S_c$

the true SCD for the clouded part of the GOME pixel. The total SCD  $S$  measured by GOME is the mean of  $S_0$  and  $S_c$  weighted by the area and the brightness of the cloud free and the clouded part, respectively:

$$S = \frac{I_c \cdot f \cdot S_c + I_0 \cdot (1 - f) \cdot S_0}{I_c \cdot f + I_0 \cdot (1 - f)}, \quad (\text{A1})$$

where  $I_c$  and  $I_0$  are the intensities one would measure for a totally clouded/cloud free scene, respectively. Solving Eq. (A1) for  $S_c$ , the true SCD over the clouded part is

$$S_c = S + \frac{I_0}{I_c} \cdot \frac{1 - f}{f} \cdot (S - S_0). \quad (\text{A2})$$

The ratio  $I_0/I_c$  is gained by comparing the maximum and minimum intensities of the PMD subpixels in the blue spectral range, resulting in  $I_c = 7 \times I_0$ .  $S_0$  is estimated taking the SCD of the respective neighboring, cloud free center GOME pixel. With these numbers,  $S_c$  is on average higher than  $S$  by 5% for pixels 1–9. The measured SCD  $S$  has thus to be corrected by a factor of 1.05. The extreme cases of  $S_{0,\min}=0$  and  $S_{0,\max}=3 \times 10^{15}$  (i.e. the maximum SCD of the center GOME pixels) lead to a range of 1.04–1.10 for the correction factor  $c_{\text{cloud}}$ .

## Appendix B

### Effective AMF and NO<sub>2</sub>/NO<sub>x</sub> partitioning

The troposphere is divided into  $n$  layers. Let  $a_i$  be the box AMF,  $s_i^{\text{NO}_2}$  and  $v_i^{\text{NO}_2}$  the slant/vertical column of NO<sub>2</sub> and  $v_i^{\text{NO}_x}$  the vertical column of NO<sub>x</sub> of the  $i$ th layer. Furthermore,  $l_i$  is the ratio  $v_i^{\text{NO}_2}/v_i^{\text{NO}_x}$ , and  $p_i^{\text{NO}_x}$  and  $p_i^{\text{NO}_2}$  represent the normalized profiles of NO<sub>x</sub> and NO<sub>2</sub>, respectively.

The total TSCD of NO<sub>2</sub>,  $S^{\text{NO}_2}$ , can be expressed as

$$S^{\text{NO}_2} = \sum s_i^{\text{NO}_2} = \sum v_i^{\text{NO}_2} \cdot a_i = V^{\text{NO}_2} \cdot \sum p_i^{\text{NO}_2} \cdot a_i \quad (\text{B1})$$

The latter is the sum of the box AMFs weighted with the vertical NO<sub>2</sub> profile, resulting in the effective AMF:

$$A_{\text{eff}} = \sum p_i^{\text{NO}_2} \cdot a_i \quad (\text{B2})$$

On the other hand,  $S$  can be expressed as

$$\begin{aligned} S^{\text{NO}_2} &= \sum v_i^{\text{NO}_2} \cdot a_i = \sum l_i \cdot v_i^{\text{NO}_x} \cdot a_i \\ &= V^{\text{NO}_x} \cdot \sum p_i^{\text{NO}_x} \cdot l_i \cdot a_i \end{aligned} \quad (\text{B3})$$

$$\Rightarrow V^{\text{NO}_x} = \frac{S^{\text{NO}_2}}{\sum p_i^{\text{NO}_x} \cdot l_i \cdot a_i} = f \cdot S^{\text{NO}_2} \quad (\text{B4})$$

with

$$f = \frac{1}{\sum p_i^{\text{NO}_x} \cdot l_i \cdot a_i}. \quad (\text{B5})$$

Alternatively,  $f$  can be written as

$$f \stackrel{(B4)}{=} \frac{V^{\text{NO}_x}}{S^{\text{NO}_2}} \stackrel{(B1)}{=} \frac{V^{\text{NO}_x}}{V^{\text{NO}_2} \cdot A_{\text{eff}}} = \frac{1}{\frac{V^{\text{NO}_2}}{V^{\text{NO}_x}} \cdot A_{\text{eff}}} = \frac{1}{L_{\text{eff}} \cdot A_{\text{eff}}} \quad (\text{B6})$$

with

$$L_{\text{eff}} := \frac{V^{\text{NO}_2}}{V^{\text{NO}_x}}. \quad (\text{B7})$$

Hence,  $L_{\text{eff}}$  can be calculated as

$$L_{\text{eff}} \stackrel{(B6)}{=} \frac{1}{f \cdot A_{\text{eff}}} \stackrel{(B5)}{=} \frac{\sum p_i^{\text{NO}_x} \cdot l_i \cdot a_i}{A_{\text{eff}}} \stackrel{(B2)}{=} \frac{\sum p_i^{\text{NO}_x} \cdot a_i \cdot l_i}{\sum p_i^{\text{NO}_2} \cdot a_i}. \quad (\text{B8})$$

This can be interpreted as weighted sum of the NO<sub>x</sub> partitionings of the single layers.

$A_{\text{eff}}$  and  $L_{\text{eff}}$  are calculated in Sect. 4.2. for illustration. For the final conversion (Eq. 3) and the estimation of errors, Eq. (B5) is used for the calculation of  $f$ , since modifications of  $p_i^{\text{NO}_x}$ ,  $l_i$  and  $a_i$  affect both  $A_{\text{eff}}$  and  $L_{\text{eff}}$ , but effects partly cancel out in  $f$ .

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