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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Lightning-produced NO_x over Brazil during TROCCINOX: Airborne measurements in tropical and subtropical thunderstorms and the importance of mesoscale convective systems" *by* H. Huntrieser et al.

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- We thank Reviewer #2 for the helpful comments.

General points: As an experimentalist I would prefer a more detailed discussion of the ozone measurements in clouds to get a clearer view of the potential reason for the artificial enhancements.

- The problem with the ozone measurement in clouds is not discussed here in detail since it is a topic for a separate paper (which is planned to be published in near future as indicated in the present paper after further studies in the laboratory). However, we

have included some additional information in Sect. 2.1 and 3.3.1, and furthermore complete O3 data in Fig. 6a, as one example of the artificial O3 measurements.

- Sect. 2.1 complemented: “During several anvil penetrations, strongly enhanced O3 signals in the range ˜100-300 nmol mol-1 were measured. In Sect. 3.3.1 measurements from such a flight on 28 February 2004 are shown as an example and discussed in detail. Influence by entrainment of O3-rich air from the tropopause region or lower stratosphere was excluded as explanation for the O3 enhancement, since CO was also enhanced in most of the anvils. Furthermore, the O3 mixing ratios are too high to be explained by photochemical production or by vertical transport from the BL, where in general lower mixing ratios between ˜20-50 nmol mol-1 were measured. These pronounced O3 enhancements were always connected to penetrations in the anvil outflow and never outside. From the discussion of the flight in Sect. 3.3.1 we consider the high O3 signals to be artefacts and excluded such elevated signals from the data set. Similar ozone-rich transients have been observed during CARIBIC and MOZAIC flights performed mainly during in-cloud sections in the tropics (Zahn et al., 2002). Recently, Ridley et al. (2006) discussed several possible reasons for observed spikes in O3 (and NO) signals during CRYSTAL-FACE and other airborne field experiments. They suggested that the observed short-term spikes (˜1 s) during CRYSTAL-FACE were measurement artefacts caused by discharges on the aircraft fuselage or inlets. However, during TROCCINOX the artefact O3 signals were much broader and covered the entire duration of the anvil penetration (˜20-40 km, ˜several minutes). In addition, measurements during the TROCCINOX field campaign in 2005 with an additional O3 instrument (with different inlet design) indicate that discharges on inlets were probably not the reason for the artefact O3 signals. Meyer et al. (1991) have reported on a water vapour interference for ground-based measurements of O3 for some of the instruments using the UV absorption technique (same technique as used on the Falcon) which may explain our observations. The interference can be significant when the humidity is varying rapidly as during anvil penetrations. The reason for the interference, as suggested by Meyer et al., could be humidity-induced variations of the extinction of UV light by the

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windows of the two optical cells in the instrument. First tests in the laboratory with the Falcon O3 instrument indicate an interference due to abrupt changing water vapour mixing ratios in accordance with Meyer et al.˜(1991). However, further laboratory studies are needed to study the interference in more detail and the results will be reported in a separate paper.

- New section added to Sect. 3.3.1: “In addition, O3 measurements are shown in Fig. 6a. However, the mixing ratios observed during the three anvil penetrations were very variable. During the first penetration, O3 mixing ratios slightly decreased down to ˜50 nmol mol-1, which can be explained by transport from the BL. However, during the second and third penetration of the same anvil at lower altitudes, O3 mixing ratios strongly increased to values above 200 nmol mol-1. We believe that these elevated O3 signals are artificial, as discussed in Sect. 2.1. During the first and second anvil penetration we would expect about the same O3 mixing ratios, since the increases in CO and NOx mixing ratios are similar during these penetrations.”

E.g. is the ozone enhancement correlated with NO (lightning source) or CO (boundary layer transport)? Is it observed only in clouds or do you also observe unexpected enhancements outside from clouds?

- We have observed the O3 enhancement during some of the penetrations in the anvil outflow and never outside. NO mixing ratios were also enhanced (due to lightning-production and transport from the BL). During the penetrations in Fig. 6a-d, CO mixing ratios generally increased (e.g. 28 February 2004), however also occasionally decreased (e.g. 18 February 2005) depending on background and BL mixing ratios. Though, in both cases it was clear that air masses from the BL were transported upward.

Personally, I believe that the question whether O3 is produced in lightning strikes is not settled yet and your data could be an important contribution. Along the same lines, I would appreciate to see the ozone data added to Figure 1. I would also prefer to

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differentiate in this figure between in-cloud and cloud free observations.

- We have added O3 data to Fig. 1, however the strong O3 enhancements that we believe are artificial were excluded. These were especially pronounced during the penetrations of an active isolated thunderstorm on 28 February 2004, but also observed on 4 and 18 February 2005 (Fig. 6).

On transport modelling: You use backward trajectories to identify the air mass origin in the convective outflow. How reliable are back-trajectory calculations that pass through convection?

- As mentioned in Sect. 2.3, the convection scheme by Emanuel and Zivcovizc-Rothman has recently been implemented in FLEXPART to account for sub-gridscale moist convective transport . A new sentence was added (page 2586, line 19): "In vicinity of thunderstorms the determination of the air mass origin with FLEXPART is not always reliable due to the small extension of many of the systems and the low background wind velocities, especially in tropical air masses. However, if the extension of the convective system is large enough (as in the MCS case presented here) and wind velocities in the UT are pronounced (as in vicinity of the subtropical jet stream), it should be resolved in the ECMWF data and therefore accounted for by FLEXPART."

Minor points:

Abstract, page 2562, line 18: Please specify whether the cited enhancements are absolute mixing ratios or enhancements above a background.

- Changed to "absolute mixing ratios". In addition, page 2562, line 10-11 was changed to: "During thunderstorm anvil penetrations, typically at 20-40 km horizontal scales, NOx mixing ratios were distinctly enhanced and the absolute mixing ratios varied between 0.2-1.6 nmol mol-1 on average."

Section 3.3.2, page 2575, line 5: Is the decrease in the maximum LNOx mixing ratio from 2 to 1 ppbv deduced from a model forecast, or where does this number stem

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from?

- Changed to “LNOx mixing ratio in the model forecast”.

- In addition, further references were added to the reference list and cited in the text:

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Salio, P., Nicolini, M., and Zipser E. J.: Mesoscale convective systems over southeastern South America and their relationship with the South American low-level jet, Mon. Wea. Rev., 135, 1290-1309, 2007.

Sato, M., and Fukunishi, H: Global sprite occurrence locations and rates derived from triangulation of transient Schumann resonance events, Geophys. Res. Lett., 30, 1859, doi:10.1029/2003GL017291, 2003.

Sauvage, B., Martin, R. V., van Donkelaar, A., Liu, X., Chance, K., Jaegle, L., Palmer, P. I., Wu, S., and Fu, T.-M.: Remote sensed and in situ constraints on processes affecting tropical tropospheric ozone, Atmos. Chem. Phys., 7, 815-838, 2007.

Thomas, J. N., Taylor, M. J., Pautet, D., et al.: A very active sprite-producing storm observed over Argentina, EOS, Transactions, AGU, Vol. 88, Nr. 10, 117-119, 2007.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 2561, 2007.