Atmos. Chem. Phys. Discuss., 4, S569–S573, 2004 www.atmos-chem-phys.org/acpd/4/S569/ © European Geosciences Union 2004



ACPD

4, S569–S573, 2004

Interactive Comment

# *Interactive comment on* "Tracing troposphere-to-stratosphere transport above a mid-latitude deep convective system" by M. I. Hegglin et al.

### M. I. Hegglin et al.

Received and published: 21 April 2004

#### Author response to referee comment #2

We thank the reviewer for his or her helpful comments.

Main comment: The referee asks to clarify the different mixing processes and how initial trace gas compositions of mixed air masses have looked like to explain the unusual  $NO_y/O_3$  relationships in flight segments I and II.

Reply: We fully agree that the mixing line in segment I must be caused by mixing



**Print Version** 

Interactive Discussion

**Discussion Paper** 

between a 'normal' tropospheric air mass with a stratospheric one with unusually high NO<sub>v</sub>- and quite low  $O_3$  mixing ratios. It is indeed the stratospheric reservoir which is unusual, not the tropospheric one. But this is exactly what we were trying to say by noting (see first sentence of last section of page 12): 'On this flight segment stratospheric NO<sub>v</sub> mixing ratios between and after the tropospheric filaments were much higher than values expected from the  $NO_v - O_3$  correlation given by Murphy et al. (1993)...'. As detailed below we believe that these unusually high NO<sub>v</sub> values in the stratosphere themselves are of recent tropospheric origin and are not representative for the lowermost stratospheric air. Some references indicated by the referee (Fischer et al., 1997; Bregman et al., 1995) present examples where high NO<sub> $\nu$ </sub>/O<sub>3</sub> ratios were attributed to renitrification. Yet, renitrification is very unlikely in our case because the flight took place in November. The referee argues that in November the lowermost stratosphere is strongly influenced by tropospheric air that crossed the tropopause during summer and autumn along isentropic surfaces as suggested by Ray et al. (1999) and that therefore the  $NO_y/O_3$  relationship might be significantly different from the stratospheric one found during other seasons. This would imply that large parts of the LMS in autumn would be disturbed due to this mixing-in of tropospheric air over the previous weeks and months. However, this is not in agreement with our observations. Hoor et al. (2004, submitted) present a new study in which they conclude that in summer isentropic mixing is confined to a tropopause following mixing layer with a vertical extent of about  $\Delta \Theta$ =30 K from the tropopause and a decreasing tropospheric influence beyond this layer. The here presented measurements however were taken in an altitude range between  $\Delta \Theta$ =35 and 55 K from the tropopause. Also, in contrast to flight I the NO<sub>v</sub>-O<sub>3</sub> correlations obtained during two subsequent flights on the following day were well within the range of expected correlations as formerly found by Murphy et al. (1993) and Fahey et al. (1996) with values between 0.003 and 0.004 (this important statement is now included in the manuscript).

In our manuscript we were talking about two temporally separated processes transport-

4, S569–S573, 2004

Interactive Comment

Full Screen / Esc

**Print Version** 

Interactive Discussion

**Discussion Paper** 

ing tropospheric air into the lowermost stratosphere. Obviously there was considerable confusion about this because we did not present this fact in a concise manner. We have therefore reworked and expanded the discussion of flight segment I (Sect. 5.1) including a new figure (now Fig. 5) which in the previous version was only mentioned but 'not shown'. The figure suggests that the stratospheric air in segment I was not a pure stratospheric air mass but had remained in the vicinity of the tropopause during at least the previous 10 days, and a majority of the corresponding trajectories had even experienced troposphere-to-stratosphere exchange some 5 to 10 days before the measurements. We argue that  $NO_v$ -rich tropospheric air (of about 4-5 ppbv of  $NO_v$ ) has been mixed into the lowermost stratosphere resulting in a highly perturbed stratospheric air mass. In a second mixing event, which generated the marked tropospheric filaments observed in this flight segment, NO<sub>v</sub>-poor (or rather normal) tropospheric air (of less than 1 ppbv) was mixed with this perturbed stratospheric air, resulting in an unusually steep correlation slope. The origin of the enhanced NO<sub>v</sub> mixing ratios mixed in through the first process, however, could not be definitely identified. We were not able, for instance, to trace back the trajectories to a single convective event or frontal activity which could have delivered the high NO<sub>v</sub> mixing ratios through transport from the polluted boundary layer or lightning activity. Rather, the individual trajectories had passed several different systems including a cold front located over the Canadian east coast on the third November.

The referee raises the question whether the observed data in segment II could not be interpreted as a transition zone between the air masses with a steep correlation slope  $NO_y/O_3$  of 0.013 in segment I and the lower slope of 0.003 in segment III. We can indeed not exclude this possibility if only considering the  $NO_y/O_3$  relationship. However, there are two arguments against this hypothesis. First, the  $H_2O/O_3$  correlation shows a bulge of elevated  $H_2O$  in segment II (see new Fig. 3b) which can not be understood simply by mixing between air in segments I and III. Second, the trajectory calculations suggest that the air in segment II had experienced a significantly different history. In contrast to segment I air parcels in segment II were not influenced by TST (at least not

# ACPD

4, S569–S573, 2004

Interactive Comment

Full Screen / Esc

**Print Version** 

Interactive Discussion

**Discussion Paper** 

by TST resolved by the ECMWF model) (see new Fig. 5), they were of polar rather than mid-latitudinal origin (Fig. 7) and had descended from high PV values (Fig. 2d) suggesting that this air had a more marked stratospheric character in agreement with the high  $O_3$  mixing ratios of 400-500 ppbv. We therefore argue that recent convective influence is the most plausible cause for the elevated mixing ratios of both  $NO_y$  and  $H_2O$  in segment II as derived by our analysis presented in the paper.

Please refer to the revised Section 5.1 for more details.

Specific comments:

Fig.10: Indeed the air parcels had traveled a long distance during the past 24 hours. However, flight segment 2 was close to the center of the streamer where wind speeds were only about 20 m/s (rather than the 50 m/s estimated by the referee). Flight segment 2 was actually close to a marked shear line where wind directions dramatically changed from northeasterly winds to the west of the shear line to southwesterly winds to the east. Figure 10 (now 11) shows that air parcels located to the east of the shear line had followed a southern path leading over regions of strong convective and light-ning activity. The convective influence plot (Fig. 10, now 12) shows this influence in an integral way for the time of the flight suggesting that the flight path was very close to the convectively influenced region. The fact that the measured wind direction in large parts of segment II had a stronger easterly component than the ECMWF analyzed wind suggests that the convectively influenced air mass (as derived from trajectories based on ECMWF winds) in reality would have been advected even more closely to the flight path.

Fig. 11: The suggestion has been adopted in a new Figure.

Additional references:

Bregman, A., van Velthoven, P.F.J., Wienhold, F., Fischer, H., Zenker, T., Waibel,

ACPD

4, S569–S573, 2004

Interactive Comment

Full Screen / Esc

**Print Version** 

Interactive Discussion

**Discussion Paper** 

A., Frenzel, A., Arnold, F., Harris, G.W., Bolder, M.J.A., and Lelieveld, J.: Aircraft measurements of  $O_3$ , HNO<sub>3</sub>, and  $N_2O$  in the Arctic lower stratosphere during the Stratosphere-Troposphere Experiment by Aircraft Measurements (STREAM) 1, *J. Geophys. Res., 100 (D6),* 11245-11260, 1995.

Fahey, D.W., Donnelly, S.G., Keim, E.R., Gao, R.S., Wamsley, R.C., Del Negro, L.A., Woodbridge, E.L., Proffitt, M.H., Rosenlof, K.H., Ko, M. K. W., Weisenstein, D. K., Scott, C.J., Nevison, C., Solomon, S., and Chan, C.J.: In situ observations of  $NO_y$ ,  $O_3$  and the  $NO_y/O_3$  ratio in the lower stratosphere, *Geophys. Res. Lett.*, 23, 1653-1656, 1996.

Fischer, H., Waibel, A., Welling, M., Wienhold, F.-G., Zenker, T., Crutzen, P. J., Arnold, F., Bürger, U., Schneider, J., Bregman, A., Lelieveld, J., and Siegmund, P. C.: Observations of high concentrations of total reactive nitrogen ( $NO_y$ ) and nitric acid ( $HNO_3$ ) in the lower Arctic stratosphere during the Stratosphere-Troposphere Experiment by Aircraft Measurements (STREAM) II campaign in February 1995, *J. Geophys. Res.*, *102*, 23559-23571, 1997.

Hoor, P., Gurk, C., Brunner, D., Hegglin, M.I., Wernli, H., and Fischer, H.: Seasonality and extent of extratropcial TST derived from in-situ CO measurements during SPURT, *Atmos. Chem. Phys. Discuss.*, *4*, 1691-1726, 2004.

Murphy, D., Fahey, D., Proffitt, M., Liu, S., Chan, K., Eubank, C., Kawa, S., and Kelly, K.: Reactive nitrogen and its correlation with ozone in the lower stratosphere and upper troposphere, *J. Geophys. Res.*, *98, D5*, 8751-8773, 1993.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 169, 2004.

# ACPD

4, S569–S573, 2004

Interactive Comment

Full Screen / Esc

**Print Version** 

Interactive Discussion

**Discussion Paper**