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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurements of NO, NO_y, N_2O, and O_3 during SPURT: implications for transport and chemistry in the lowermost stratosphere" *by* M. I. Hegglin et al.

M. I. Hegglin et al.

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We thank the reviewer for his or her helpful comments and suggestions, including stylistic advices.

Comment 1: The referee's main comment is that the conclusions are too soft and that they rely on a limited data set. He/She asks for an over-arching statement which says that further research (e.g. model studies) is required to proof the interpretation of the data set.

Reply 1: We elaborated on the presentation of the conclusions and added a corre-

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sponding statement: *However, the conclusions drawn in this paper apply strictly only to the UT/LMS in the longitude sector from - 20°E to 30°E in the Northern hemisphere. Finally, to confirm the presented conclusions, further investigations should be made by applying the presented evaluation methods to larger data sets. More measurements, preferentially over an even broader range of* ϕ_e *and* Θ *are needed to better characterize the latitudinal gradients of these tracers. Future aircraft observations should be conducted at different longitudes and extended into the southern hemisphere in order to enhance global knowledge of tracer distributions in the UT/LMS.*

Comment 2: Suggestion to consult Ray et al. (1999) in order to improve the discussion what air moves into and out of the LMS and when.

Reply 2: We acknowledge the value of the approach and evaluation of Ray et al. (1999) and included a discussion about their findings in the text. Nevertheless, we note that the conclusions of Ray et al. (1999) were based on two balloon soundings at 34.5°N, one at high-latitude at 65°N, and two in the tropics at 7°S. The conclusions were therefore based on an even more limited data set than the SPURT measurements, latter offering extended knowledge of tracer distributions in the LMS in both respects, the latitudinal and temporal range covered. Ray et al. (1999) did for example not yet consider the rapid transport of young air in the tropically controlled transition region in their mass balance calculation (cf. Hoor et al., 2005).

Comment/reply 3: The intention of the tracer tracer correlation section is to learn something about the origin of the 'background' LMS air. To do so, one has to focus on tracer tracer correlations which are less influenced by STE processes. This is given for the O_3/N_2O correlation. $\Delta NO_y/\Delta N_2O$ due to local sources and sinks shows much higher variability and is used as additional evidence only. Showing the scatter plots would be instructive for information about tropospheric influence and would trigger the discussion of NO_y sources. This discussions goes beyond the scope of this paper and we refer interested readers to Hegglin (2004). See also answer to a comment by K.

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Pfeilsticker.
We agree with the reviewers second comment, that equivalent latitude - potential temperature presentation of NO_x is less meaningful than for longer lived tracers such as O_3 or N_2O. Nevertheless, it has the advantage to separate tropospheric from stratospheric air masses as opposed to regular latitude-altitude presentations. The lifetime of NO_x further is around 11 days in the tropopause region (Jaegé et al., 1998), which is a longer timescale than the typical meridional transport timescale of Rossby waves. 11 days is also long enough that NO_x can have a substantial effect on the O_3. We consider therefore both the NO_x profiles and the equivalent latitude plots to be informative and important especially for CTM modelers or for studies which aim to address the influence of aircraft NO_x emissions on ozone chemistry in this region. We added further the a figure and discussion of vertical profiles of NO_x/NO_y since we agree that this is a very instructive value too (please see revised manuscript).

Comment 4: We further specified the used model run of the Grooss et al. study: *'It is based on the results by Grooss et al. (1998) using the Mainz two-dimensional photochemical model for upper tropospheric conditions assuming CO and HNO_3 mixing ratios of 60 ppbv and 0.2 ppbv, respectively.'*

Comment 5: We used the results by Grooss et al. (1998) that different background O_3 mixing ratios change the NO^{crit} values in order to motivate the need for an approximation of a NO^{crit} valid in the LMS. The paper did not intend to oppose the results in Grooss et al. (1998) to the ones presented in our study, nor to show a major disagreement. Their result that CO does not affect NO^{crit} is correct for the troposphere, where their study was focusing on. However, along with the discussion of two new figures (see reply to comment 7) the text has been changed to resolve this issue.

Comment 6: We acknowledged the source of CO. Please see revised Sect. 4.4.

Comment 7: In order to clarify this point, we produced two different figures 12 and 13 which separate the twofold and (admittedly) unclear information given in the original

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figure 12. Figure 12 now shows excess NO calculated for each single measurement point as a function of $\Delta\Theta$ relative to the tropopause. The figure therefore shows how much NO is available in surplus to the NO^{crit}. It further indicates up to which height the sampled air masses are in the O_3 production regime. This presentation is in our eyes more informative than just a scatter plot between NO and NO^{crit}. Figure 13 finally shows mean vertical profiles of NO^{crit} for each season. It demonstrates how the seasonally changing background composition of the LMS (as given in Sect. 'Tracer-tracer correlations') has a strong impact on calculated NO^{crit} and therefore on ozone chemistry itself.

Comment 8: Text changed: *'The slopes of the NO_y to N_2O correlation show changes by 20% between October 2002 (slope = -0.102) and April 2003 (slope = -0.082). This result is significantly higher than the maximum change in slope of 6% derived from ER2-data above 400 K during the ASHOE/MAESA and the STRAT campaigns (Keim et al., 1997). The corresponding slopes were -0.069 in April 1994 and -0.073 in January 1996. The difference of the two observations may be explained by the different altitude range of the observations, the inter-annual variability in the strength of the mean meridional circulation, or by tropospheric influence which has a higher impact on the NO_y to N_2O slopes than on the O_3 to N_2O slopes.'*

Comment 9: This general statement was not concise and we removed the text on page 8660 L21.

Comment 10: The calculations were updated by using the JPL recommendations for chemical reaction rates.

Comment 11: done.

Comment 12: Text improved.

Comment 13: Seasons accordingly defined.

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