**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Measurements of NO, NO$_y$, N$_2$O, 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 reviewer T. Birner for his careful reading and valuable suggestions.

Mayor comments:

Comment 1: Number of plots does not fit the number of outcome. The fields are noisy and don't show strong differences within a given season.

Reply: Seasonal means of the observed tracers are given in the SPURT overview paper by Engel et al. (2005). It would be scientifically incorrect to reproduce the same

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Discussion Paper

EGU

figures in two different papers. We also only partly agree with the reviewer's concern. Showing tracer mean distributions for each month does add valuable information e.g. for chemistry climate modelers who aim to validate their models against observations. One example for such a validation is given in Hegglin (2004). The month to month comparison of the SPURT data with simulated O_3 and NO_y tracer distributions in the chemistry transport model MOZART (version 2.4) revealed a shift in maximum tracer distributions of 2-3 months. The problem could be traced back to a wrong initialization of the stratospheric tracer fields and could be fixed easily. However, we improved the discussion in Sect. 4.1 in order to address the concern that the number of plots does not fit the number of outcome.

Comment 2: Use of ECMWF fields for the mapping of the data may introduce uncertainty in the observed structures.

Reply: We would like to draw the attention of the referee to the fact that we used measured potential temperature (p and T) when plotting the data in the equivalent latitude - potential temperature framework or in the vertical profiles. Only the calculation of equivalent latitude was based on the ECMWF fields. We agree that the use of ECMWF because of its limited resolution leads to a limitation in accounting for small scale structures in the tracer fields. It therefore leads to a potential 'smearing out' of the real gradients at the tropopause as stated in the new version of the manuscript. Nevertheless, the use of the equivalent latitude - potential temperature framework represents a major improvement of the representation of tracer distributions in the LMS compared to the use of standard geometrical coordinates. For the presentation of the vertical profiles relative to the height of the tropopause finally, the use of ECMWF PV is the only way to determine the location of the tropopause during SPURT since we lack measurements of a temperature profiler.

Comment 3: Wrong use of the potential vorticity concept.

Reply: The intention of the PV vertical profiles was to have a proxy for static stability,

since we lacked static stability data. We were not aware of this not being consistent with the PV concept and removed the figure.

Comment 4: Demand for clearer conclusions and improved consistency in the paper's structure.

Reply: We improved the paper's structure by addressing different general and minor comments see below. We further moved the description of the NO_x calculation from the Result-section to a separate paragraph in Section 3 where all methods are described. We also renamed the last section to 'Summary and conclusions' and improved the conclusions drawn from the presented evaluations (please see revised manuscript).

Comment 5: Lack of citation of Pan et al. (2004) paper.

Reply: Reference included. A thorough discussion of the main result of the paper, namely the difference between the use of the dynamical versus the thermal tropopause is, however, beyond the scope of this paper. As stated above, the use of low resolution ECMWF fields for the calculation of the dynamical and the thermal tropopause leads to a smearing out of the real tracer gradients at the tropopause leading to uncertainties in both cases.

Minor comments are incorporated into the revised manuscript as follows:

General

Comment 1: Limitation of the number of flights and covered geographical region.

Reply: Please see reply to comment 1, Referee # 2 who shared the concern.

Comment 2: Suggests emphasizing equivalent latitude being a PV coordinate.

Reply: We added the following text to Sect. 3.1: *Since equivalent latitude is a PV coordinate, its calculation relies on monotonically increasing PV values. A recent study*

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Interactive Discussion

Discussion Paper

EGU

by Birner (2005) found, however, that PV in the UT/LMS exhibits only small meridional gradients away from the tropopause. An interpretation regarding geographical dependencies of tracers based on the equivalent latitude framework should therefore be treated carefully in regions away from the tropopause.

Comment 3: Suggests comparison between median and mean values.

Reply: The difference between mean and median tracer mixing ratios are rather small and the discussion of it does not add important information to the evaluations made in the paper. It is well known in the literature that medians are more robust against outliers. We added the following text to make an overall statement: *Medians have been used since they are less sensitive to extreme values in a data set than mean values. For the longer lived species N_2O and O_3, mean and median distributions show no significant differences. NO_y and NO_x, however, due to localized sources and sinks and especially in the tropopause region show mean values which are often up to 30% and 100% higher, respectively.*

Specific

P8650 L2: Text has been changed to:*'The measurements cover all seasons between November 2001 and July 2003. They span a broad band of latitudes from 30° N to 75° N and a potential temperature range from 290 to 380 K.'*

P8650 L15: Text has been changed to:*'Concurrent large gradients in static stability in the vertical direction, and of PV in the meridional direction, suggest the presence of a mixing barrier.'*

P8651 L7: done

P8651 L13: Text changed.

P8652 L5: done

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

P8652 L6: Text has been changed to:*'...and extratropical stratosphere-troposphere exchange (STE) (Holton et al., 1995).'*

P8652 L7: done

P8652 L13/L14: The text has been changed to: *'Vertical profiles of CO measured during SPURT have revealed the existence of a mixing layer 20 to 30 K above the local dynamical tropopause during all seasons (Hoor et al. 2004). The chemical composition of this mixing layer exhibits characteristics of both the troposphere and the stratosphere.'*

P8652 L24: See next comment.

P8653 outline: Text has been shortened from page 8652 line 24 on: *'The data reveal new information about the seasonal distributions of the measured trace gas species and represent a valuable contribution to the existing climatology between the tropopause and $\Theta=380$ K (Θ–potential temperature). This paper is part of the SPURT special issue in ACP where other tracer data together with tracer-specific evaluations are presented. An overview of the SPURT project is provided by Engel et al. (2005).*
A short description of the measurement systems used, and their performance and overall accuracies is given in Sect. 2. Section 3 describes the evaluation methods. Section 4.1 presents median tracer distributions in equivalent latitude - potential temperature coordinates most suited to be used as diagnostics for the validation of chemical transport models (CTMs) and global circulation models (GCMs). Section 4.2 discusses the role of the tropopause in shaping tracer distributions. Section 4.3 shows tracer-tracer correlations which give insight into the seasonal variability of the Brewer Dobson circulation. Finally, in Sect. 4.4, we present a new calculation of the NO mixing ratio at which net O_3 production goes from positive to negative (NO^{crit}), which takes both the tropospheric and stratospheric character of the LMS into account.'
The description of the subsections of the results were shortened but not removed since they allow the reader to find a certain topic faster (the paper is quite long and addresses

issues interesting for both dynamicists and chemists).

p8654 L15: We added a specific statement: *A possible error in accounting for the NO_y artifact signal may introduce an offset of maximal 20 to 50 pptv to the data.* We note, that according to newer evaluations the NO_y measurement error changed (cf. Paetz et al. (2005), ACPD, submitted) and the text had to be corrected accordingly: *For the NO_y-channel the accuracy contains also the uncertainties in the conversion efficiency for different species ($\pm 10\%$), an additional calibration gas uncertainty of ($\pm 3.7\%$), the reproducibility of the conversion efficiency experiments ($\pm 3\%$) and an experimentally determined pressure correction ($\pm 4\%$). The overall accuracies of the NO_y, NO and O_3 then are $\pm (0.126 \cdot [NO_y] + 11$ pptv), $\pm (0.045 \cdot [NO] + 9$ pptv), and $\pm (0.05 \cdot [O_3] + 149$ pptv), respectively. These specifications refer to a 2σ confidence level (σ is the standard deviation).*

p 8655 L4-6: Text changed to: *The two systems agree within 6% with some deviations after calibration and during ascent and descent phases of the aircraft where the differences lie outside the range of the combined stated accuracies.* The ascents and descents were included in the presentation of the data set.

p 8655 L10/22: done

p 8655 L11 and p 8656 L8/L12: Text modified as follows: *Rossby and smaller scale waves lead to deviations of the local tropopause from the climatological mean. The associated north-south excursions of air parcels are adiabatic to first order and largely reversible. They lead to large variability in trace gas distributions in a geometric (longitude, latitude, pressure) coordinate system, since chemical constituents are advected from regions with higher or lower climatological mean concentrations at the same time. In an attempt to remove this variability we therefore use a two-dimensional potential vorticity based equivalent latitude (ϕ_e) and potential temperature (Θ) framework to present our data. This framework follows the meridional deformations in PV contours and uses the fact that PV behaves like a passive tracer under adiabatic and friction-*

less motions. The equivalent latitude of a PV contour is defined as the latitude of a circle centered about the pole enclosing the same area as the PV contour (Butchart and Remsberg, 1986). This is illustrated in Fig. 1. If A(PV) is the area enclosed by the PV contour, then its equivalent latitude is given by ϕ_e (PV)=$\sin^{-1}$ (1- A(PV) / ($2\pi r_e^2$)). For the SPURT measurements the PV-equivalent latitude relationships were calculated for 27 individual isentropes from Θ=270 K up to Θ=400 K in steps of 5 K. In this way we obtained a field ϕ_e(PV, Θ) for discrete values of PV and Θ. The equivalent latitude for each measurement point was then calculated by bilinear interpolation onto [PV(t),Θ(t)]. PV(t) and Θ(t) are obtained from a combination of 6-hourly analyses and 3-hourly forecast ECMWF model fields interpolated in space and time onto the flight track [lon(t), lat(t), p(t)]. Since equivalent latitude is a PV coordinate, its calculation relies on monotonically increasing PV values. A recent study by Birner (2005) found, however, that PV in the UT/LMS exhibits only small meridional gradients away from the tropopause. An interpretation regarding geographical dependencies of tracers based on the equivalent latitude framework should therefore be treated carefully in regions away from the tropopause. Hegglin et al. (2005) used the equivalent latitude-potential temperature framework in a 2-D advection-diffusion model of the extratropical LMS in order to simplify the complexity of atmospheric transport processes and obtained correlation coefficients (R^2) between 0.72 and 0.94 between modeled and observed carbon monoxide distributions.

p 8657 L1/L15: done.

p 8658 L20: done.

p 8659: First sentence: No redundancy is obvious to us. The other comments have been addressed. Please see revised manuscript with the NO_x calculation moved to an additional paragraph 3.4 in the method section.

p 8660 L5: The influence of the Brewer Dobson circulation on tracer distributions in the stratosphere is well known and the conclusion obvious.

p 8661 Text added:*Vertical transport is hindered by strongly increasing potential temperatures, whereas quasi-horizontal transport along isentropes in the meridional direction is impeded by a strong gradient in PV at the tropopause.*

p 8661 L15: Text improved: *During the autumn and winter campaigns with little sunlight, NO_x mixing ratios are around 0.1 ppbv. The calculation of NO_x fails, if no sunlight is available (see Sect. 3.4).*

p 8664 L12/13: done.

p 8665 L1: Text added: *Well mixed is used here for the mixing of air masses with different characters (e.g. air masses of tropical and mid-latitudinal origin) down to scales below the resolution of the observation.*

p 8665 discussion of figure 10: We added the description of the error bars. The meaning of the blue circles is explained. Not all the campaigns are indicated to keep the figures more concise. Finally we leave it to the reader's judgment if the seasonal cycle in $\Delta NO_y/\Delta N_2O$ is convincing or not, since the slopes from October to April change significantly by about 20%. Furthermore, our interpretation of the seasonal cycle and the origin of the air masses mainly relies on the clearly convincing $\Delta O_3/\Delta N_2O$ slopes while we use the $\Delta NO_y/\Delta N_2O$ only as additional evidence.

p 8667 L18/L22/L24: done.

p 8668 L23/24: done.

p 8670: Section improved and renamed. Please see revised manuscript.

p 8671: Section improved. Please see revised manuscript.

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Discussion Paper

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