

## ***Interactive comment on “Atmospheric tracers during the 2003–2004 stratospheric warming event and impact of ozone intrusions in the troposphere” by Y. Liu et al.***

**Y. Liu et al.**

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We thank both the anonymous referees for their valuable and encouraging comments and suggestions. We would submit a revised version of the paper, which focus our emphasis much more on the interannual variations of the cross-tropopause ozone flux (CTOF) between the warming and cold Arctic winters. In the revised paper, there are 2 new model simulations, which are included in the manuscript.

First, we re-did the simulations using a recent version of MOZART-3 model, which includes 106 chemical species (the old one includes 48 species) at a higher horizontal resolution of 192x96 (the old one is 128x64). These jobs are really time-consuming and prolong our response. In order to highlight the potential impacts of the SSW events

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on the downward ozone flux into the middleworld and even across the extratropical tropopause, Parts 4.1 and 4.2 in the old manuscript have been reorganized and merged into one part in revised version (general Comment 1 from Reviewer #1 and Comment 6 from Reviewer #2) and an additional part concerning the variations of CTOF is added as Part 5.2. Technically, in order to give a more convincing result of the ozone flux between stratosphere and troposphere (general Comment 2 from Reviewer #1 and Comment 1 from Reviewer #2), the 3-dimensional ozone fluxes on each model grid are totaled daily and saved in the outputs.

Second, in order to compare the difference between "SSW" and "non-SSW" event, we made another model run for 1999-2000 winter, when it is cold and is under "non-SSW" condition. The results of the typical cold Arctic winter (1999-2000) are analyzed, and the interannual variations of the CTOF can be compared and a relatively general effect of the SSWs can be concluded (Comment 2 from Reviewer #2).

Moreover, the developing stages of the warming and cold winters are divided into two one-month-long periods (15 Dec-15 Jan, 15 Jan-15 Feb), by which the different the features of each winter can be more easily described.

Finally, a few typo errors and some potentially misleading statements are corrected and improved (Comments 2, 3, 4, 7, 8, 9 and 12 from Reviewer #1; Comment 8, 9 and additional comments from Reviewer #2). Validation materials of the MOZART model and the MIPAS are also added in Part 2. The detailed responses to each comment are listed below. The "Reviewer Comments" are noted first and then we give our "Reply" to the comment. We are submitting a revised manuscript that includes all the responses noted below.

### 1. Reply to Reviewer #1

The first general comment concerns Section 4.1., where temporal changes of ozone and N<sub>2</sub>O associated with the SSW are qualitatively described on the basis of zonally averaged values (Figs. 3 and 4). The description in the text should be more precisely,

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because some of the discussed features cannot directly be seen in the 2D fields presented in the figures (see also detailed comments 8, 9).

Reply: In the old manuscript, the vertical and horizontal N<sub>2</sub>O and O<sub>3</sub> distributions are separated as Parts 4.1 and 4.2. We have reorganized and merged them into the new Part 4 in the revised paper. The vertical and horizontal distributions of N<sub>2</sub>O and O<sub>3</sub> are much more combined with each other so that the temporal changes of the 3-dimensional features can be better highlighted. Detailed reply to Comments 8 and 9 can be found in the following text.

The second general comment concerns the diagnosis of the downward flux of ozone into the upper troposphere. It is not immediately clear how Figure 11 (weaker vertical flux in 2003/04 over the whole period) and Tables 1, 2 collaborate with each other.

Reply: We have replaced the Table 1 with the averaged heat flux over (45°-75°N, 100hPa), which can serve as a proxy of the Brewer-Dobson circulation strength. The total downward ozone fluxes during the three winters (2002-2003, 2003-2004 and 1999-2000) are quantified in Table 2. We find there is a consistency between the tendency of zonal mean eddy heat flux (as in Table 1) and the tendency of total downward ozone flux over East Asia (as in Table 2). This consistency implies that the downward transport of stratospheric ozone across the 100hPa into the 'middleworld' is closely associated with the strength of the Brewer-Dobson circulation, especially the descending branch over the East Asia. Since the downward ozone flux is also associated with some other factors such as the horizontal advection of ozone, the magnitude of the total ozone flux is not perfectly proportional to the eddy heat flux on a regional scale. More details about Table 1 and 2 are listed in the 'Reply' to Comment 3 from Reviewer #2.

Comment 1: Introduction, p13637, line5: A detailed investigation of the effects of the EPP event in late October/early November 2003 on ozone is given in a recent paper of Vogel et al. (ACP, 8, 5279-5293, 2008). This paper could be added to the references.

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Reply: This paper provides a good quantification of the EPP event on stratospheric ozone losses. We have added it in the Introduction, reference, and in Part 4. Further more, the main conclusions from this study can reply the 10th comment from Reviewer #2.

Comment 2: Chapter 3, p 13639, line 26: It is stated that "Several small and shallow downward intrusions, however, were observed after January 2003 (lower panel of Figure 1)". I assume that this can be seen by the wave-like contour line just below 30 km in Figure 1. Unfortunately, the pattern above this line is not so easy to identify (at least in print outs), as a result of the colour code.

Reply: In order to highlight the temperature differences between the two cases, especially the structures of the downward intrusions, the colorbar has been adjusted. Also, the contour lines have been added in Fig. 1 of the revised manuscript.

Comment 3: Chapter 3, p13640, line 1: "The major differences between the two events are highlighted by the white circles appearing in Fig. 1". Comment: I guess that you mean Fig. 2. Moreover, what do you mean by "white circles"? I guess that you mean the areas that indicate negative wind speeds (easterlies).

Reply: Very sorry for the missing "white circles" in Fig. 1! The both "circles" in Fig. 1 are used to highlight the differences of the Arctic temperature in two SSW cases, especially in the lower stratosphere. The two "circles" have been added in Fig. 1 of the revised manuscript.

Comment 4: In the next sentence, it is stated that a prolonged reversal of the zonal wind occurred near 10 hPa (Fig.2). Since it is obvious from Fig. 12 that this reversal occurred over a wide altitude range, the sentence should be replaced by something like " ... the zonal-mean wind exhibited a prolonged reversal at high latitudes between ... hPa, as illustrated in Figure 2 for the 10hPa level.

Reply: This suggestion has been adopted in Part 4 of the revised manuscript.

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Comment 5: Same page, line 23: I suggest to replace "vertical distribution" by something like "latitude-height cross-section" or just say "Zonal mean N<sub>2</sub>O and Ozone distributions".

Reply: According to the comment 7 of reviewer #1 and comment 6 of reviewer #2, we have re-organize Part4.1 and 4.2 into one part. Both the zonal-mean and horizontal characteristics are discussed together.

Comment 6: Page 13641, line 4: I suggest to replace "...is represented realistically. The calculated N<sub>2</sub>O concentrations are low inside the polar vortex, ..." by something like "... is represented realistically by the model results. Before the occurrence of the SSW, the calculated N<sub>2</sub>O concentrations are low inside the polar vortex, ..."

Reply: This suggestion has been adopted in Part 4 of the revised manuscript.

Comment 7: Page 13641, line 21: "The intrusion of high N<sub>2</sub>O concentrations into the polar region occurs through the tongues of high N<sub>2</sub>O concentration". This section should be formulated more precisely to avoid confusion. The reader could get the impression that the tongues can be seen somewhere in the 2D presentation of Figure 3, 4. However, Figure 3 just shows that the area of low N<sub>2</sub>O values (originally associated with the vortex) is filled by higher values after the SSW event. One could mention that this is related to (3d) tongues that have been previously observed by MLS.

Reply: In order to avoid the confusion mentioned above, we have reworded above sentence into "The dramatic increase of mid-stratospheric N<sub>2</sub>O concentration in the polar region can generally be attributed to the enhanced poleward transport of N<sub>2</sub>O-rich air with mid-latitude origin (see Figs. 9c and d)."

Comment 8: In line 28, you use again the expression "ozone-rich tongue". However, you refer to the meridional gradient of the mixing ratio maximum of the ozone layer. This may cause confusion. In the following text, expressions like "a high ozone tongue" should be replaced by something that is more appropriate to describe the modelled

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and observed ozone changes in the 2D presentation of Figures 3 and 4.

Reply: In the revised manuscript, all the "high ozone tongue" has been replaced as the "poleward transport of ozone-rich air" or just as "high ozone concentrations".

Comment 9: Page 13642, line 26: It is stated that "For example on 10 January 2004 the already elongated vortex on the 10 hPa surface splits into two parts". From my point of view, the situation shown in Figure 7 is better described by something like "On 10 January 2004 two remnants of the diluted vortex can be seen"

Reply: Thanks. This suggestion has been adopted in Part 4 of the revised manuscript.

Comment 10: Page 13643, line 22: You could add (if appropriate) something like "In addition, part of the difference in the strength of the structures in ozone and N<sub>2</sub>O may be attributed to the MOZART-3 transport scheme".

Reply: This sentence is modified as: "In addition, part of the difference in the strength of the structures in ozone and N<sub>2</sub>O may be attributed to the MOZART-3 transport scheme and the resolution of ECMWF dynamics".

Comment 11: Page 13645, line3: You could add a sentence that the decline in wave amplitude during period 2 and 3 is also reflected in the temporal development of the shape of the polar vortex (Figs. 7 to 10).

Reply: This sentence has been added in Part 5.1, going as "The decline in wave amplitude during period-2 was also reflected in the disturbance of the polar mid-stratospheric temperature (see Figs. 1) and the temporal development of polar vortex changes very rapidly (see Figs. 7 to 10)."

Comment 12: Figure 3: There is an inconsistency between the figure legend (7 January) and the figure caption (10 January).

Reply: All the MPV contours and figure legends have been consistent in the new figures (see Figs. 3 and 5). This comment is closely related to the comment 8 from Reviewer

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#2, where more explanations can be found.

## 2. Reply to Reviewer #2

Comment 1: In section 5, the paper shows the increase of the downward ozone flux at 100 hPa and the increase of ozone column between 100 and 300 hPa. Since this region includes part of the overworld (above 380 K, which corresponds to the stratosphere), the lowermost stratosphere and the uppermost troposphere, it fails to evaluate the ozone flux that will mix irreversibly in the troposphere, impacting thus directly the tropospheric ozone budget. A much more convincing diagnostic of the stratosphere-troposphere exchange should be given here.

Reply: In order to avoid the confusion about the ozone flux between "overworld" and "middleworld" and the ozone flux between stratosphere and troposphere, we separate Part 5 into two parts. In Part 5.1, we analyze the zonal mean eddy heat flux between 45°-75°N on 100 hPa (proxy for the Brewer-Dobson Circulation strength) and the vertical ozone flux on 100 hPa (proxy for the ozone flux between "overworld" and "middleworld"). We find the downward ozone flux over East Asia contributes predominantly to the total ozone budget. The variations of the extratropical and East Asian ozone fluxes are similar to that of the B-D circulation during the three winters. In Part 5.2, we calculate the total meridional and vertical cross-troposphere ozone flux (CTOF). In order to examine the different impacts of SSW in different latitudes, we separate the vertical CTOF into components in mid-latitudes (30°-60°N) and polar latitudes (poleward of 60°N).

Comment 2: In the same section, only one paragraph is dedicated to the increase of ozone in the upper troposphere. It is not demonstrated that this increase is only due to the SSW events and more detailed explanation should be given. Furthermore, in order to be able to provide general conclusions of the impact of such event on the tropospheric ozone budget, which would largely enhance the interest of this study, a comparison with downward ozone flux in normal winter conditions should be given.

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Reply: We have analyzed the situation in the cold 1999-2000 winter and the comparison results of the two warming winters and one cold winter have been given out in Part 5 of the revised manuscript. Generally, compared to the cold 1999-2000 winter, the enhanced wave activity during the typical warming winter (2002-2003) enhanced the downward ozone flux into the extratropical lowermost stratosphere (or "middleworld"), especially over East Asia. However, because of restricted wave activity associating with the long persistence of stratospheric easterlies, the total downward ozone flux on 100 hPa surface was about 10% lower during the 2003-2004 event compared to the situation in 2002-2003. Moreover, during the both the warming winters (2002-2003 and 2003-2004), the downward cross-tropopause ozone flux (CTOF) poleward of 60°N have increased ~10 times than the cold winter (1999-2000). More details can be found in Part 5.

Comment 3: The authors should quantitatively explain why they compute the heat flux over East Asia and relate the increase of the heat flux in this region with the increase of the downward ozone flux over this region.

Reply: In the previous manuscript, we try to correlate the variation of downward ozone flux over East Asia to the descending branch of Brewer-Dobson circulation over East Asia. Thus, we calculated the regionally averaged heat flux over East Asia. But, after the discussion with some colleagues, it seems the regionally averaged heat flux fails, theoretically at least, to represent the descending branch of the B-D circulation over the same region. Thus, in the revised manuscript we try to merely examine the relationship between the downward ozone flux over East Asia and the general strength of Brewer-Dobson Circulation. Currently, the vertical component of the EP flux which is proportional to the zonal mean eddy heat flux ( $v'T'$ ) is widely used as a proxy of wave forcing of the residual circulation (Fusco and Salby, 1999; Newman et al., 2001; Randel et al., 2002). Here, we use the averaged heat flux over (45°-75°N, 100hPa) as a proxy of the B-D circulation strength. The quantitative results are listed in Table 1. Moreover, the major centers of the downward ozone flux on 100hPa are located over

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East Asia (Fig. 11). That means the descending branch of Brewer-Dobson circulation over East Asia contributes most to the total downward ozone flux across 100 hPa. So, we calculated the total ozone flux over East Asia during the two periods for the three winters. The quantitative results are summarized in Table 2. We find there is a consistency between the tendency of zonal mean eddy heat flux (as in Table 1) and the tendency of total downward ozone flux over East Asia (as in Table 2). This consistency implies that the downward transport of stratospheric ozone across the 100hPa into the "middleworld" is closely associated with the strength of the Brewer-Dobson circulation, especially the descending branch over the East Asia.

Comment 4: Section 2.1, MOZART model: This paragraph should include an evaluation or references to validation studies of the model. A paragraph should be devoted to the validity of the MOZART model for the simulation of ozone in the 100-300 hPa region.

Reply: The following paragraph is added in new manuscript: "In this study, we are using MOZART-3 to examine constituent STE processes, specifically ozone, in the UTLS region. This work builds on three previous studies that have evaluated the validity of STE processes in MOZART-3. The first is the Park et al. (2004) evaluation of seasonal variations of several trace constituents near the tropopause. This study examined methane, water vapor, and nitrogen oxides (NO<sub>x</sub>) derived from Halogen Occultation Experiment (HALOE) satellite observations. The model results showed good agreement for methane and water vapor, but underestimated the nitrogen oxide abundance. It was postulated that this model low NO<sub>x</sub> was related to the lightning and convective parameterization used in MOZART-3. This work highlighted the importance of the Northern Hemisphere (NH) monsoons as regions for transport of constituents into the lowermost stratosphere. This work was extended in Gettelman et al. (2004) where the impact of the NH summer monsoon circulations on STE was specifically examined. This study concluded that a simulation using observed winds (same ECMWF winds used in this study) was able to represent transport events from aircraft and that

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these events can explain the global correlations of ozone and water vapor around the tropopause. A third study, Pan et al. (2007) was recently completed using MOZART-3 driven with both ECMWF and climate model meteorological fields. Diagnostics were created to evaluate model performance in the extratropical UTLS region. Overall the model results showed qualitative agreement with the observation in the location of the chemical transition across the extratropical tropopause. These studies give confidence that the MOZART-3 model driven with ECMWF meteorological fields is appropriate for use in STE studies."

Comment 5: Section 2.2, MIPAS observations: A summary of the results of the validation studies (particularly for the species and altitude region of interest to this article should be provided.

Reply: "The retrieved products have been validated by Cortesi et al. (2007) and Ridolfi et al. (2007)." in Section 2.2 has been explained in more detail as followed: "The retrieved ozone profiles have been validated by Cortesi et al. (2007). The MIPAS O3 partial columns were compared with coincided measurements from ozone sondes and ground-based lidar and microwave radiometers, it shows MIPAS O3 vertical profiles had the mean relative difference of 10% with the individual correlative data sets in the stratosphere. The retrieved temperature profiles have been validated by Ridolfi et al. (2007), in which MIPAS temperature is compared with correlative measurements from radiosondes, lidars, in-situ and remote sensors operated either from the ground or stratospheric balloons. The results prove that the bias of the MIPAS profiles is generally smaller than 1 or 2 K within the stratosphere. Intercomparisons of ground-based FTIR and MIPAS N2O profiles at 5 NDACC-sites in both hemispheres show good agreement between MIPAS and FTIR N2O partial columns: the biases are below 5% for all the stations and the standard deviations are below 7% for the three mid-latitude stations, and below 10% for the high latitude ones (Vigouroux et al. 2007)."

Comment 6: Section 4 on the impact of SSW events on stratospheric N2O and O3 is much more detailed and documented than section 5. Since the interest of this paper

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relates mainly to the latter section, a better balance between both sections should be found. In particular, I don't see the interest of section 4.2, which repeats results that were already found in previous studies. Can general conclusions be drawn from results presented in section 4?

Reply: The Part 4.1 and 4.2 have been merged into a new part as Part 4 and the analysis about the zonal mean and horizontal distributions are combined with each other. Moreover, both the horizontal distributions of N<sub>2</sub>O and O<sub>3</sub> on 10 hPa (in Figs. 7 to 10) have been interpolated onto the 30km height, which is an original level of the satellite observations.

Comment 7: Section 4.1: Figures 3 and 4 show clearly that N<sub>2</sub>O from MOZART simulations is overestimated as compared to MIPAS measurements. In the same way, some features are not reproduced, such as the positive N<sub>2</sub>O meridional gradient above 30 km. The authors should provide more comments on the various discrepancies between MOZART and satellite data.

Reply: More sentences about the discrepancies between MOZART and satellite have been added at the end of the new Part 4 such as "However, there are also differences in the magnitude of the concentrations of both compounds: some observed features are not reproduced, such as the overestimation/underestimation of upper stratospheric N<sub>2</sub>O/O<sub>3</sub> concentrations and the positive N<sub>2</sub>O meridional gradient above 30 km." The reply to this comment is further complemented by the Comment 10 from Reviewer 1.

Comment 8: It is not clear why figures 3 and 5 show N<sub>2</sub>O and ozone on January 10 2004, while the strength of the 2004 SSW event reached its maximum on January 7 as mentioned in the text. Furthermore the MPV distribution is different between the N<sub>2</sub>O and ozone figures on that day. Both figures should thus be revised.

Reply: This comment is complementary to the comment 12 of reviewer #1. There was an inconsistency in choosing the MPV contour values between the two figures. Both the dates in the two figures should be "Jan07". The revised figures (Figs. 3 and

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5) are consistent now. What should be added is that the 2003-2004 SSW reaches a maximum: the vortex becomes unstable and starts to split, according to the study by Manney et al. (2005). However, in the horizontal map on 30km, the vortex prominently splits after 10 Jan, 2004. Thus, we choose 10 Jan in Figs. 7 and 9.

Comment 9: The feature described in page 13642 lines 3-5 is not clear from the figure. Another layout could be given, in order to explain this feature.

Reply: In order to avoid the confusion, we have adjusted the sentence into "As highlighted above, in the middle of January, the split vortex shifts away from the polar region, causing a relatively low O3 concentration just at the location of the remaining vortex (see Figs. 5c, d and 9c, d)."

Comment 10: The effect of the EPP event in 2003 on ozone should be quantified.

Reply: A recent study by B. Vogel et al. (i.e., Model simulations of stratospheric ozone loss caused by enhanced mesospheric NO<sub>x</sub> during Arctic Winter 2003/2004, Atmos. Chem. Phys., 8, 5279-5293, 2008) has already given a good quantification of the EPP event on stratospheric ozone. Their main conclusion is that 10<sup>18</sup> DU accumulated ozone loss occurred until end of March 2004 caused by the transport of mesospheric NO<sub>x</sub>-rich air in early 2004. According to this study, the major ozone loss caused by the EPP-generated NO<sub>x</sub> occurs after the recovery of the upper stratospheric polar vortex (after the late-January). Thus, the stratospheric ozone in early January is little affected by the EPP-event. Vogel et al. also suggest that the impact due to enhanced mesospheric NO<sub>x</sub> values on ozone loss processes in the lower stratosphere (350-700K) is about 5% but lower than 9% for the Arctic winter 2003/04. Compared to the cold Arctic winters with strong halogen-induced ozone loss, this value would be negligible. So, generally, we don't need to consider the effect of the EPP event when we analyze the UTLS ozone flux.

Additional comment 1: Page 13642, lines 8-10: This sentence is not clear and should be rewritten.

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Reply: This sentence has been changed into "However, when compared with the 2003-2004 warming, the observed polar ozone concentrations in the middle stratosphere are obviously higher during the 2002-2003 winter (see Figs. 5c and 6c)."

Additional comment 2: Use hPa units for pressure throughout the text and figure legends.

Reply: Both the "mb"s in Figs. 2 and 12 have been replaced with "hPa".

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13633, 2008.

**ACPD**

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