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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ozone loss and chlorine activation in the Arctic winters 1991–2003 derived with the TRAC method" *by* S. Tilmes et al.

S. Tilmes et al.

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General comments

The specific comments of the anonymous referee 2 are very helpful for improving the manuscript. In some paragraphs of the manuscript the text was not as specific as it should have been, this will be improved in the revised manuscript as suggested. Further, thanks for finding many typing errors in the manuscript.

Specific comments

Referee 2: *A large part of the discussion in this paper is based on weak polar vortex, strong polar vortex, less strong and moderately warm vortex etc. How do you determine the strength of the vortex? How do you determine if a vortex is or not perturbed?*

In Section 3.2 of the manuscript the evolution of the polar vortex of each year is de-

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scribed in detail. Based on meteorological analyses (see cited studies) it was described when the vortex was disturbed. While no exact quantification for the extent of perturbation of the vortex is possible, we have defined what we consider a cold, moderate or warm vortex based on UKMO meteorological analysis. In the last paragraph of this section, the temperature conditions of the winters are classified in three categories: cold, moderately cold and warm winters, depending on the averaged area of possible PSC existence. In the revised manuscript the volume of possible PSC existence is used for this definition.

An average value of the strength of the vortex was not determined. This can be done considering PV of the vortex edge, using the Nash et al., 1997, algorithm (please find Figure 1 in the reply to the first referee).

The number of days when the poleward boundary of the vortex (as defined by Nash et al. (1996) criterion) exceeds a PV value of 40 PV-units may be used as a measure of the strength of the polar vortex. Therefore, the last paragraph of Section 3.2 is changed to:

"To summarise the temperature conditions for winters between 1991–92 and 2002-03, five winters are characterised as being cold (1992–93, 1994–95, 1995–96, 1996–97 and 1999–2000). These winters show a strong decrease of the HCl mixing ratio in the HCl/HF relation in spring and strong deviations of O_3-tracer profiles from the early winter reference function. For the cold winters the daily V_{PSC} average in 400–550 K between mid-December and March is between 20 * 10^6 km^3 and 40 * 10^6 km^3 (shown below in Section 6). Moderate deviations from the O_3/HF reference were found in 1991–92, 1993–94, 1997–98, 2000–01 and 2002–03. The daily V_{PSC} average is between 5 * 10^6 km^3 and 15 * 10^6 km^3. little chlorine activation and very little deviation from the early winter ozone-tracer reference function. The value of the daily V_{PSC} average does not exceed 3 * 10^6 km^3 for the very warm winters.

The temperature conditions are closely related to the strength of the vortex. A measure

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of strength of the vortex can be derived summarising these days of each year over the entire winter, when the poleward boundary of the vortex (as defined by the Nash et al. (1996) criterion) exceeds a certain threshold value of PV. Here 40 PV-units (PVU = 10^{-6} K m^2/(kg s)) at 475 K is used. For four of the cold winters 1992–93, 1994–95,1995–96 and 1999–2000, this is the case for more than 100 days of the winter; in 1993–94, 1996–97 and 2002–03, it is 80–90 days (moderately warm or cold winters), in 1991–92, 1997–98, 2000–01, 2001–02, 40–63 days (moderately warm or warm winter), and 1998–99 only 20 days of the year (warm winter)."

Referee 2: *Can you explain what you consider as sufficiently long lifetimes for tracers? Or later in the same paragraph: very long lifetimes. What is the order of magnitude?*

Using the TRAC technique, relations of long-lived tracers are considered. Long-lived or dynamical tracers are chemically long-lived gases. The dynamic processes of these gases, such as vertical advection and horizontal transport, are fast compared with their local chemical lifetime. Ozone also can be considered as a long-lived tracer in winter at high latitudes where the lifetime of O_3 is larger than $\approx$ 100 days, as long as no halogen-catalysed chemical ozone loss process is active (e.g., Proffitt et. al, 1992 and WMO 1990). The stratospheric chemical lifetime of CH_4 is 93 $\pm$ 18 years (Volk et al., 1997). The only known sink of HF is transport to the troposphere (e.g. Chipperfield et al., 1997).

This discussion about the lifetime of the tracers used in connection with the tracer-correlation technique is already reported in Müller et al. (2002) and Tilmes et al. (2003). We agree that in the relevant section these citations have to be added.

Referee 2: *Discussion about the very strong ozone losses in January as deduced from SAOZ and HALOE measurements:*

On Page 2196, line 10 in the submitted manuscript it is written: "The very large ozone loss rates in the early winter 1993–94, 1994–95, 1995–96 and 1997–98 derived by SAOZ can be neither confirmed nor falsified here due to the lack of observations.

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However, there is also no sign of such strong ozone losses as deduced from SAOZ for January considering HALOE measurements within the vortex boundary region in 1993–94 and 1995–96."

Because no results of ozone loss of winter 1991–92 are available from SAOZ this winter was not compared with TRAC. Therefore, the sentence in the abstract indeed applies for all winters were SAOZ and HALOE are comparable. However, we agree that the sentence in the abstract is not clear. It will be rewritten in the following way:

"Results estimated here are in general agreement with the results obtained from other methods. Differences occur mainly owing to different time periods considered in deriving accumulated ozone loss. However, there is no sign of very strong ozone losses as deduced from SAOZ for January in winters 1993–94 and 1995–96 in a TRAC analysis of HALOE measurements."

(please find further comments on this subject in response to Referee 1 to the same subject)

Section 3:

Page 2177 line 26. *For the winter 1996, using one single observation of HALOE, you see strong chlorine activation (fig 1 bottom) but no O_3 loss. Can't it be explained by the latitude of HALOE observations between 23-31/1/96.*

The latitude of this profile is 50 degree North. Nevertheless, this profile was measured inside the vortex (defined by Nash et al., 1996). At this time and location solar illumination is expected, but the vortex was illuminated only for a very short time before 23-31 January (please find Figure 2 in the reply to the first reviewer, which is the Figure 1 of the revised manuscript). Therefore, the HCl was already strongly reduced but only little ozone loss was found (as described in detail in Section 3.1 paragraph 3).

Page 2177 line 26 to page 2178, line 9 will be rewritten in the following way:

"Changes in the O_3/HF relation due to isentropic mixing are not expected during De-

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cember and January, because at that time the vortex was already very strong. Further, at this time and location ($\approx$ 50) of the profile solar illumination is expected, but the vortex was illuminated only for a very short time before 23-31 January 1996 (see new Figure 1). Therefore, the HCl was already strongly reduced but only little ozone loss inside the range of uncertainty of the reference function was found. Of course, the HCl-tracer relation decreases much faster than the O_3-tracer relation, because chlorine activation occurs on much shorter time scales than ozone loss. The fact that in 1996 – in spite of the early vortex having been cold and strong – no significant ozone loss occurred during January may be explained by the very small amount of sunlight that has illuminated the early vortex."

Page 2181, line 10: *How do you deduce from figure 4 that the strong chlorine activation in February and at the beginning of March occurred only in the lower stratosphere below 420 K.*

The sentence will be rewritten in the following way:

"In January, February and at the beginning of March very low HCl mixing ratios are clearly noticeable and strong chlorine activation (Fig. 4) occurred in the lower stratosphere. HCl mixing ratios are nearly zero in January and less than 0.3 ppbv in February below the 0.6 ppbv HF level, which is below the 420 K potential temperature level."

Referee 2: *Page 2181 line 19: The paragraph should probably be rewritten for clarification, insisting on the years and on the altitude range at which the loss occurs.*

The paragraph will be rewritten in the following way:

"The strongest local ozone loss of all the years considered, about 2.4 ppmv in 1995–96 and 2.5 ppmv 1996–97, was found in the altitude range from about 450–490 K for both winters. In the cold winters of 1994–95 and of 1999–2000 the maximum of local ozone loss profiles was similarly strong. It occurred for both of these winters in the altitude range from about 410–460 K."

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Section 5: reference will be corrected

Referee 2: *Page 2196 line 18: Be more specific.*

“Such values are approximately in agreement with the moderate ozone loss deduced from ILAS 0.5–1.0 ppmv ($\pm$ 0.2 ppmv) and HALOE 0.9–1.4 ppmv ($\pm$ 0.2 ppmv) at the 475 K level (Tilmes et al., 2003).

“approximately” will be canceled, because the values ARE in agreement within the range of uncertainty. New sentence is:

“Such values are in agreement with the moderate ozone loss within the range of uncertainty deduced from ILAS 0.5–1.0 ppmv ($\pm$ 0.2 ppmv) and HALOE 0.9–1.4 ppmv ($\pm$ 0.2 ppmv) at the 475 K level (Tilmes et al., 2003).

Section 6:

This section was rewritten substantially, please find further comments in the reply to the first referee.

Section 7: Page 2202 line 25:

The paragraph will be rewritten: “SAOZ results for the winters 1996–97, 1999–2000 and 2001–2002 seems to be somewhat large.”

Minor comments:

Page 2173 Line 27: will change to:

“Further, the use of different meteorological analyses may result in differences of up to $\approx$ 25 % for the deduced value of V_{PSC}.”

Page 2191, line 28:

The signal saturation problem is an instrumental problem, it may be a systematic problem. It is becoming noticeable only for some years considered in this study.

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Technical corrections

Further suggested changes will be implemented in the revised version of the manuscript. Typing errors will be corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2167, 2004.