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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 N. Harris are very helpful to improve the manuscript and will be addressed when preparing the revised manuscript. Some misunderstandings may have been caused by not very clearly written paragraphs – these sections will be improved in the revised manuscript.

Specific comments N. Harris: *It would be interesting to have brief discussion of the interannual variability of the early winter reference function shown in Fig. 3, especially if the HF and CH_4 could be de-trended. I think there is a general view/assumption that the November reference functions (vortex spin-up rather than the pre-ozone loss ones needed here) should be the same from year to year....*

We agree, that to calculate chemical ozone loss using the TRAC technique, the pre-ozone loss reference function is needed rather than the spin-up reference in November.

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This is discussed in detail in Section 2.1 page 2172 line 13 to page 2173 line 9.

Indeed, significant interannual differences of de-trended early winter reference functions are found in this study. To emphasize this fact and describe it more clearly, Section 3.1, page 2179, line 25 to page 2180, line 1, will be rewritten as explained below, and a further figure (new Figure 4) will be added including de-trended O_3/HF early winter reference functions as suggested.

'The deduced ozone-tracer relation in the early winter has its own characteristics each year. This is due to inter-annual differences in polar vortex development and not due to chemical loss (Manney et al., 2003a). Considering ozone-tracer relations, year to year variations should be influenced by a trend of mixing ratios of the long-lived tracer used. This is the case using HF as the long-lived tracer (Figure 3, top panel), because HF mixing ratios increased by $\approx$ 0.4 ppbv in the middle stratosphere from 1991 to 2000 (Tilmes, 2004).

On the other hand, the growth rate of 60 ppbv in 12 years of CH_4 – taken from the tropospheric growth rate derived by Simpson et al, 2002, – is very small compared to the observed stratospheric CH_4 mixing ratios (between 0.5–1.5 ppmv) and therefore is not significant for the present analysis. Further, ozone was relatively constant during the 1990s in northern mid-latitudes (WMO, 2003). Nevertheless, a decrease of ozone was found in the polar regions, but mainly in the southern hemisphere (WMO, 2003). Therefore, considering O_3/CH_4 reference functions (Figure 3, bottom panel) the interannual differences of the early winter reference functions are not a result of a significant trend of methane. There is a hint of a trend of ozone in high northern altitudes towards lower ozone mixing ratios, but interanual differences in ozone mixing ratios are possible for different reasons (as described below). O_3/HF early winter reference functions can be de-trended with regard to HF, using the HF growth rate deduced from the HALOE HF/CH_4 relationships (Table 1) (Tilmes, 2004).

New Figure 4.....

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The range of interannual differences of ≈ 1 ppmv ozone mixing ratio– in altitudes above the 0.8 ppbv level of HF or the 1.0 ppmv level of CH_4 – are similar for both O_3/HF and O_3/CH_4 relationship (see Figure 3, bottom panel and new Figure 4). The largest ozone mixing ratios are found in winter 1991–92. This is possible due to enhanced global transport in this winter owing to the eruption of the Pinatubo in June 1991. Very small ozone mixing ratios are found for the three winters 1999–2000, 2001–02 and 2002–03. This may be due to an earlier isolation of the polar vortex for example in winter 2002–03. Additionally, ozone loss may have already occurred at the time when the reference function was derived in winter 1999–2000 and 2001–02 (see new Figure 1) [discussed in interactive comment to M. Rex (Figure 2)]. Due to the different influences that control ozone mixing ratios in high northern latitudes in the early winter, a possible trend of ozone cannot be determined in this study."

N. Harris: *There was a major warming near the end of January which resulted in transport of air into the vortex reported by various authors ....*

In winter 1991–92 no HALOE profiles are available at the end of January, thus ozone loss and the homogeneity of ozone loss profiles cannot be investigated during or shortly after this major warming. Only one HALOE profile was located inside the outer vortex in January 14, 1991, and inside the vortex in February 1992. Ozone loss was found to be homogeneous for a substantial number of HALOE observations in the Arctic vortex during the second part of March and April 1992. This is an indication that the air from outside, that entered the vortex at the end of January, is well mixed within the vortex during March and April. At this time of the year, no significantly inhomogeneous temperature distribution was observed, and no PSCs can be expected any more. Further, there is a clear distinction between inside and outside vortex air O_3/tracer profiles indicating again that the vortex in February and March 1992 was well isolated from mid-latitude air.

Isentropic mixing may change the tracer-tracer relationship as described in Section 2.1, page 9, paragraph 3: "Further, Müller et al.(2001) used balloon-borne measurements

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in the Arctic winter 1991-92 to show that the impact of mixing between air masses from outside the vortex with air inside the vortex would result in a tendency to greater ozone mixing ratios in the ozone-tracer relation." Such mixing should therefore result in an underestimation of chemical ozone loss.

Using HALOE measurements, we cannot determine the precise influence of the mixing events in January 1992, but defenitely it is not so strong, that profiles scatter above the derived early winter refernce function. Additinally, the vortex in the lower stratosphere was not much affected. The zonal winds at 60 $^{\circ}$ N considerably weakened, but remainded westerly (Naujokat et al. (1992). Therefore, the stop of the calculation of ozone loss is not neccesary for this winter. Actually, considering the tracer-tracer corrlations, no significant increase in the relation is getting obvious in February. Therefore, we can give a lower limit of the ozone loss for this winter.

In the relevant section of the revised manuscript the issue of the impact of the major warming in January 1992 is discussed as suggested. It will be emphasized more clearly, that March and April ozone loss values are discussed and it will be argued that the major warming in January 1992 should result in an underestimation of chemical ozone loss in this winter.

N. Harris: *... the authors imply that inhomogeneous temperature distribution must result in inhomogeneous ozone loss*

On page 2188, line 24, it is written:

"The meteorological developments during various winters, described above, may be responsible for inhomogeneous temperature distributions inside the vortex and, therefore, are responsible for the inhomogeneities in ozone destruction inside the entire vortex."

This is indeed the case for winter 1996–97. Considering March and April 1992, there are no PSC regions any more, and no sign of a significant inhomogeneous temperature

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distribution.

In view of the comment, this paragraph will be change to:

"The meteorological developments during various winters, described above, may be responsible for inhomogeneous temperature distributions inside the vortex. They may be responsible for the inhomogeneities in ozone destruction inside the entire vortex, most obvious in winter 1996–97. In this winter inhomogeneities in temperature resulted in inhomogeneties in denitrification within the polar vortex (McKenna et al., 2002b) that are the likely cause of the inhomogeneties in ozone loss (Schulz et al., 2000 and Tilmes et. al, 2003)."

N. Harris: *Section 5*

We agree that the structure of Section 5 is somewhat unclear. Some subsections will be implemented in the revised manuscript and the structure will be improved.

N. Harris: *In the second paragraph (and some other places), I think the authors should give an upper limit on the ozone loss they think occurred rather than say it is zero.*

The upper limit of very small ozone loss values is already shown in Table 4 and 5. It will be added in additions in the text in the relevant paragraphs.

N. Harris: *In the seventh paragraph, the rationale for differences with MLS is very vague. Why can't the authors integrate over the same range as MLS.*

MLS ozone loss was calculated over pressure levels for the height range above 100 hPa. The calculations of ozone loss using TRAC was performed considering potential temperature levels as a measure of altitude. Potential temperature is a conservative quantity that is rather robust, whereas using pressure levels to calculate chemical ozone loss may be influenced by small pressure variations due to adiabatic fluctuations that influence mixing ratios differently in different years.

Nevertheless, for the revised version of the manuscript in addition we calculated

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HALOE ozone loss between the pressure range of 100 to 40 hPa to provide a more exact basis for comparison with MLS results. In the revised manuscript, Table 8, page 57, will be changed using the HALOE ozone loss calculated in pressure levels. The discussion will be rewritten. The new values (Table 1) show that significant differences occur between MLS and HALOE results in winters 1991–92 and 1994–95, because the time intervals considered using both techniques are not exactly the same. This is also a likely reason for differences between result from HALOE and the vortex average approach, as descirbed in the reply to Referee 1. The results of the other years fit quite well inside the range of uncertainty.

Table 1: Calculated chemical loss in column ozone loss (DU) in the Arctic over seven winters, HALOE results and MLS results (Manney et al., 2003), are compared.

date	MLS[a] above 100 hPa	HALOE entire vortex in 100–40 hPa	HALOE vortex core in 100-40 hPa	HALOE - MLS entire vortex
March 1992	29	56±15	57±15	27 ±15
March 1993	54	67±16	77±16	13 ±16
March 1994	35	26±15	34±15	-9 ±15
March 1995	36	68±17	73±17	32 ±17
March 1996	63	74±20	75±20	11 ±20
March 1997	35	35±14	45±13	0 ±14

[a] taken from Manney et al. (2003), Table 3

N.Harris: Section 6: *There are some interesting results presented here but it is not really clear what the authors think they mean...*

Here, the important results of this paper seem to be not expressed clearly enough, this

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will be improved in the revised manuscript. Section 6 will be completely rewritten, see interactive comments to Referee 1.

Here, the comparison between results of this study and these of Rex et al. (2004), Section 6, will be performed for exactly the comparable values (time interval, altitude range and meteorological analysis) of the volume of PSC. Further ozone loss in DU will be compared for the same altitude range and time interval. Minor differences of time intervals will be discussed in the revised text. Additionally, different time intervals will be considered to calculate the average value of potential PSCs and results of this study will be supported. The reason for differences of the two approaches are outlined in the revised section.

Part of the new Section 6: "Considering a time interval for estimating V_{PSC} from mid December to the end of March further important factors controlling ozone loss are masked by averaging V_{PSC} over nearly the entire winter. In the next part of this section, different time intervals to average V_{PSC} will be used to perform a comparison between chemical ozone loss calculated using the TRAC method and V_{PSC}. For this purpose, both parameters (ozone loss and V_{PSC}) are estimated in the time same interval between the time of the early winter reference function and end of March (February in 2000–01 and 2002–03) using the TRAC technique."

Further discussion, see interactive comments to Referee1, Section 6.

N. Harris: *the discussion of early January losses in the Conclusions section (it should be retitled Discussion) also seems dated as there is no reference to the recent Rex et al GRL paper (2004) on available measurements*

First, the Section 'Conclusions' will be devided into two separat sections called Discussion and Conclusions. We assume here, the paper Rex et al. (2003 b) was meant. This citation will be added on page 2202, line 29 and ozone losses will be changed in ozone loss rates (discussion below):

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"Further, the unexpected large ozone loss rates during January in the winters 1995, 1996 and 2000 based on data from Match, POAM II and POAM III and MLS (Rex et al., 2003 b) could not be found using the TRAC method."

N. Harris: *To me the tracer technique seems to be the outlier as there is reasonable agreement between the other techniques.*

The strong early January losses found by the SAOZ method in winter 1993–94 to 1995–96 cannot be found in this study, because of the lack of measurements as described in the revised text, Page 2196, line 10:

"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. 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, where a comparison is possible."

Additionally, strong ozone loss rates were found using other approaches as described by Rex et al. (2003 b). These ozone loss rates may be quite large in some cold early winters, nevertheless only to a very small value of accumulated ozone loss that cannot be clearly identified using the TRAC method. Thus, there is no contradiction between these results. This discussion will be added on Page 2202, line 31 in the revised manuscript.

Further, the accumulated ozone loss values in March of TRAC and the vortex average approach agree quite well except for the differences discussed in Section 6. Therefore, we do not see that the result of the TRAC method are an outlier.

Minor comments N. Harris: *Given a similar approach has been used by others, I am not sure that it is worth giving this approach its own name (TRAC)*

TRAC is a shortening for 'tracer-tracer correlations'. The name should imply to calculate chemical changes of tracers, e.g. ozone, excluding transport processes inside of

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the isolated polar vortex region. Here, it should describe a specific technique, including all the technical procedures used, for example including the Nash criterion to derive the vortex edge an so on.

To prevent confusion in the future (see interactive comment by M.Rex) the name TRAC method will be changed to 'Tracer-Correlation Method' in the title and the shortening will be only used in the text.

N. Harris: *The start of the description of the 1995/96 winter section 3.2 is a bit vague....*

The start of the description of the 1995/96 winter section 3.2 will be changed to:

"The winter 1995–1996 was classified as "the coldest winter" recorded by the US National Meteorology Center (NMC) in 18 years (Manney et al., 1996a)."

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