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Interactive comment on "Early unusual ozone loss during the Arctic winter 2002/2003 compare dto other winters" *by* F. Goutail et al.

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At first, we would like to thank Dr R. Salawitch and the anonymous referee for their helpful comments and suggestions.

Three important modifications have been done to the manuscript:

1) Referee #1 noted a possible offset between SAOZ and the REPROBUS simulation at the beginning of December 2002. We have investigated this issue and found that the passive ozone was not properly initialized in the lower troposphere, resulting in too large total column in early December. Note that the chemically-integrated ozone was not affected by this problem. We have corrected the initialization of the passive ozone and have performed a new model simulation, which is now in line with the SAOZ

measurements at the beginning of the winter (see Figure 1). Consequences of this are a delayed onset of the ozone loss in December as well as a slightly reduced final loss in late March (Figure 2) compared to the initial run that was presented in the submitted manuscript (20% vs. 23%). The final chemical loss computed by the model is also reduced in the revised version of the paper (13% vs. 20%).

2) The SLIMCAT team has provided new simulations from an improved version of the model which corrects problems found in the transport code in the lowermost stratosphere (between the tropopause and 350 K). This caused too large O3 mixing ratios and had thus a large influence on the column. The new version of SLIMCAT allows an evaluation of the SAOZ/SLIMCAT ozone loss added in the revised manuscript.

3) According to both referee remarks, Section 5 and figure 4 were a little confusing. They have been simplified. The model results with the version available at the end of each winter has been removed. Indeed both models have received too many changes since 1993 for useful comparisons. The figure only show now the SAOZ/REPROBUS results. In addition, the indication of a "cold" winter based on one temperature level (475K) was not enough for the discussion. A second level (550K) has been added.

1. Reply to Referee R. Salawitch

1a. description of the rate constant for ClO+ClO and cross section for ClOOCl used by each model for the 2002/03 simulation; In Reprobus the rate constant used for the ClO + ClO + M reaction is that recommended by the latest JPL compilation (Sander et al. [2003]). This recommendation is based on a simultaneous fit to data from Bloss et al. [2001], Nickolaisen et al. [1994], and Trolier et al. [1990]. The references given in the submitted manuscript were not up to date. Reaction rates recommended by Sander et al. [2003] now supersede in Reprobus those given by Sander et al. [2000]. Cl2O2 cross-sections used in Reprobus are also those recommended by JPL (Sander et al. [2003]). The long wavelengths tail measured by Burkholder et al. is not taken into account in the simulations presented in this paper. For the recent SLIMCAT comparisons

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(1999/2000, 2002/3, 2003/4), the ClO + ClO rate from Sander et al (2003) has been used. All model photochemical rates were taken from this except for the absorption cross sections of Cl2O2. For this, the data of Burkholder et al (1990) extended to 450 nm by extrapolation was used. The above information are now included in the revised manuscript.

1b. description of how much Bry was present at 435, 475, and 550 K for each model, at a particular time of the 2002/03 run (these levels picked to correspond to data shown in Fig. 2; other levels would be OK). If possible, it would be quite useful for many readers if a column for BrOx plots be added to Figure 6. It might make sense to break this figure up so that only 3 columns are shown per page, to allow each panel to appear larger than the present version, which is difficult to read. The REPROBUS BrOx plots for the 2002/03 winter have been added to the figure 6 and the figure itself has been divided in four parts to have it more readable. For the old SLIMCAT runs, a Bry loading of 20 pptv has been considered. For the 'new' run (presented here for 2002/03) the model used a time-dependent loading of tropospheric source gases from WMO (2003), with an additional 6 pptv to account for short-lived Bry species. Overall, this gives a Bry loading of around 21 pptv

Finally, it would also be useful, if possible, to show in the published paper a version of Figure 6 from SLIMCAT. It was not possible to add in the paper a complete figure 6 for SLIMCAT, however a "reduced" figure showing HCl, ClOx and O3 loss the same way as for REPROBUS has been added in the manuscript for one day, December 5 and same two levels 550 k and 475K

2a. Clarify whether the SLIMCAT results shown in Figure 4 are for the version of the model available at the time of observation; 2b. Would be nice if somehow, the major changes to both models relevant to the comparison (e.g., photochemistry of ClOx; any changes in assumptions regarding levels of BrOx) were noted, perhaps in a table; 2c. Paragraph starting on line 14, page 5025, is confusing as written. First sentence says 1994/95 loss is not reproduced well. Third sentence says Goutail et al. (1999) found

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loss was "well captured". Very confusing which results are even shown in Figure 4; are they the results of Goutail et al. (1999) that according to the text compare well to observations? Clarification of this paragraph would be helpful. Figure 4 has been simplified, all the model O3 loss results have been removed from the plots, only the SAOZ/REPROBUS results are presented for each year.

Now, the paper is limited to modelled results for the winter 2002/03 only.

3. I have some concern that the temporal evolution of ozone loss for a given winter might be influenced by the introduction of new stations, as they become illuminated. Since the stations rely on solar illumination, the early data period contains only southerly stations. As time progresses, data from more and more stations becomes available. I suspect that the distribution of ozone loss within the vortex, combined with mixing, is such that this is not a major concern. However, it is not discussed at all in the paper. Might the "stabilization of ozone loss" in early February, discussed on page 5023, line 19, be due to this effect? The final published paper would be improved if, to whatever degree possible, there was some discussion of possible bias in the temporal evolution of the observed ozone loss rate due to introduction of the northward stations over the course of winter. A discussion has been added in the paper. The loss simulated at all stations throughout the winter, including thus in the darkness, has been plotted. No significant change could be observed.

4. The next comment follows point 3. For the ozone evolution over 11 winters shown in Figure 4, there are some stations always present, while data from a few appear when the station turns on, as indicated in Table 1. I suspect the introduction of these new stations has a minimal effect on the year-to-year comparisons, but this point is not addressed at all in the present paper. The final published paper would be improved if, to whatever degree possible, there was some discussion of this point as well. There was a mistake in table 1: Harestua station started measurements in 1993 and not 1994. There is only one station - Salekhard - which has been introduced since the beginning of the analysis starting in December 1993. This "added" station located on

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the western side of Siberia, on the polar circle, is alternatively IN and outside vortex as the other stations located also on the polar circle. The result is an increase of the number of measurements in order to get better statistics.

Minor Points: All minor points have been taken into account in the new version of the manuscript and the confusing sentences have been rewritten..

2. Reply to Referee #1

In Section 5, Fig. 4 is discussed. SAOZ results are shown as well as REPROBUS and SLIMCAT results. I would suggest to interchange Section 5 and 6. Then, the authors would finish the discussion of the winter 2002-03 and afterwards compare with the results of other winters and additionally, with different measurement results. The idea behind presenting section 5 before section 6 was to show first the measurements for the winter 2002/03, then the comparison with other winters (comparison with model results as they were available at the end of each winter), and finally discuss the model for the winter 2002/03 (up-to-date version of both models). We have now simplified fig4. Only SAOZ/REPROBUS results for all winters are presented now with is no reference to REPROBUS and SLIMCAT modelled loss (too many changes in the models since 1993)

In general the discussion of the uncertainty of results is missing as well as the comparison with different published results. More details have been given including references to intercomparison papers etc..

Abstract: The abstract could do with some more specific information. It should be mentioned that the authors use the REPROBUS model for the eleven years, and that SLIMCAT and REPROBUS results are compared for the winter 2002-03. Further, it should be clearly stated, that ozone loss is estimated inside the poleward boundary of the polar vortex and that it is an average over 7 stations. Uncertainty ranges of the results are missing. Could the authors emphasis more clearly, why ozone loss is unusual in this winter. The abstract has been completely changed.

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Section 3: Page 5022, line 16. 'the edge of the vortex', How is the edge defined? A complete description has been added in the manuscript.

Page 5023, line 13-16: 'Consistently with the meteorology, the loss started very early (Fig 2, bottom), during December, a 10 % total column ozone reduction ....' The scatter of ozone loss within one station and within all stations considered are quite large (up to 15 % at the beginning of January for Sodankyla, and at the beginning of February for Thule). How is this possible? At the beginning of December 2002, ozone loss of up to 5 % is obvious, considering Sodankyla (Fig.2, bottom panel, red open circles), at a time when no ozone loss should have been occurred (as shown in Fig.6). Is there maybe an offset that wrongly increases accumulated ozone loss? You were right. There was an offset at the beginning of December due to a wrong initialisation of the ozone field in the troposphere. The REPROBUS simulations have been corrected, leading to significant changes.

Further, at the beginning of January 2003, a decrease of the 10-day average measured total ozone reduction (increase of the black line from -12 percent to -10 percent) is obvious in Fig.2 bottom panel. In considering accumulated ozone loss, what does this decrease mean? Looking at Fig.2 bottom panel, it is hardly possible to derive the exact time, at which time 10 percent ozone loss were reached using these results, owing to the strong scatter. Would an average of less days change the results? The uncertainty of the total column ozone reduction and of the average rate per day should be added. The uncertainty of the total column ozone reduction and of the average rate per day have been added.

Section 5. 'While during most of the winters, the loss begins in January or eventually in mid-December like in 1995/96, in 2002/2003, it started very early in December in coincidence with very low stratospheric temperatures.' This paragraph could be enhanced with regard to the following issues. What about 1997-98 and 2003/2004? In these years, ozone loss starts very early, derived by SAOZ/REPROBUS (Fig 4). Further, in 1994/95 temperatures are very low in December as well, and in 1999/2000 in the sec-

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ond half of December. However, with SAOZ much less ozone loss is calculated. Next paragraph: The comparison between SAOZ/REPROBUS results and model results should be extended to all winters considered. Here, only 1994/1995 and 1995/1996 are discussed. Why is ozone loss in 1994/1995 and 1995/1996 still not reproduced by REPROBUS and SLIMCAT although 'improvements have evidently been made in the two 3-D CTMs'? The 1994/95 and 1995/96 simulations have been performed with the Reprobus version available by that time. Many improvements (ECMWF 60 levels, chemical rates, PSC schemes ..) have been added since then. It is not possible to reprocess previous winters with this "new" version because ECMWF is not available above 10 hPa.

By the way, in 1994/1995, strongest ozone loss was calculated in this study, however other published results do not agree with this result. The range of uncertainty of all winters should be added. Difficult to estimate the error in 1994/95. A good comparison between the results of the various methods is available for the winter 2000 in Harris et al. Section 5 has been modified. The model ozone loss has been removed from the right part of the figure and the temperature at 2 level is presented now on the left part.

Section 6.1: First paragraph. In Fig.5, at the beginning of December, an offset of about 3 percent ozone loss is obvious (all symbols scatter below zero). This may influence the result. As said above, this offset was due to an incorrect initialization of tropospheric ozone and has been corrected in the revised manuscript. See introductory remarks.

Third paragraph: '..., but HCl was completely depleted', at which altitude? The text has been modified: On December 25 (Figure 6, middle), few PSCs could be still seen, HCl was completely depleted at 550 K and almost completely at 475 K and chlorine and bromine were largely activated in the whole vortex. However, the vortex was now elongated towards illuminated latitudes, and significant ozone losses appear at its periphery, more apparent at 475 K than at 550 K.

Fig. 6: In December 25, ozone depletion occurs at the vortex boundary of 5-10 % in

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475 K and less in 550 K. It would be interesting to calculate an entire vortex average, that is certainly less than 5 % in 475 K and much less than 5 % in 550 K. Would this mean that the results of this study depend on the location of measurement stations? The study shows that the loss starts at the periphery of the vortex, (where the stations are located). In addition figure 6, on 25 December shows that inside the vortex, even in sunlit areas, some stations may see depleted ozone and some non-depleted ozone. This may explain the large scatter in figure 2, observed for example, at Sodankyla around January 5.

Section 6.2 last paragraph: In Appendix A and B, two models are described in detail, but during the discussion of different results, I cannot find a reference concerning the difference between these models. Which differences can be seen in the results? Some more discussion about this issue should be added in this section and in Section 5. The description of the model has been introduced in section 6 now. Differences between the 2 CTM are discussed in the section

Conclusions: 'An unusually early ozone loss was observed in 2002/2003, at least one month earlier than during any of the previous eleven winters' It is not 'at least' one month earlier (see 1997-98 and 2003/2004). last sentence: There is not indication that ozone loss has taken place without illumination of the vortex in this paper. What is meant by 'low sun?'. An unusually early ozone loss was observed in 2002/03, at least ten to twenty days earlier than during any of the previous eleven winters. The conclusions have been rewritten.

Minor comments All minor comments have been taken into account

Fig. 6.: scales of the figure cannot be read at all This figure is now devided in four parts for better clarity: Figure 6a and 6b at 550 K and Figure 7a and 7b at 475 K. Figure 6a and 7a show PV, PSC surface area and BrOx. Figure 6b and 7b show HCl, ClOx, O3 loss.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 5019, 2004.