

Answers to N.R.P. Harris (Referee)

Thanks for your comments, very helpful for improving the paper.

Major Comments

A good description of the Reprobus photochemistry and microphysics is required since that underlies some of the later discussion and there have been many changes to our understanding since 1994. For example on p316, line 17 a conclusion is made about the importance of HOx - NOx chemistry based on the Reprobus calculations. The validity of this conclusion is significantly weakened by the lack of (a) a real description of the Reprobus chemistry and microphysics, and (b) a comparison between the observed and modelled column NO₂.

a) Description of Reprobus chemistry

Indeed, the description is very short and needs obviously some update. However, here Reprobus is used in two configurations only, passive and gas phase, but not heterogeneous. Since a discussion of the model capacity to capture the loss is out of scope of the paper, our choice was not to show any full simulation of the ozone loss by Reprobus. The description of the model (section 2.1, p 314, l 21) has been revised as follows:

The passive mode of REPROBUS has no chemical component and considers only a single “passive ozone” tracer. This tracer is initialised identically to the ozone analysed by ECMWF at the beginning of the simulation and is then transported by the winds without any chemical production or loss. This allows diagnosing the ozone chemical loss by making the difference between the “passive ozone” column calculated every day by the model over each station and the ozone column measured by SAOZ. In addition, a REPROBUS “gas phase” simulation with full chemistry but without heterogeneous processes is carried out to quantifying the contribution of gas-phase chemistry (without PSCs) to the observed ozone loss. The « gas phase » simulation employs the comprehensive chemical package of REPROBUS, with 45 species and 120 reactions and photolytic processes. Absorption cross-sections and kinetics data for this run are all based on the latest JPL compilation (Sander et al., 2011).

Additional ref

Sander, S. P., J. Abbatt, J. R. Barker, J. B. Burkholder, R. R. Friedl, D. M. Golden, R. E. Huie, C. E. Kolb, M. J. Kurylo, G. K. Moortgat, V. L. Orkin and P. H. Wine "Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation No. 17, JPL Publication 10-6, Jet Propulsion Laboratory, Pasadena, 2011

b) Comparison between observed and modelled column NO₂

Reprobus is not providing sunrise and sunset columns as observed by SAOZ but every 3 h at 00, 03, 06..... UT. The comparison with the observations at 90° SZA is thus not straightforward. Following your suggestion we have attempted to do this by interpolation. But since for example the minimum NO₂ in the early morning is not available, this leads to an overestimation of the column at 90°. The same applies in the evening where the interpolated column at 90° is strongly weighted by the max NO₂ column in the early night. The only way to provide the right information would be to run Reprobus every 10-15 minutes during the full winter which is not possible for the moment.

The section on the ozone loss/ PSC relationship needs refining. First, given that SAOZ measures total column ozone, how are ozone losses between 400-675K calculated? Second, given the importance of ozone loss at potential temperatures below 400K in some winters,

why is 400K chosen as the lower limit?

We are not calculating the loss between 460-675 K but the PSC volume between 13.5-25.5 km from ECMWF temperatures at 400, 475, 550 and 675K within 3 km thick layers at each level. We are always showing total ozone columns, which include thus all levels.

Third, what is the proposed mechanism relating sunlit VPSC with ozone loss? VPSC is an empirical proxy for activation, and sunlit vortex is an empirical proxy for ClO/photochemistry. But they do not have to occur at the same time. Finally, given the long periods of low temperatures over a large altitude range in which extensive denitrification probably occurred (e.g. Fig 3c in Manney et al (2011)), it is not clear why the authors propose denoxification as the additional parameter.

The use of the word “denoxification” seems confusing. The idea was to use it to say “absence of NO_x” not considering the mechanism responsible. But it’s obviously wrong. It has been thus changed everywhere for “denitrification”

Sunlit VPSC is indeed a proxy for ClO photochemistry. The use of sunlit VPSC instead of VPSC comes from the better relation found with ozone loss in Fig.6 and Fig.12. Indeed a large amount of PSC in early January as shown in Fig. 5 has little impact on ozone, but in contrast a smaller amount in March leads to fast depletion.

The SAOZ-based observations are consistent with the analysis in Section 5.3 of Harris et al (2010) - but without the offsetting reduction of the ozone loss at lower altitudes due to renitrification. The SAOZ measurements (Fig 9) show that the ‘column denitrification’ was similar to other years (i.e. probably zero) on Feb 1 and 15, but thereafter became progressively more of an outlier with only 2000, a winter with directly observed, extensive denitrification, anywhere close.

Indeed the deep vortex denitrification on Feb 1 in 2011 is similar to that of other cold early winter years described by Harris et al (2010). The non-zero NO₂ diurnal variation is coming from the non-denitrified high altitude layer above 25 km as seen for example by Pommereau and Piquard (1994) during EASOE. What is very different in 2011 is the late renitrification compared to all other years, even to the previous latest denitrified 1997 and 2000 winters.

The discussion of Fig. 9 p 320 has been revised as follows.

Figure 9 shows the amplitude of the NO₂ diurnal cycle, a proxy for denitrification, on each year on Feb. 1, Feb. 15 March 1 and March 15 since 1994. The 2011 denitrification on Feb 1 is similar to that observed on other cold early winters such as 1995/96, 1999/2000, 2004/2005, 2007/2008, 2009/2010 and 2010/2011 (Rex et al., 2006, Harris et al., 2010, Pitts et al., 2011, Khosrawi et al., 2011, Arnone et al., 2012 and references herein). The non-zero NO₂ diurnal variation is coming from non-denitrified high altitude layers above 25 km as observed for example from balloon measurements in the vortex in January during the cold winter of 1992 by Pommereau and Piquard (1994). But less frequent is the extension of the denitrification until early March such as in 1996, 1997, 2000 and 2011. And even less frequent is its persistence, that is the absence of renitrification until mid-March in 2000 and 2011 only, the last being the record year. The proposed explanation for that is in the unusual strength of the vortex in 2011 shown by the maximum PV gradient larger than on all previous years (Fig. 10) as pointed out by Manney et al. (2011). Such strength prevented renitrification by import of NO_y and HNO₃ rich air masses from the outside of the vortex as frequently observed in the Arctic after minor warming episodes and subsequent elongation and

reformation of the vortex. However, as for the ozone loss in Fig. 7 or denitrification in Fig. 9, there is no sign of trend in the vortex strength since 1994, but high inter-annual variability only.

Ref added

Pommereau, J. P. and J. Piquard, Ozone, Nitrogen dioxide and Aerosol vertical distributions by UV-visible solar occultation from balloons, *Geophys. Res. Lett.*, 21, 1227-1230, 1994

I do not think the shading in the figures helps. Make the lines stronger if anything.
Modified.

Minor comments

Thanks for editing. All corrected.

313, 11/14 This does not accurately reflect what Rex et al. say. We reported that the extreme cold winters were getting colder, but there was no trend in the warm winters.

Right. Sentence replaced by:

Although stratospheric climate conditions have been suggested to become more favourable for large Arctic ozone losses over the past four decades (Rex et al., 2004) and the eventuality of a large depletion in case of an unusually long cold winter recognised for long (WMO, 2011), such extreme loss was unpredicted by climate models.