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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A closer look at Arctic ozone loss and polar stratospheric clouds" *by* N. R. P. Harris et al.

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Major comments

1. The present study should distance itself from the previous one. Some figures (e.g. figures 4 and 7 are very similar to figure 2 and 3 of the previous article). Some discussions (e.g. activation and ozone loss period) are also very close. The authors should build on the previous study and emphasize the new results.

For historic reasons there are similarities between parts of the two papers. This present paper contains significantly more material and analysis, including the recalculation of the relationship using the self-consistent ERA Interim analyses rather than operational products.

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Interactive Discussion

Discussion Paper

We think the presence of the information in Figures 4 and 7 helps this manuscript. However we understand the reviewer's point and we have revised the text describing them. We have merged the old Figure 4 with a new version of Figure 5 so that we can show the comparison between the continuous and episodic PSC cases as well as the impact of lower activation temperatures. We are also replacing Figure 7 with a new version which also includes the ozone loss rate for the same three cases.

2. The main incentive for the study is the compact relationship between integrated ozone loss and the volume of PSC as represented in figure 2. However, the error bars in the figure are barely explained nor is evaluated the effect of these error bars on the sensitivity slope of ozone loss as a function of cooling. A value of 15 DU per 1° cooling is provided without error bars. If the error bars are large, could it affect the usefulness of this relationship in climate models?

Establishing Figure 2, its potential usefulness for assessing climate model results, or quantifying the error bars or the slope in Figure 2 is not the purpose of this paper – this has all been done in previous publications (e.g. Rex et al., 2006; WMO, 2007). We need to rminimise overlap with those publications here. The only reason for repeating Figure 2 in this publication is to update it and to base it on a homogenous meteorological data set. The present paper focuses on explaining the compactness and linearity of the relation rather than on establishing it or analyzing its uncertainty.

3. The discussion is very qualitative and sometimes fuzzy as detailed below. An explanation based on the factors that mainly affect the temporal evolution of ozone and other major constituents, as expressed by the continuity equation would give more weight to the demonstration.

The discussion has been deliberately made fairly qualitative. The basis for the work is that both observations and 3D CTMs show the existence of a linear, compact relation between ozone loss and VPSC. The aim of this paper is to unravel the important factors already present in the model as well as in the atmosphere rather than to show it can be

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reproduced quantitatively – that has already been done in more sophisticated models that can reproduces the observed relation. We therefore chose to use a relatively simple tool (a photochemical box model with idealized trajectories) to identify the main processes rather than a more complex model which is harder to diagnose.

We have made a number of revisions to the paper which make the discussion more quantitative, but it is hard to go as far as reviewers 2 & 3 seem to be suggesting. We have also made the rationale for this clearer at the end of section 1.

**Sensitivity figures demonstrating cancelling effects of e.g. the ozone loss and deactivation would be useful.

Figure 7 shows exactly that.

4. The whole methodology is based on a photochemical box model run along idealized trajectories. How these trajectories mimic the real atmosphere should be better demonstrated in the article.

For the same reasons discussed above, we do not want to compare too closely to the real atmosphere as that is inappropriate for a photochemical box model on idealised trajectories. We have chosen conditions that broadly resemble the Arctic. We have tried to make the details of the trajectories used, and their relation to the real atmosphere clearer in the text.

**Some sensitivity studies (e.g. denitrification) should also described in the context of observations.

We used a degree of re/renitrification similar to that which has been observed in the Arctic (Popp et al., 2001). This is now made clearer in the text.

Detailed Comments

1. 6683, l13: Bodeker et al., 2005 should not be the only reference for the evaluation of Antarctic ozone loss in 2002. More references should be provided on this rather well

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documented event.

We are trying to make the specific comparison with the size of the Arctic losses (rather than provide a more general set of references to studies of the event), and Bodeker et al is a very good reference for that. A statement referring to Hoppel et al (2003) and Ricaud et al (2005) has been added in order to include similar information about how the vertical distribution of ozone.

6683, l19: the sentence on the capacity of 3D models to reproduce ozone loss is too vague.

We now say that they “have been continually improving”.

2. Figure 1: how is the vortex defined? The definition of the vortex should be clearly stated in the article.

The definition of the vortex (36 s-1 normalized potential vorticity) is now stated in the figure caption.

3. 6684, l8: PSC “fall” should be replaced by “sediment”.

This has been reluctantly changed as ‘sediment’ has become widely used as a verb to describe PSCs settling to lower altitudes. However, we would like to point out that the actual definition of sediment (as a verb) is “to deposit as a sediment” and that the definition of sediment (as a noun) is “Matter composed of particles which fall by gravitation to the bottom of a liquid”. (This is analogous to the meaning of precipitation as “Water that falls to or condenses on the ground as rain, snow, dew, etc.)”. Fall on the other hand is defined as “To descend freely (primarily by ‘weight’ or gravity)”, which seems a better word to describe what the PSCs are doing. (All definitions from the Oxford English Dictionary.)

4. 6685, l15: Figure 2 that is introduced here needs a detailed explanation on how the various quantities and their error bars are derived. An explanation is provided later in the methodology section but the organization of the article should be revised so that

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the figure and the various terms are adequately described, including the sensitivity relationship of ozone loss versus temperature.

Establishing Figure 2 is not the purpose of this paper – this has been done in previous publications (e.g. Rex et al., 2004, 2006; WMO, 2007) and we need to minimise overlap with those publications here. The main reason for repeating Figure 2 in this publication is to update it. The present paper focuses on explaining the compactness and linearity of the relation rather than on establishing it or analyzing its uncertainty.

5. 6686, l15. Provide reference for the assertion that PSC are unlikely to form above 24 km.

We have added Pitts et al., 2009 as a reference and also clarified that the 24 km refers to the altitude of the airmasses in March so that they were high earlier in the winter (say 28-30km).

6. 6686, l20-25 & discussion on PSC page 6688-6689: The discussion on Vpsc computation is somewhat fuzzy. Could the various assumptions on HNO3 and H2O mixing ratio translate into error bars in Vpsc? It is also not clear how the estimate in the article compare with estimations from CALIPSO. A much more focused discussion is needed here.

The definition of PSC formation and their exact composition is not critical to the analysis presented here. TNAT is a good proxy for the efficiency of chlorine activation. The validity of this is shown that a different proxy based on activation on sulphate aerosol makes very little difference to the results. The reason is that the proxies are highly correlated. The discussion about this has been significantly revised to make this point and the underlying arguments clearer.

7. 6687, l11-18: Provide explanation for the design of the idealized trajectories. What is the reason for the sinusoidal 6 day cycle of 20°?

That is a typical time for air to once round the vortex and the position chosen is that of

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the vortex in its preferred state. Some text has been added to make it clearer.

8. 6688, l5: how good are the diabatic heating rates calculated in the SLIMCAT model, e.g. how do they compare with N2O isopleths?

They agree well. This has been shown in Feng et al (2005). This is now stated and now referenced at this point in the manuscript.

9. Figure 3: The description of this figure is quite fuzzy.

We have amended the figure caption to define the subsiding surfaces clearer and given a reference.

(1) There is a typo on the O3 loss units (it should read 1012 mol cm-3.

No it is not a typo - it is molec cm-3. (as abbreviation for molecules). We are not giving the loss in mol per cm-3 (then the numbers would be much much smaller).

**Further, in order to show the contribution of each layer to the loss, I suggest to use DU/km.

We are adding this on the right hand x-axis.

(2) Why is there no error bar in the figure as in figure 2? What are the quantitative values of the slopes and "scatter" at the various levels?

Adding error bars would be very hard to do properly – it's either fairly arbitrary suggesting a knowledge of the uncertainties that we don't really have, or would require a comprehensive study comparable to the Harris et al., 2002 paper. However we do not think the message (higher slope at higher pressures / lower potential temperature would be different.

(3) Is it pertinent to relate the Apsc value and ozone loss at the same isentropic level, since air masses can be in the presence of PSCs at a certain level in the beginning of the winter and then descend to another level due to diabatic heating at the end of the

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winter?

As already stated in the text, the isentropic level quoted is the one in March of each winter. We assume that surfaces descend according to the heating rates calculated by SLIMCAT and average the ozone loss and PSC volume in that way. We now use the term 'spring equivalent potential temperature (eθ)' in the paper to make this is clear.

10. 6690, l1: A summary of the main heterogeneous and homogeneous chemical reactions playing a key role is needed here.

The main photochemical processes in the Arctic have been summarized in many publications and we do not think that we need to provide another description here with the resulting expansion of the paper. We are happy to follow the editor's guidance on this matter as it is relatively easy to do.

** These reactions could then be used to derive quantitative relationships for the temporal evolution of ozone and other key compounds (e.g. ClOx, HNO3).

The problem with this suggestion is that the system becomes quickly non-linear as more chemical reactions are considered. We feel that the box model is already simplifying things enough.

**In addition bromine compounds are also barely mentioned in the article. How the uncertainties on the effective BrO levels in the lower stratosphere affect the O3 loss/ Vpsc relationship?

The O3 loss/ VPSC relation is empirically derived and so does not depend on any particular assumption on the BrO abundance. The model runs shown are based on 20.2 ppt of Bry and standard Br photochemistry (as now stated in the revised model description). The calculations in Figure 7 have also been done using Bry = 24ppt (a standard 'high' Br case, possibly more realistic), and the integrated ozone losses by day 60 of the runs are found to be only $\sim$10% higher. We do not think this is large enough to justify mention in the manuscript. The assumed Bry level is now stated in

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the paper.

11. Discussions p. 6691& 6692: as emphasized previously the discussion on the cancelling effect of ozone loss and deactivation would greatly benefit from quantitative relationships.

The new version of old Figure 7 should partly address this by including the ozone loss rates as well as the ozone itself. The much higher ozone loss rates and faster deceleration in the latest case is apparent. It is extremely hard to develop quantitative relationships in such a coupled chemical system – we have looked at this suggestion but not found anything which would help.

12. 6694, l14-17: The counteracting effects should be quantified. The adjective "small" is very vague.

The photolysis frequency at 550K is actually <5% larger than at 500K – this effect was only mentioned because people had raised this as being possibly important when this work was presented at meetings. Given the influence of SZA, ozone profile, etc., we do want to give an over-simplistic quantification in the text. The effects are small.

13. 6695, l15: To what extent the sensitivity study on denitrification correspond to denitrification observed in Arctic winters? Reference should be provided.

We used a degree of re/renitrification similar to that which has been observed in the Arctic (Popp et al., 2001). This is now made clearer in the text.

14. 6697, l15: The approaches for parameterization of Arctic ozone loss according to the methodology developed in the article should be developed in a more detailed way, e.g. what would be the advantage of the second approach with respect to the first one? An evaluation of the temperature range for which the simple "scaled" relationship can hold would also be very useful.

The section has been revised a little. However we are not sure we can follow the reviewers' suggestions until such models have been developed and compared.

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