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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions" *by* M. von Hobe et al.

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Review of the paper 'Reconciliation of essential process parameters'

General comments: This paper's author list includes a very large number of the most distinguished stratospheric scientists in the world. I want to express my very high regard for these individuals, but I must recommend that this paper should be rejected, for the reasons indicated below. I wish I could be more positive. I intend this review to be constructive and helpful for a new paper that might be written instead.

Primary problems with the paper are:

1) The paper isn't suitable for a scientific publication in part because in many places it reads like a proposal rather than a scientific paper on results; it further refers often to institutions and funding programs rather than describing scientific findings. Please edit the text carefully to remove these, and provide a paper that is results-oriented rather than funding-oriented, institution-oriented, or plan-oriented. I give some specific comments below to try to help with this.

2) Since the focus is on the results of this big project, there is not enough attention to a balanced report across the work of others for this to be a review – but in a number of places it tries to suggest that it does constitute a review of the literature. This isn't so. Other work and views are frequently cited selectively, and the discussion of past work is far too limited to represent a review paper. A review paper would have to be far longer, would have to include far more references and discussion, including to work that stands in contrast to what is here when appropriate. The current paper isn't the proper approach to reporting scientific results of particular research, and it also isn't the proper approach to writing a comprehensive review paper. It therefore becomes very unclear as to what the goal is – a proposal, a scientific research paper, a review paper, etc.; in the end I think it is an unpublishable mix of the three as it stands. I indicate specifics below, and I recommend that if the authors do write a new paper that it be shortened, that its focus sharpened to the results of this project only, and that any significant conclusions be self-contained, with results shown here instead of references to other work (see next comment).

3) The paper makes strong claims regarding a number of important questions in our science, but because of its nature in many places very little justification/specific results are shown to convince the reader that the reported conclusions are justified. Please present enough detail on key points for them to be demonstrated here, rather than referring to your other papers. This would mean bringing material in.

4) In a number of places, the paper's claims seem to me to be simply incorrect. I indicate specifics below.

Detailed comments:

5) Title. Notwithstanding the title of the project, I think this paper cannot claim to have 'reconciled' enough information about stratospheric ozone loss and its links to climate to bear the title that it carries. We really do not know why the coldest Arctic winters appear to be getting colder. We really do not know how much of the observed Arctic ozone changes are linked to chlorine versus climate, or internal variability. We really have not yet seen a compelling quantitative evaluation of the ozone losses in 2011 from a first-principles sense adequate to allow us to predict the future. Estimating how much has been destroyed in 2011 falls well short of a goal to 'enhance predictability' and I see no evidence that this claim is justified. So the current title is not appropriate, because it does not describe what has actually been learned, and that's what the title of a paper must do. There are some important new results here – particularly in the particle microphysics section – but the assertions that the project has comprehensively addressed a very broad range of issues has led to imbalance and overstatement. Please use a title that reflects the content of the work if a new paper is submitted.

6) Abstract. I have many concerns about the abstract. The first 12 lines are too basic to be useful – this should be dropped, since the paper doesn't really review in sufficient detail to represent a review of the important issues of e.g. geoengineering, or ozone recovery, etc. The remarks about nucleation are helpful. The material regarding meteoritic input needs to be clearer and highlighted more strongly – that is a useful new result. I do not think there is adequate justification for the strong claims about binary aerosols; this should be removed, see comments below. If CCMs have been improved, say how and why – what is said is too vague to be useful. I do not think the statement about links of the 2011 ozone loss to climate change are justified – justify or remove.

7) Statement about 'ozone-hole' like conditions (page 30666). It is not at all obvious to me that we can expect 'ozone-hole' like conditions in the Arctic as long as stratospheric chlorine is high. . ..we have had one year that some claim was 'hole-like' but how do we know this is not just a rare exception? Also, what is your definition of 'hole-like'? What

is the uncertainty on that statement?

8) In scientific papers, it is not appropriate to refer to 'EU-funded project'. Say what was learned, not what institution provided money or the institution where the work was done. More examples are indicated below.

9) Section 2 is a brief discussion of some past work. It is highly selective, far too short, and incomplete. It gives the impression of being a review paper when it is not. As an example, the remarks at the top of page 30668 refer in a vague way to the idea that ODS is only part of the story of recovery, but gives far too little information to allow the reader to gauge what work on climate coupling actually has shown. A review would be much longer and would read very differently. Remove the appearance of being a review and be specific about what this work has shown.

10) page 30667. I don't think this section is needed. If this is a review paper, this is much too short to be comprehensive, and figure 2 is hugely oversimplified. Remove this part and the figure.

11) page 30668 contains many examples of highly selective and limited referencing of work by others. Thompson and Solomon (Science, 2002) should have been referenced on this page regarding the ozone hole coupling with SH climate if this were reviewing the subject; this paper began that line of investigation. Regarding Antarctic sea ice and links to ozone loss, this is far from proven and there are numerous papers that should have been referenced and are not, including some making the opposite claim, e.g. Sigmond and Fyfe (GRL, 2010). Is this something you are addressing in this paper? If not, why is this here? Again, it is very clear that this is not a proper review. Please restrict the focus to what new science is here and show that science better, and avoid the appearance of being broader than this paper actually is.

12) page 30668. We do not know that the trend of cold winters getting colder in the Arctic is due to climate change, yet this is stated as if it were well proven. Delete this.

13) page 30669. The section on 'major breakthroughs through previous projects and field campaigns' is unbalanced, misleading, and wrong. To say that 'virtually all major breakthroughs have been based on outstanding experimental results gathered during large scale field campaigns' is far off the mark. Theoretical studies first suggested that chlorine radical chemistry involving heterogenous chemistry would enhance ClO and were the dominant cause of the ozone hole (Solomon et al, Nature, 1986) – well before Anderson et al. measured high ClO on a large field experiment. . Laboratory studies also pre-dated field work and had shown that chlorine radicals were liberated through heterogeneous reactions (Molina et al., Tolbert et al., Science, 1987). Further, DeZafra et al. (Nature 1987) had already measured high ClO in Antarctica, and its association with ozone loss in the vertical profile, before Anderson did it in the horizontal – DeZafra et al. was a small project, and not everything important has happened in 'big projects' as suggested here. Similarly, laboratory and model studies had already demonstrated that NAT should be present (Hanson and Mauersberger, Toon et al.) before it was seen in the field, yet this is neither stated nor are the appropriate references provided So there were actually little or no 'unexpected' and surprising results in the big campaigns of the late 1980s as you claim, rather a confirmation of what had already been predicted by theory, measured in the lab, and measured by smaller field campaigns. Such confirmation was certainly important, but it was not where the major breakthroughs came from. Delete this, and delete Figure 1, which is a misleading depiction of the history for the reasons stated above.

14) page 30670. What is meant by 'most palpable'? This is an undefined term but I am guessing you mean most important or most critical to better understand. But many of the things listed were not particularly uncertain, and there are other things far more uncertain and likely to be important that are not listed, such as the Bry content of the lower stratosphere, i.e., the very important work by Salawitch. Again, this section gives the appearance of a review paper when it is actually highly selective in its focus, this paper fails to appropriately cite many excellent works by others.

15) page 30670. The issues of mixing are certainly important – but what exactly has 'reconcile' done to reconcile them? Please remove material not supported by the results.

16) page 30671. What exactly has reconcile done to reconcile the issues associated with errors in reanalysis temperatures? Or the role of mountain waves? Again, remove discussion of issues that are not supported by the text.

17) 30671. The fact that some ozone loss can occur in association with heterogenous reactions on background aerosols, and that PSCs are not necessarily a prerequisite for some chlorine activation did not originate with Drdla (2005) or Drdla and Mueller (2012). The importance of heterogeneous activation not only on volcanically enhanced sulfate/water aerosols but also on background aerosols was covered years earlier in Hanson et al., JGR 1994, whose abstract includes the following statement, well justified in the text: "Substantial direct chlorine activation and consequent ozone destruction is shown to occur due to heterogeneous reactions involving HCl for volcanically perturbed aerosol conditions at high latitudes. Smaller but significant chlorine activation also is predicted for background aerosol loadings at extreme high latitudes, suggesting chlorine activation can occur on background sulfuric acid aerosol in these regions." Please ensure appropriate reference to earlier work.

18) 30671. There is no question that reaction of HCl with ClONO2 can occur on cold background aerosols – we have known that since at least the early 1990s as noted above and it should not be presented here as a new finding. But just because binary aerosols 'can' activate some chlorine, does not mean they 'do' so often enough, fast enough, at the key times of year for ozone loss, and over large enough areas to be important for ozone depletion when compared to other processes. The question is not IF a rx can occur but how MUCH of an effect do these particles actually have not in isolation or in a few 'example episodes' but explicitly compared to other processes in a full simulation over the entire season and range of relevant latitudes and altitudes. This has not been adequately addressed in any published study, either for the Antarctic,

Arctic, or for other latitudes, contrary to the claims made here. It is important that this paper not create a controversy where none really exists. No published paper has demonstrated the claim made here that ‘clearly, the process of chlorine activation on binary aerosol could have consequences for stratospheric ozone on a global scale’. This statement should be dropped since it is speculation. Further comments on this issue follow below.

19) by the bottom of page 30672, we are 10 pages into the paper and we have yet to see any new results. This is followed by a further long section on ‘strategy and activities’. Much of this is unnecessary and distracting in a scientific paper, although it surely made a nice proposal. The paper should be greatly shortened and these sections removed.

20) page 30673. In a scientific paper please do not refer to work being done in collaboration with a particular institution, ie. Centre de spectrometrie. . ... Please remove all references to institutions and programs.

21) page 30674-end. Please remove all references to programs and funding sources, e.g., PremierX funded by ESA, Deutsches Zentrum fur Luft und Rahmsfahrt should not be needed, apparently the proper scientific reference is Rautenhaus et al.,; Lapland Biosphere Atmosphere Facility, Finnish Met Institute Arctic Research Center, etc. Keep acronyms to a minimum. Describe instrument type and mode, and list published peer-reviewed references rather than institutions per normal standards in scientific papers.

22) Mixing in CLAMS is parameterized – see page 30680. Hence, more explanation is needed of how the modeling work here provides advances in understanding on issues of mixing, which are potentially quite important as noted earlier.

23) 30681-30687. First it is suggested that Jackson and Orsolini’s method is more accurate than other approaches, then several statements are made that seem inconsistent with that claim. Please correct.

24) 30688-30693 contain the first real results of this work regarding particle physics and chemistry. Yet, I don’t see much of this material in the abstract or conclusions. Please fix that, and consider elaborating this section a bit more.

25) 30694 first suggests PSCs are key to ozone loss in this period then suggests that binaries were important. Which is it? If PSCs were present, then why turn them off and say that the reactions on binaries must be important?

26) 30695-30696. These pages should be substantially rewritten or dropped to avoid being misleading. I have read the Wegner paper and find that its content does not back up the claims as presented here and elsewhere in this paper. I have also read the earlier paper by Drdla and Mueller, which again is not sufficient. Concerns include:

a) Episodic evaluations of what might happen if PSCs were turned off, and analyses for a few early season flights as in the Wegner paper are interesting but clearly do not prove that binaries are what actually drives ozone loss. The key driver of ozone loss is how much active chlorine can be maintained in sunlight over the full vortex, not what happens episodically or at times too early in the season to matter much for ozone loss; studies of satellite data on HCl and HNO3 variability in mid-winter as in Wegner et al likewise do not prove this. Behavior in December and January, or even in early March, are not the key issue for Arctic ozone loss. If that were important we would have a lot more ozone loss in the Arctic. The chlorine that may be activated in winter does not necessarily stay activated; we have abundant examples of Arctic years with very cold winters, lots of ClO observed in winter - yet very little ozone loss. Even in Antarctica large ozone losses do not occur in August – only a little does – large losses occur when plenty of sunlight is available. This section needs to be removed or greatly revised to acknowledge these important facts and avoid drawing conclusions of ‘significant’ or ‘substantial’ effects based on studies that are only episodic and/or do not cover the important time of year.

b) Activation alone is not the whole story, deactivation is also extremely important and it is the net between these that matters. Here you are often referring to gross activation as if it represents the net - which is misleading and inappropriate. Numerous papers have shown that in the sunlit conditions that matter for ozone loss (not the winter when it doesn't), activation has to compete heavily with very rapid deactivation (see e.g., the paper by Portmann et al., JGR, 1996, also Groos et al., ACP, 2012). That is why PSCs are very important just when it matters for ozone, since faster net activation is required the more the sun is present and as ozone begins to drop. Observations in 2011 in the Arctic, along with Antarctic ozone depletion characteristics all support the view that colder temperatures and PSCs in well-lit air are required for substantial ozone loss (indeed, this paper points this out later, and the paper thus becomes extremely inconsistent on this key point). This section of the paper is not appropriate. It presents cherry-picked material rather than a balanced discussion including these fundamentals.

c) As an example among other papers, Groos et al. (ACP, 2012) show that even in Antarctica, rapid ozone loss cannot occur until activation gets substantially faster than deactivation, at temperatures that are below those for which binaries are relevant. Yet this is not acknowledged, and the statement about the Groos paper on page 30700 quotes a different and in my view less relevant conclusion of the Groos paper. State here what the Groos paper shows regarding PSCs vis a vis binaries and the role of deactivation, if any of this material is retained.

d) Because of the concerns raised above, Figure 16 is highly misleading and should be removed – activation in a day is not significant if deactivation is occurring in about a day too, so this diagram is far too incomplete and is misleading.

e) The referenced study by Wegner et al. and the one by Drdla and Mueller stack the deck by turning off PSCs; this doesn't prove binaries were actually responsible for any changes observed, even at times too early to be important for ozone loss. Whether binaries might have activated chlorine if temperatures had not been colder and PSCs had not formed is misleading if indeed PSCs probably did form and ultimately were

responsible for the chemistry. Figure 3 of the Wegner et al. paper seems to show that PSCs were probably present for the Mar 7 flight back trajectory, but the paper shows results for what would happen if PSCs are turned off and only binaries are allowed, and does not show a parallel calculation in which binaries are turned off and only PSCs are allowed, which does not seem to me to be balanced. Along with other problems, a degree or two of error in temperature will make the difference between PSCs and binaries, so great care needs to be taken in drawing conclusions on PSC versus binary chemistry. This is not even mentioned. Observations of PSCs would be required to know whether or not PSCs are present, not inferences using temperatures that may be high biased, given this extreme sensitivity. Significant bias errors in reanalyses have been shown by many papers, and one degree is a lot. For all of these reasons, I find the discussion unbalanced and misleading.

f) Any comprehensive analysis of the role of binaries would need to consider not only any role they might have in ozone loss today but also the evolution of ozone loss from the 1980s onward. In Antarctica for example, ozone loss is so fast that it is essentially saturated now – but that wasn't so in the beginning of the development of the ozone hole in the 1980s. In particular, for an atmosphere with lower chlorine levels such as at the onset of ozone depletion in the early 1980s, faster reaction rates can be expected to be more important. It is inappropriate to make broad claims about the role of binaries in ozone depletion when the evolution of the phenomenon since 1970 has not even been looked at.

g) Bottom line on binaries 30695-30696: This paper contains misleading conclusions here, in the abstract, and in the conclusions (30704-30705). Change the text to make clear that activation is not the whole story irrespective of particle type - deactivation is also key for the seasons that matter for ozone loss, and hence the competition between activation and deactivation is key, as shown by others. Change the text to make clear that no study has established a significant role in ozone depletion for binaries as compared to PSCs, both for the present day atmosphere and for the evolution of polar ozone since the 1970s. Remove the statement in the abstract (30665) about 'strong evidence for significant chlorine activation. . ...' – it is unlikely to be very significant for ozone loss so it is misleading to call it significant here; also remove the statement on page 30704 that the paper shows 'substantial evidence for the importance of chlorine activation on cold binary aerosols'. Published work to date has not established that this is 'substantial' as compared to liquid and solid PSCs for the net activation at the times and places when it matters for ozone loss in the Antarctic or the Arctic, much less that it would be consistent with the development of the phenomenon in the 1980s. The material in this paper needs to be removed to avoid being misleading.

27) 30698, change 'EU project SHIVA' to 'project SHIVA'

28) 30698-30699. How definitive is the study by Kreycy et al. (2012)? Why is this evidence deemed better than previous work? While these are interesting results, too little information is provided to allow the reader to gauge their value compared to other work.

29) 30699. The statements regarding the Laube et al. paper are interesting, but are not presented in enough detail to allow the reader to understand why this evidence is deemed better than previous work. Please elaborate or remove, also please provide quantitative information (perhaps a table?) to allow the reader to judge how big an effect is implied relative to earlier work.

30) 30700. The key finding of Groos et al. was that very low ozone mixing ratios can only be achieved when temperature are cold enough for rapid activation processes to out-compete deactivation. Add this statement if this is retained.

31) 30700. The paper by Kuttipurath et al. is interesting. Its conclusions state that recovery is currently camouflaged by natural variability, but what is said in this paper does not seem consistent with that. Please reconcile the wording.

32) 30701-20702, and figure 19. Is this material more appropriate for a model development paper by a smaller group of authors? It doesn't seem to address fundamental goals here. Please clarify why this is key or delete.

33) 30704. Please clarify why the UV changes in the Arctic do not match those of the Antarctic. Larger remaining column amounts? Shorter depletion season due to earlier/stronger final warming? Both?

34) 30704. Remarks about the SPARC effort and other papers on ClOOCl photolysis should not be first introduced in the conclusions. Please move this up to where dimer photolysis is first discussed, to ensure referencing of important work by others.

35) 30704. As pointed out above, remove statement about 'substantial evidence for the importance of heterogeneous chlorine activation on cold binary aerosol', because it is misleading; the published work on this does not 'substantially advance our understanding of the processes destroying ozone in the polar vortex' because it has not been shown to be important for ozone loss. The material just before it on 30703 and following makes a case for the key role of PSCs in Arctic ozone depletion especially in 2011, not binaries; this merits discussion since a key goal of the paper is to discuss what matters for ozone depletion in the Arctic.

36) 30704. Say how representation of polar ozone depletion and processes in global models have been improved, or delete this statement since it is too vague to be useful.

37) 30705-30706. The closing paragraphs on the 'gate to the stratosphere' and wave breaking raise new issues not addressed in the paper. I think this isn't appropriate here; elaborate what you can conclude instead. A pointer towards future work, if any, should be much briefer.

38) Figures 20 and 21 are not very useful without error bars. Uncertainties also need to be discussed in the related text on 30703-4. If errors cannot be fully quantified and could be very large, then some kind of statement needs to be given to put these results in context. As it stands, the reader can't judge if the Arctic losses were as

large as Antarctica, 80% as large, or perhaps only 30-50% as large within error bars, etc – very different implications in the various cases. This is an important issue to quantify but at present the results are only qualitative since they lack uncertainties. A basic challenge for determining Arctic ozone loss is uncertainty in the dynamics that fill in ozone that may be chemically destroyed, making quantification much more difficult than for Antarctica; this needs to be highlighted better.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 30661, 2012.