

***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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I have reviewed this paper, even though three other reviews had been submitted, because I like to follow through on my commitments and also because I was interested in learning more about RECONCILE. I am not sure why four people were asked to review this paper, but c'est la vie. I have quickly scanned the other reviews but have not studied them in detail.

I believe this paper needs a major revision. If changes are made in response to the comments below as well as the major points of the other reviews, the paper will be a fine addition to the scholarly understanding of polar ozone depletion.

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Major Issues:

1. I take exception to the notion that all of the major breakthroughs in our understanding of the processes involved with ozone depletion have resulted from large scale field campaigns, as stated in Section 2.3. While the references given on lines 15 to 18 of page 69 are important papers, other earlier papers should be cited if the intent is to give a historical overview of polar ozone depletion.

The first published account of PSCs appeared in 1929 (Stormer, Nagture, 1929). The modern understanding of PSCs was initiated by satellite observations: the early work of McCormick and Hamill should be part of the historical narrative (i.e., McCormick, M. P. et al., Polar stratospheric cloud sightings by SAM II, *J. Atmos. Sci.*, 39, 1387, 1982). The notion that reactions occurring on surfaces at low temperature would be responsible for chlorine activation was a theoretical suggestion (Solomon et al., 1986; McElroy et al., 1986) based on a wealth of available observations, albeit none involving chlorine species at high latitude. This theory was supported by many laboratory observations (which for some reason tend to get lost in the narrative) as well as the landmark ground based NOZE campaign, including key observations of the depletion of column HCl, HNO₃, and the appearance of OCIO. These papers all preceded Anderson et al. (1989). Salawitch, R. J. et al. (Chemistry of OCIO in the Antarctic stratosphere: implications for bromine, *Planet. Space Sci.*, 36, 213-224, 1988) and Salawitch, R. J. et al. (Influence of polar stratospheric clouds on the depletion of Antarctic ozone, *Lett.*, 15, 871-874, 1988) can be consulted for many possible citations as well as the understanding of polar ozone depletion prior to Anderson et al. (1989). The first ground-based FTS observations from Antarctica appeared in Farmer et al. (*Nature*, 1987); observations of elevated ClO in the Antarctic vortex were first reported by DeZafra et al., (*Nature*, 1987) and P. Solomon et al. (*Nature*, 1987).

While Anderson et al. (1989) provided the “smoking gun” that remains one of the most famous atmospheric composition measurements, it is not proper to only cite this paper, and refer to the work as a “major breakthrough”, if the goal is to provide a historical

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Discussion Paper



narrative. Much was known about polar ozone depletion before the ER-2 ventured into the Antarctic vortex!

Another glaring omission from the citation list is Toon, O. B. et al., Condensation of HNO₃ and HCl in the winter polar stratospheres, Geophys. Res. Lett., 13, 1284-1287, 1986, who first suggested denitrification could be important.

Stimpfle et al. (JGR, 2004) is also not cited; this omission is especially egregious. Section 4.3.2 states that RECONCLIE has provided a major advance in our understanding of ClO_x catalyzed ozone depletion, even though the Young et al. study is not yet submitted. Von Hobe et al. state that studies RECONCILE studies shown great consistency between field observations and the ClOOCl cross section measurements of Papanastasiou et al. (2009). The Pope et al. (2007) paper stirred the pot and led to a critical re-assessment of our fundamental understanding of polar ozone depletion. Yet at the end, J ClOOCl from Papanastasiou et al. (2009) is nearly identical to J ClOOCl found using the Burkholder et al. (1994) cross sections upon which Stimpfle's earlier analysis was based. After the investment of so much time and effort, RECONCILE reaches the same conclusion as Stimpfle's work based on the Burkholder cross sections. RECONCILE has been important, particularly in light of the Pope et al. study. But if a historical narrative is to appear in this paper, it should that that RECONCILE has confirmed the work of Stimpfle et al. and Burkholder et al. Upon revision, the statements within Stimpfle et al. regarding Keq should also be folded into the narrative.

The existence of NAT rocks was predicted by a favorite paper of mine, Salawitch, R. J et al., Denitrification of the Antarctic stratosphere, Nature, 339, 525, 1989. Given the prevalence of NAT rocks in our modern understanding of ozone loss, the figures and analysis in this 1989 paper have stood the test of time in a remarkably accurate manner. This paper should therefore also be part of the historical narrative of polar ozone depletion: indeed, the notion that NAT rocks would exist at T above the water frost point (page 89, line 24) was not only predicted in this early paper, it was set forth as the reason the polar vortex could experience denitrification in the absence of

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dehydration.

On page 92, first paragraph, the importance of the superposition of small scale T fluctuations unresolved by ECMWF is described. Lack of any citation to the work of Bruce Gary, such as Murphy, D. M. and B. L. Gary, Mesoscale temperature-fluctuations and polar stratospheric clouds, J. Atmos. Sci, 52, 1753, 1995, is another important, unfortunate omission.

I shall end my major comment #1 by stating science does not occur in a vacuum and the copious omission of reference to so many key papers should be addressed if the historical narrative thread remains in the revised paper.

2. The paper describes many “trees” yet there is fleetingly small description of the proverbial “forest”. In other words, the paper is written as a series of sonnets about this detail or that detail, with little attempt to synthesize. This paper would have much more impact if the findings of RECONCILE could be synthesized in a coherent manner.

Particularly problematic are:

2a) the notion that the RECONCILE winter was “the third warmest winter in the 21 yr period from 1989 to 2009 measured by the polar cap temperatures at 50 hPa” (bottom of page 85) yet “during this unusual winter, more PSCs were observed by CALIPSO than in the previous three Arctic seasons combined” (bottom of page 87). I believe these statements refer to the same winter. What climatology was used to decide the RECONCILE winter was so warm? Was this sorting based on T (hope not) or V_PSC (hope so)?

Figure 13 shows profiles of HNO₃, used to argue for significant denitrification between 30 Jan 2010 and March 2010. Yet, according to the T time series in Figure 7, T never got low between these two dates. This is a glaring inconsistency. Usually, denitrification is diagnosed using plots of HNO₃ vs a tracer such as N₂O. DO such plots support the interpretation of the data given in the paper? How much descent occurred between

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30 Jan 2010 and early March 2010, and does this degree of descent account for the differences in the various HNO₃ profiles. Why are the data shown as a function of potential T, and not equivalent potential T?

2b) the discussion of ozone loss by data assimilation and ATLAS. Section 3.5 states that the data assimilation technique leads to an estimate of ozone loss on the low end of values provided by other techniques. Section 4.1.3 states data assimilation “points to an underestimation in the ozone loss estimate [sic]”. Figure 15 shows ATLAS runs of ozone loss that are discussed in the text for showing little sensitivity to perturbations of model parameters, yet no attention is given to the considerable difference between the MLS-based and model based ozone loss.

What bromine level was used in the model?

How was the dimer cross section treated?

How about denitrification in the model?

Of course ozone loss should be sensitive to bromine, dimer photolysis, and denitrification. Why is this sensitivity not apparent in Figure 15 and what is causing data assimilation to consistently yield ozone loss estimates that are on the low end of ozone loss estimates from other techniques?

2c) the issue of the importance of chlorine activation on cold sulfate aerosols. I supported the publication of Drdla and Mueller 2012 as can be discerned from the public record of the reviews of this paper. Yet, it has LONG been known that temperature controls chlorine activation. I gave a talk about this in Boulder in the late 1980s or early 1990s and did not think this detail was publishable as a stand alone focus of a paper (i.e., all models showed and many observations showed ClO_x depends mainly on T_{min} during the past 10 days or ~2 weeks; see Kawa et al, JGR, 1997 for example). It has also long been known that the primary role of PSCs is to promote denitrification, which allows activated chlorine to be sustained as the returning sub warms the vortex (e.g.,

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the first Salawitch et al. 1988 paper given above). Much of the discussion in the submitted paper on chlorine activation seems misguided, as if the team is set to “prove” something that has long been known. At the same time, the harping on this point results in the paper reading as if the importance of denitrification to prolong ozone loss, which of course does require PSCs, is no longer of interest.

I understand many textbooks have “gotten in wrong” and incorrectly state that PSCs are required for chlorine activation. Something along these lines could be stated in the paper, but if one of the key highlights of RECONCILE is purported to be that cold conditions result in Cl activation regardless of particle habitat, others may view RECONCILE as “reinventing the wheel”.

3. One of the glaring problems in our modern understanding of polar ozone depletion is the inability of CCMs to provide a realistic quantification of the abundance of ozone in the Arctic vortex. The tact taken in the submitted paper is focused on the lack of predictive capabilities for specific, cold winters (e.g., page 105, lines 1 to 6). In my opinion, a diagnostic understanding of why CCMs fail to properly simulate Arctic ozone is a crucial first step for advancing our knowledge, one that is not examined in enough detail in the submitted paper. Predictive capability of Arctic ozone levels for a specific winter is far off in the future.

Discussion of why CCMs fail to provide accurate simulations of Arctic ozone should be improved. Is this problem caused by an inability of models to move enough chlorine into the model Arctic vortex? Is the problem due to the inability to activate chlorine that is present within the modeled vortex, perhaps on cold sulfate aerosols? If proper modeling of denitrification involves detailed modeling of nucleation and microphysics, can suitable parameterizations be developed to allow denitrification to be properly simulated within CCMs.

In my opinion, CCMVal2 posed the problem regarding inability of CCMs to simulate Arctic ozone in rather vague terms. The submitted paper does little to advance upon

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CCMVal2 in this area.

4. While observations have clearly shown that the coldest Arctic winters are getting colder, especially in terms of the metric important for ozone depletion, the volume of vortex area exposed to PSC temperatures (V_{PSC}), this observation raises a series of questions that I believe will drive the future of Arctic ozone research, yet are barely discussed in the paper:

To what degree are rising levels of GHGs responsible for the coldest winters getting colder?

Might the “coldest winters getting colder” and the less frequent appearance of cold winters both be related to rising levels of GHGs: i.e., as the abundance of GHGs rise, there tends to be more wave activity that makes it less likely to have a winter where the vortex is not disturbed. However, when nature conspires to provide an ozone season with an undisturbed Arctic vortex, might rising levels of GHGs lead to more radiative cooling, and hence larger V_{PSC} and more ozone depletion?

These questions were posed in Rex et al. (2006). In my opinion, these questions should be driving the CCM component of RECONCILE. Yet there is little mention of forcing by GHGs in the manuscript.

Note: the notion that GHGs could be responsible both for decreased frequency of cold Arctic winters, yet more extensive cold conditions when a year with an isolated vortex ensues, is remarkably similar to the modern understanding of the effect of GHGs on hurricanes. High resolution models show the frequency of hurricanes diminishing, due to a lessening of the latitudinal temperature gradient, yet the intensity of hurricanes that do form rises, due to larger values of CAPE (Convective Available Potential Energy) associated with the warmer tropical ocean. If the authors see fit, they are welcome to use the hurricane analogy in the revised paper.

Minor points:

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- a. Page 71, lines 10 to 16: the paper of Prather and Rodriguez, GRL, 1986 entitled “Antarctic ozone: Meteoric control of HNO₃” should be cited!
- b. Page 71, line 17: the word “cold” should appear in this bullet
- c. Page 75, line 20: which major warming is being referenced here?
- d. Page 78, line 5 to 8: Text states “We use observations of . . .”. Not clear what these observations are used for. Same comment applies to lines 16 to 18 of this section. I do not know what “stratospheric mode measurements” means on line 21 of this page. It is hard for the reader to plow through this section . . . I do not think the paper needs to state the inclination of the MLS orbit, for instance. Too much detail and at the same time, too little substance.
- e. Page 82, line 15: CCMs do not provide the only tool for predictions and projections: Multiple Linear Regressions, simplified empirical models that would not be classified as CCMs, etc can provide very meaningful projections, indeed often more useful than projections from CCMs.
- f. Page 85, line 23: should text state “late Feb.” rather than “early Feb.” ?
- g. Page 85, line 26: why the surprise? Indeed, as noted above, if the analysis were based on V_PSC, doubt the RECONCILE winter would rank as the third warmest.
- h. Page 86, line 9: should be “was affected”
- i. Page 87, top: text leaves me wanting to know more. The Match estimate of ozone loss is based on statistics of large numbers; the self Match flight is one realization. Did Markus and Peter sign off on the statement that ozone loss estimate by Match could be impaired, based on the inability to execute a self Match flight?
- j. Page 91, line 21: perhaps “dissipated” rather than “dissolved”
- k. Page 93, lines 12 and 13: I suspect the most critical parameter for denitrification is still the fraction of available CN that nucleate, as we had suggested in our 1989 paper.

This detail seems to be lost in the discussion. Also, when the new nucleation scheme is used in CLAMs, for which winter does it “compare better to observations” (line 21). Early in the paper good agreement of CLAMs with data from a prior winter is described. Can CLAMs explain NO_y for different winters using a single parameterization?

l. Page 98, top: I do not know that a talk I gave needs to be cited but, it so, the text should make clear that the suggestion for alternative ClO_x chemistry was put forth in response to the challenge to account for observed ozone loss rates by halogens if ClOOCl truly photolyzed as slow as suggested by Pope et al. I know the team understands this, but the paper leaves out this key detail.

m. Page 98 and 99, top: We’ll continue the Bry discussion in other venues, I am sure. The suggestion that the partitioning between BrO, BrONO₂, and NO₂ is not well understood is an important advance. However, I do not place much stock in the organic bromine method (top of page 99), because it has long been known that if a very large portion of bromine crosses the tropopause from something other than CH₃Br and halons, it must do so as product gases. So I would say the apparent agreement between different estimates of Bry and the organic bromine method suggests that injection by product gases is small. One of the issues that remains to be addressed is that when many folks look at the stratospheric bromine budget, they fail to account for tropospheric loss of CH₃Br, which can be considerable.

n. Page 99, 18: how much longer. Text too vague.

o. Page 100, line 18: I thought Cl+CH₄ → HCl was crucial. Why is this not mentioned?

p. Page 100, bottom: if recovery of the Antarctic ozone hole is to be discussed, please cite some of the early papers on this topic. I have not had time to go through Kuttippurath et al. 2012b, but they are not the first to write on this topic

q. Page 102, lines 4 to 14: this paragraph is, I am afraid, clear only if known

r. Fig 1: only polar ozone campaigns are given and perhaps the most important, NOZE,

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is absent. Also, the timeline should be recast either as “Illustrative timeline of polar ozone research” or else other campaigns that did not deal with the cold polar vortex, such as SPADE and POLARIS, should be added. The satellite era is shortchanged in this timeline as well.

s. Fig 6: the caption of Figure 6 is politically loaded. How many of the grey stations had funding redirected to other, nearby stations? Is it possible to be more specific regarding when these stations were dropped (i.e., “during recent years” is not nearly as specific as “since year 2010”)?

t. I do not understand how, given what is being shown in Fig 17, the reader can deduce whether a particular cross section is consistent with field data.

u. Fig 19 is a microcosm of this entire paper: clearly, a lot of work went into making a figure showing the difference in ozone for two runs of a CTM. But which run agrees better with observations. Without an observational context, this figure does not relate much information.

v. Fig 20: what is the DFA2 correlation exponent and what do we learn by examining this quantity? This figure and the accompanying text are, as I have noted above, “clear only if known”. It is obvious this part of the paper was written by a different person than other parts.

I think RECONCILE has been very successfully and has contributed important, new information to our understanding of polar ozone depletion. I support an overview paper; I think many pieces for an outstanding overview paper are present in the submitted paper, and if the editor decides to proceed with a revised paper, I wish the team good luck with addressing the concerns of this review as well as the three others.

END OF REVIEW.

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