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Interactive comment on "Temperature thresholds for polar stratospheric ozone loss" *by* K. Drdla and R. Müller

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I appreciate the authors' work on this issue, and I have several comments below that I hope will improve the paper. In particular, I feel that more needs to be done and displayed to show the robustness (or not) of the findings. My major concerns are as follows:

1) Uncertainty in kinetic parameters. An earlier excellent paper by Drdla and Schoeberl JGR, 2003 studied many of the same issues, with the same basic approach, but reached very different conclusions, i.e., that much of the ozone loss was driven by PSCs. The current paper doesn't use the JPL kinetics recommendations, which is a departure from standard practice in the field. It appears that this paper uses a selected set of gamma values, choosing a single 'fast' set for binary sulfate and a 'slow' set for

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NAT among available kinetic studies. This set of choices will have a large effect on the importance of one set of particles versus the other. The choices made don't seem to be consistent with the published kinetics literature, which has not reconciled the different lab studies, as far as I am aware. If the issue remains unresolved in kinetics studies, then a more balanced presentation would include a range across kinetics uncertainties for sulfate gammas, and another similar kinetics range for NAT. The implications of the possible ranges of gammas should then be discussed, as in the paper by Carslaw et al. GRL, 1994. That paper is mentioned briefly but it needs to be more fully acknowledged since it deals with some of the same issues dealt with here; it would also be important to be clear as to what is new here relative to that study, once the uncertainties have been examined as appropriate.

2) Role of deactivation. The paper states that deactivation of chlorine is not important, and that only activation is. Yet Table S1 suggests that Arctic and Antarctic activation are about 63% and 68% respectively in the 'fullpsc' case (more comment on that table and associated discussion is found below), while ozone loss is about 36% in the Arctic and 87% in the Antarctic for the specific year studied (also see below). Aren't these differences in ozone loss between the two hemispheres for the same activation level due to some degree to deactivation in the Arctic? If so, then how can the statement that deactivation doesn't matter be correct? The paper currently states that deactivation can't matter if solar zenith angles are less than some value, but this should be shown rather than asserted, and compared to previous work on the subject; I believe that a number of other studies have suggested that deactivation can be quite important, including Drdla and Schoeberl JGR 2003. An explicit set of calculations, explanation of the contrasts between the Arctic and Antarctic, and better discussion of prior published work on the topic is needed here.

3) Selection of single winters, and uncertainties in temperatures within those. The paper presents results for one Arctic and one Antarctic winter. Why choose only those cases? what kind of uncertainty range on outcomes occurs if different winters are

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picked? I would expect the results to be sensitive to small differences in temperature in different years. Also, what about uncertainties in the imposed temperatures themselves– an error of one or two degrees could make a big difference depending on temperature histories, and we really don't know polar temperatures to that high a degree of accuracy do we? Again, a discussion of the sensitivity to these choices is needed, and I suspect that the outcome will be very sensitive to those choices, which could change the level of confidence in the conclusions, and their application in different years.

4) Selection of a single point in time in the development of the Antarctic ozone hole. The paper presents results for the year 2000. But it is clear that chlorine levels are very high now, making the ozone loss process largely saturated. The year 2000 therefore doesn't provide a sensitive test, especially of Antarctic ozone loss, since ozone loss now runs so efficiently there. A much more useful test would be to show whether a binary-background aerosol only mechanism can be consistent with the full evolution of the Antarctic ozone hole from the late 1970s to present. Previous work (e.g., Portmann et al., JGR, 1996) presented results for background aerosols only versus observed (from SAGE) changing aerosol abundances based on satellite data, and could not reproduce the onset of the ozone hole in the 1980s using background liquid aerosols alone; enhanced surface areas (probably volcanically enhanced, but note that some of those particles are surely ternary) were shown to be important in that work for the early period. It is worth noting that background aerosols alone were shown to be sufficient to produce a year 2000 ozone hole in that study, and this ought to be acknowledged (but see comments below, particularly number 8 and 9). An explicit examination of the time dependence is needed. I doubt that this mechanism can produce enough ozone loss to develop an ozone hole early enough in the time series to be consistent with the view that background liquids are sufficient on their own.

5) How sensitive are the results obtained here to HCl and ClONO2 abundances at the start of the winter. How well do we know those, and how much could they vary

depending upon chemical conditions such as temperatures and parcel trajectories in summer/fall? I would expect that if HCl is in larger excess over ClONO2, then the role of liquid binary aerosols is reduced since you will run out of ClONO2 early, requiring full activation to occur later, in association with wave-driven exposure to sunlight. If this happens later in the winter, then that activation could be on PSCs. It is also worth noting that Drdla and Schoeberl 2003 showed evidence for September Antarctic activation on PSCs, among other studies. Again, the paper needs a discussion of how sensitive results are to the assumptions made.

6) Strong claims are made about processes and their relevance for data, but the paper doesn't present the required comparisons to observations that would be needed to back them up. The four figures presented in the main body of the paper, and much of the material in the supplement, show idealized material, with no comparisons to atmospheric data. For example, how do the results match the distributions of the Antarctic ozone loss profile? Can the upper or lower ends of the distribution help to discriminate between where different types of particles are important, and how well the ideas here match observations? Of interest would be the upper end where liquids alone may not do enough, as I believe would be consistent with findings in earlier papers.

7) How sensitive are the results to assumptions about PSCs, and how do these compare to a range of new data? There are recent data from CALIPSO (e.g., Pitts et al., ACP, 2010) that seem to me to be highly relevant beyond the references to S10 and S12, with different apparent surface areas and compositions. My understanding is that the new CALIPSO data give evidence for much more mixed cloud than earlier work, how does that affect these results? Also, in some years, PSCs are observed in September in the Antarctic (again see Pitts et al., ACPD, 2010); I would expect that to be important and discussion is needed.

8) The paper presents results in Tables S1 and S2 for ‘fullpsc’, liquid only, and liquid binary, and attempts to use this to argue that liquid binary must be responsible for most of the activation of chlorine that occurs and for the subsequent ozone loss. This is

a core matter for this paper and I don't see why it is in the supplement instead of in the main paper: it should be moved but it also needs changes. If temperatures are cold enough long enough to drive enough chlorine activation in sunlight without much deactivation, ozone loss will be extensive regardless of what kind of particle does the activation. The present table stacks the deck for binary liquids by the limited set of tests performed, and the order in which they are done. If a 'fullpsc' case had been done followed by a case with, e.g., ternaries only, or similarly a case with e.g, NAT only, wouldn't the key results have been similar – but with activation deemed to occur on very different kinds of particles (especially in the Antarctic)? If you only allow liquid binaries to be present then these will activate the chlorine when it gets cold enough – but how different would results be if the only kind of particle allowed were a different one? Just because binary aerosols could do the job alone doesn't mean that binary aerosols alone are what actually does the job in the real world. That issue has been discussed in many earlier papers, and a fuller set of references and acknowledgement of this is needed (e.g., Hanson and Ravishankara's classic 1996 JGR paper on this issue; also Solomon 1999). Also, the tables make very strong claims with no discussion of uncertainties. Surely the sensitivity to uncertainties in temperature, temperature history, HCl/ClONO2 ratios, kinetic assumptions, etc. would lead to large ranges in these numbers, but values are being presented here to the tenth percent level. Such a table, if it is presented, would need to present a range of uncertainties including the factors I describe above, and avoid 'stacking the deck' for binary liquids alone, but instead test other cases as well.

9) Does it matter? The paper is arguing for major changes in the way people do large scale modeling and analysis. But it would be important to show that it actually matters what parameterization is used. It has been known for a long time that liquids activate some chlorine at warmer temperatures than PSCs do and that they can drive important chemistry even as binaries at temperatures above 195K, especially at low altitudes (see Solomon 1999 and references in that review). Not all models include this explicitly not because people didn't know about it, but because the distinction doesn't matter much

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to ozone loss over the Arctic and Antarctic (see, e.g, and previous work has pointed out that it could matter on the margins, e.g. Hanson et al., 1994). Is the distinction significant in the face of the many uncertainties (e.g., temperature, air parcel histories and mixing, kinetic parameters, surface areas, etc.) as noted above? The paper needs to show that this parameterization would give robustly different results for ozone loss that match observations (from 1980 to present, not just in 2000), how much different, and where, and with what uncertainties, before it can expect to provide a new basis for modelling/analysis.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 28687, 2010.