

Interactive comment on “Ice condensation on sulfuric acid tetrahydrate: implications for polar stratospheric ice clouds” by T. J. Fortin et al.

T. J. Fortin et al.

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We would like to address the comments of Anonymous Referee #3 in their order of presentation.

1) Stratospheric relevance

First, we agree that the literature does not support an argument for an abundance of SAT in the stratosphere. As a result, we do not make this argument in the paper. Instead, we would argue that the presence of SAT in the stratosphere explains several observations of solid particles at temperatures above the NAT temperature and cannot be ruled out; an argument that is supported by the cited literature. However, in an effort to further clarify our position, we have changed the line in question to read “sulfate aerosols could, even if only in small numbers, exist in a frozen state.” Second, the author brings up a good point concerning the role of HNO_3 . We agree that this issue

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should be addressed and have done so by adding a discussion of this matter in a number of sections of the final version of the manuscript. For example, clarification of how our experimental conditions compare to stratospheric conditions has been added to the end of the second-to-last paragraph of section 1. Additional discussion of the possible complications due to the presence of HNO_3 has been added to section 3. Finally, to complement the reviewer's discussion of observed denitrification, we note that, on occasion, values of NO_y as low as 2ppbv have been observed (e.g., Fahey et al., Nature, 344, 321–324, 1990; Kawa et al., Geophys. Res. Lett., 17, 485–488, 1990), suggesting that HNO_3 can be at least as low as 2ppbv under extreme conditions.

2) Modeling of dehydration.

Since the modeling simulations are intended as a sensitivity study showing the relative effect of adding a new ice formation mechanism, we chose to use as the baseline the most prevalent theory of Type 2 PSC formation and dehydration, namely synoptic scale cooling. We are unaware of any existing literature describing the role of mesoscale temperature fluctuations in dehydration. Therefore, it is not entirely clear what mesoscale perturbations the reviewer believes to be important in this context. Since the duration of individual wave events is too short to allow sufficient particle growth and sedimentation, it appears necessary for temperatures to be lower than the frost point at the synoptic scale for dehydration to occur. Even below the ice frost point, small-scale temperature perturbations on the order of 1 to 2 K will not necessarily have a significant effect on the model results. For example, if the temperature remains above $T_{ice} = -3$ K, homogeneous freezing is unimportant regardless of the freezing rate. As a result, heterogeneous nucleation on SAT can play a role and the number of SAT particles will determine the ice particle concentration. Furthermore, if these ice particles are given enough time to grow (hours), the removal of H_2O from the gas phase will further suppress homogeneous freezing, again regardless of freezing rate. Therefore, in short, it is not obvious that mesoscale temperature fluctuations will allow homogeneous freezing to play a significant role in ice formation if SAT is present for heterogeneous

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nucleation.

The choice to set SAT particle number densities less than that of the background sulfuric acid aerosol was based on several factors. First, observations made during the SOLVE campaign show that the majority of the aerosol remains liquid throughout the winter (e.g., Drdla et al., 2003). Second, nucleation, whether homogeneous or heterogeneous, is a selective process, thereby making the nucleation of only a fraction of the aerosol very likely. Finally, if all of the background aerosol is frozen, homogeneous nucleation to form ice becomes impossible. This would require that another mechanism be responsible for ice formation and dehydration.

Once again, the modeling results presented here are not intended for comparison with measurements. They are intended only as a sensitivity study. Consequently, a proper measurement-model comparison would require a more detailed analysis than is possible in this paper. However, preliminary research in that direction is currently underway, in which the model temperature and ice formation mechanisms are both varied. Thus far, the best agreement between the model and SOLVE measurements occurs when ice nucleation on SAT is assumed and the model temperatures are increased, consistent with other analyses of the 1999–2000 winter. No matter how the temperature is varied, no agreement is found if synoptic-scale homogeneous ice nucleation is assumed. Therefore, there is no indication that analysis of dehydration measurements will alter the conclusions of this paper.

Finally, the baseline simulation (i.e. no SAT nuclei) in this paper is identical to the "IceFrz" run in Drdla et al. (2003). A comment to that effect has been added to the manuscript (see section 4, paragraph 5). This should be sufficient to allow these results to be compared with Drdla et al. (2003).

3) Varia

a) Abstract, line 13: The abstract has been modified to read 10^{-3} cm^{-3} . In addition, the body of the manuscript now includes a more complete discussion of the model results

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for low SAT concentrations (see section 4, paragraph 6).

b) Results and Discussion, Figure 5a: We have added a plot of the S_{ice} required for SAT dissolution to the figure as suggested. While we cannot rule out the possibility of SAT dissolution followed by ice nucleation, particularly at $T \geq 190$ K, we feel the evidence suggests vapor deposition as the mechanism being observed. A detailed discussion of this point has been added to the text (see section 3).

c) Atmospheric Implications: First, as suggested, additional details of the model have been added to the text. Second, "...some dehydration" has been modified to read "...a maximum dehydration of 0.63 ppmv". Third, while a discussion of the black points in Figure 7b above T_{ice} was already present at the end of the paragraph in question, the text has been modified slightly for further clarification. Finally, a discussion of the reasons for the less intense dehydration associated with ice nucleation on SAT has been added to the text (see section 4, paragraph 6).

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