

Interactive comment on “Ice supersaturations exceeding 100% at the cold tropical tropopause: implications for cirrus formation and dehydration” by E. Jensen et al.

E. Jensen et al.

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General Response:

The interactive comment by Azadeh Tabazadeh primarily argues that the suggested mechanism of full surface coatings of organics reducing accommodation coefficients is implausible. The primary purpose of this paper is to present the measurements of very large ice supersaturations, indicate their inconsistency with the current understanding of ice nucleation, present hypotheses for possible physical explanations, and demonstrate implications of high ice nucleation thresholds for cirrus formation and dehydration in the tropical tropopause layer. As emphasized in the revised manuscript, the sugges-

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tion that organic surface coatings might reduce accommodation coefficients for uptake of water is speculative and just one of several possible explanations for the observed large supersaturations.

Specific Responses:

Azadeh Tabazadeh argues that formation of full surface active organic coatings on upper tropospheric aerosols is unlikely. We agree that our suggestion that surface active organics reduce the accommodation coefficient is speculative and that the formation mechanism for such organic coatings is unknown. We have added discussion of the arguments for and against this hypothesis in the revised manuscript. However, we are simply presenting this mechanism as a possibility. The speciation of organics in atmospheric aerosols is poorly constrained, and the potential effects of surface active organics on water uptake at low temperature is unknown. Additional measurements and laboratory experiments are required to address these issues.

Dr. Tabazadeh suggests that surface layers of organics in excess of a monolayer would tend to turn on itself and produce a micelle. We agree that surfactants tend to form micelles; however, laboratory experiments (referenced in the manuscript) have shown that organics can form complete layers and reduce the accommodation coefficient. Again, whether this actually occurs in the upper troposphere is unknown.

The comment also suggests that discussion of thermodynamic mechanisms for suppression of aerosol freezing should be included in the manuscript. Specifically, the impact of organics on surface energies (either the ice embryo/sulfate interface in the case of volume freezing or the ice embryo/air interface in the case of surface freezing) could have a large impact on ice nucleation.

We agree that discussion of surface freezing and the potential effect of surface active organics on nucleation rates is warranted. Surface active organics would likely alter the surface energy of the ice embryo/air interface thus affecting ice nucleation rates if surface freezing dominates. As mentioned in the comment, small changes in surface

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energies can have strong impacts on nucleation. However, as with the accommodation coefficient hypothesis, this mechanism is speculative, and laboratory measurements indicating either whether surface freezing dominates or whether surfactants inhibit surface freezing are not available. We have included discussion of these issues in the revised manuscript.

Regarding affects of dissolved organics on the surface energy of the ice embryo/solution interface for volume freezing, the argument made by Koop et al. [2000] is that the ice/solution interface energy and the diffusion activation energy for a water molecule to cross the solution/ice interface are dependent on activity but are not dependent on the nature of the solute. As a result, they assert that the nucleation rate can be expressed solely as a function of water activity. As pointed out in the interactive comment by Thomas Koop, the Wise et al. [2004] paper actually strongly supports this assertion for the dissolved organics used in their study (dicarboxylic acid). Since the available laboratory data supports the Koop et al. argument, it would be unreasonable to vary the surface energy independently of activity. We have added discussion of these issues in the revised manuscript.

Lastly, the comment suggests sulfate aerosols in the the atmosphere might actually have greater purity than those used in laboratory experiments. New particle formation does occur in the tropopause region, but these new particles are generally very small and unlikely to be important for ice nucleation anyway. Also, not all laboratory experiments are subject to the contamination reported by Middlebrook et al. Experiments using gas-to-particle conversion also do not indicate inhibition of freezing for sulfate aerosols. Perhaps a stronger argument is that the direct observations of ice nucleation on atmospheric particles reported by Cziczo et al. [ref,2004] also indicate that pure sulfate aerosols freeze at the conditions indicated by the laboratory experiments.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 7433, 2004.

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