

Interactive comment on “The role of organic aerosols in homogeneous ice formation” by B. Kärcher and T. Koop

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Comment 1: Please see my comments to the paper by Jensen et al. regarding accommodation coefficient, which should apply to this paper as well. Basically, why is the accommodation coefficient a more important issue with organics as compared to the effect that they must have on the solution surface tension?

Comment 2: It may be helpful to explain why the change of surface tension of an aerosol solution, containing organics, should not be a part of your model on ice nucleation. According to the classical theory of homogeneous freezing, the free energy of ice crystal formation has an exponential dependence on surface tension (of ice against the solution), which is further raised to the power of three in the exponent. Why doesn't

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this dependence appear anywhere in your ice nucleation model?

Comment 3: A recent study by Wise et al. (JGR, 2004) shows that solutions of dicarboxylic acids, the ones which you use as a surrogate here, tend to nucleate ice at a WARMER T, than sulfate, not a COLDER T than sulfate. This observations goes against your explanation for this specific system, which is only theoretically modeled in this work. Since laboratory data is now published on this specific system, it is only fair to explain why your model only shows organics to hinder ice formation in solution. Apparently from this laboratory work, the organic solutions studied, nucleated ice more readily than sulfate. It is possible that solutions of other organics can hinder ice formation, but this one is certainly not the one to use an example for your case. Also, since the laboratory organic solutions nucleated ice more readily than sulfate, the authors may want to add more components to their model to allow for both possibilities. To properly account for all possible cases, the importance of ice-solution surface tension on nucleation needs to be a part of the theoretical treatment of ice formation from solution. In fact I find it a little odd that the authors have no parameter for this dependence in their ice nucleation model. Slight changes in this parameter (up or down) can shift the nucleation rate and/or critical ice saturation one way or another by large amounts. In the absence of consideration for this important surface tension parameter, one needs to impose all sorts of kinetic arguments to resolve the discrepancies observed between the lab and the field data regarding organics. I am not convinced, by the arguments presented in this paper, that the limitation of the water uptake is the only parameter preventing the authors from matching the observational data to the general universal relation proposed by Koop et al. (Nature, 2000) on homogeneous ice nucleation. It is possible that the model proposed by Koop et al. is too general and that the effect of surface tension, because the dependence is exponential raised to the power of 3, is far too sensitive for all different systems of homogeneous particles to be formulated into a universal equation.

I am concerned with the direction that this field is headed because a lot of emphasizes

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is placed on why the observational data gives ice saturations in the real atmosphere in excess of what is permitted by Koop et al.'s ice homogeneous freezing line because theoretically one should not see ice saturations in the air above this line. However, if one goes back to the classical theory of homogeneous freezing, where an explicit relation exists between nucleation rate and surface tension, then one can readily see why organics, for example, change the rate of nucleation of ice in solution. I am not saying that kinetic explanations are not valid, but that one should not just through the key out the door for likely thermodynamic explanations to resolve at least some of the observed discrepancies.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 6719, 2004.

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