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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice nucleation from aqueous NaCl droplets with and without marine diatoms" *by* P. A. Alpert et al.

Anonymous Referee #2

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Review of "Ice nucleation from aqueous NaCl droplets with and without marine diatoms" by Alpert et al.

This manuscript investigates 1) homogeneous nucleation of NaCl droplets and 2) heterogeneous nucleation of NaCl droplets containing marine diatoms. The measurements appear to be carefully performed, and the results are quantitative in nature. Because of this they are useful for assessing the importance of marine diatoms in ice cloud formation. However, I have several comments regarding the interpretation of the data that need to be addressed before publication in ACP.

Page 8302, line 20: "It should be noted that if the homogeneous freezing curve parameterization suggested by Koop et al. (2000b) were used in place of the one suggested by Koop and Zobrist (2009), then a significantly better agreement of our ice nucleation

data with predictions at lower aw would be achieved." This statement suggests that only the parameterization by Koop and Zobrist (2009) was used in Figure 1 to predict homogeneous freezing. However, the caption for Figure 1 suggests that the homogeneous freezing parameterization is based on Koop et al. (2000b) AND Koop and Zobrist (2009). I am confused by these two conflicting statements. Some explanation is required here.

Page 8304, line 23: "Deviations of observed ice nucleation distributions from the fits were mostly found at higher temperatures due to possible heterogeneous freezing events." There are also significant deviations from the fits at low temperatures for some of the measurements. Can the authors suggest an explanation for these deviations?

Page 8306, line 3: "however, this effect is not critical for deriving Jhom as a function of T due to the goodness of the fit in Fig. 2 and the insignificant number of heterogeneous compared to homogeneous freezing events as previously discussed." I don't think the fits in Fig. 2 show that heterogeneous freezing will not impact the calculations of Jhom. Figure 2 is a fit to the entire data set; whereas, Jhom was calculated for small temperature intervals. In a small temperature interval a few heterogeneous nucleation events can drastically change the calculation of Jhom. I think a better procedure would be to exclude the first 15% of the freezing events when calculating Jhom.

Page 8305, line 9: "Within the theoretical uncertainty, JKoophom agrees with the experimental data." I do not come to this conclusion when looking at Figure 3. For example at a water activity of 0.806 the measured J values can vary from the theory by more than 5 orders of magnitude for some temperatures! Also the slopes of the measured J values are very different from the slopes of the theoretical J values at lower water activities.

Abstract: the authors suggest that homogeneous nucleation rate coefficients were in agreement with water activity theory. Similar to the above point, this statement should also be modified to reflect the differences in Figure 3.

Page 8309, line 15: “According to classical nucleation theory, omegahet and Jhet reflects an exponential dependence on temperature (Pruppacher and Klett, 1997) suggesting that heterogeneous ice nucleation due to intact and fragmented diatoms follows a time-dependent freezing process, in line with classical nucleation theory.” I do not thing that this conclusion is well supported by the data. For example, the data could follow a time independent process and still have an exponential dependence on temperature.

Abstract: “Our results confirm, as predicted by classical nucleation theory, that a stochastic interpretation can be used to describe this nucleation process.” Similar to the above comment, I think this statement is too strong and not supported since the data doesn't differentiate between the stochastic or the singular model.

Page 8312, line 7: “Assuming typical sea salt concentrations of 80 cm−3 with mean dry diameter of 200nm (O'Dowd et al., 1997) and a wet diameter of 480nm at 90% RH (Zhang et al., 2005; Lewis and Schwartz, 2006), and applying Jhom = 106 cm−3 s−1 at a temperature of 215 K (Fig. 3), Phom could reach 0.3 ice particles L−1 (air) min−1.” The authors are using sea salt concentrations measured in the boundary layer and applying these numbers to cirrus cloud conditions in the free troposphere, I think. Is there any evidence that these number densities of sea salt particles measured in the boundary layer are applicable to the free troposphere and cirrus conditions? If not, the authors should weaken their conclusions significantly.

Page 8315, line 17: “At T = 220 K and RH = 85% (Haag et al., 2003; Strom et al., 2003) applying the same diatom concentration for cirrus cloud formation assuming ice nucleation does not depend on surface area, we derive omegahet = 0.51 s−1 and Phet = 3.5 ice particles L−1 (air) min−1.” These numbers seem to be misleading. For example after 10 minutes the number of ice particles would be 35 L-1 due to diatoms, but the number density of diatoms used in these calculations was 0.1 L-1, unless I misunderstood the calculations. In this case the number of ice particles is 350 times larger than the number of ice nuclei. This comparison suggests that there is a problem

in the method used by the author to extrapolate their laboratory data to the atmosphere. If the number of ice nuclei are 0.1 L-1 then I would expect that the maximum number of ice particles would be 0.1 L-1 assuming no multiplication mechanism. Perhaps I am missing the authors point. In this case the authors should clarify their point.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 8291, 2011.