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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The Leipzig Cloud Interaction Simulator (LACIS): operating principle and theoretical studies concerning homogeneous and heterogeneous ice nucleation" *by* S. Hartmann et al.

Anonymous Referee #2

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General:

The present manuscript describes the operational principle of the Leipzig Cloud Interaction Simulator (LACIS), and at the same time presents a new method to numerically simulate homogeneous and heterogeneous ice nucleation in LACIS experiments. In that respect, the manuscript contains new material and is of great interest to the ice nucleation community. It is also definitely within the scope of ACP. I appreciate the careful description of the FLUENT/FPM model and the thoroughness of the manuscript in general, which I find well written and structured. However, I have some major con-

cerns about the interpretation of the different approaches to simulate heterogeneous freezing, and hence about the interpretation of the comparison of model results and measured data. If these concerns should not be justified, the authors should at any rate elaborate on the explanations about the respective topic.

Major comments:

1) From the data shown in Fig. 6 and Fig. 7 the conclusion is drawn that classical nucleation theory (CNT) together with the assumption of a constant contact angle fails to predict immersion freezing, whereas the simulated freezing behavior according to the parameterization given in Niedermeier et al. (2010) is in good agreement with the measured data. My concern about this comparison is that I do not see the fundamental difference of these two approaches. Throughout the paper, the reader gets the impression that two different theoretical concepts of describing immersion freezing were applied, which in my opinion is not the case. Equation (11) has been derived from equation (10) by Niedermeier et al. (2010), so the basic concept in eq. (11) is the same as for CNT with a constant contact angle (or constant parameter f_het). To me, the major difference between the two formulations is that nucleation kinetics and some IN size information has been incorporated in the fitting parameter "a". I presume that the curves in Fig. 7 were obtained with one value for f_het, so I don't see why these curves are not equally based on CNT with constant IN surface properties. Therefore, comparison of Fig. 7 and Fig. 6 raises the question if the better agreement of the simulations based on the parameterization by Niedermeier et al (2010) isn't simply due to the fact that two free parameters had been used to fit eq. (11) to the measured data, whereas for the curves in Fig. 6 nucleation kinetics had to be calculated explicitly, and the IN surface area is fixed. At any rate, the authors should make the substantial difference in the description of immersion freezing by CNT with constant contact angle and by the approach in Niedermeier et al. (2010) clearer, if there is any.

2) I suppose that a conclusion from the paper (even if not stated explicitly) would be that immersion freezing should be parameterized according to the formulation in eq.

(11)? In my opinion, the authors should discuss potential implications of their results to the description of immersion freezing more extensively, when they state that the model simulations were performed for the evaluation of different theoretical approaches to describe homogeneous and heterogeneous ice nucleation (page 25601, line 3). Does equation (11) well represent the physics of immersion freezing, or is the good agreement with the measured data rather due to a sufficient amount of free parameters (one of which incorporating the IN active surface area and the ice nucleation kinetics) in the fitting function? To answer this question, experiments with different particle sizes would be useful (whereas this possibly exceeds the scope of the present manuscript). I guess if the physics of immersion freezing is well represented by the model, the fit parameter "a" in eq. (11) should just change by the increase or decrease of the IN surface.

Minor comments:

p.25587, l.23: I appreciate the detailed description of the numerical model. However, equations (4) and (5) are difficult to understand without further explanation.

p.25589, l.15: I would mention that this calculation of S_hom relies on the assumption that each nucleation event leads to one additional frozen droplet, which is justified as long as the droplet volume is small enough.

p.25592, l.16: The term "thermodynamic effects" should be explained more precisely. Actually f_het describes the reduction of the nucleation energy barrier by the IN surface, i.e. the influence of the IN surface on thermodynamics.

p.25594, l.15: It has been stated on p. 25585, l. 18 that supersaturation in LACIS establishes due to the coupled water and heat diffusion which occur at a slightly different rate. To my knowledge the diffusivity of water is higher than the one of temperature, therefore one might raise the question why subsaturation does not establish rather than supersaturation upon cooling of the air in LACIS (in a water-based CPC, supersaturation is established with a transition from cold to warm temperatures). I suppose that

the influence of the absolute temperature and water vapor gradients prevailing in the chamber on saturation profile in LACIS is at least as important as the diffusion constants of water vapor and temperature. As the saturation profile in LACIS is of high importance for the droplet activation process, it would be beneficial to discuss the influence of diffusivities and the temperature and water vapor gradients on the resulting saturation profile a bit more. E.g. are the flow velocity and the tube diameter crucial to the formation of supersaturation?

p.25595, l.6: I suppose the critical supersaturation refers to Koehler activation of the seed particles? How is CCN activation treated for pure mineral dust particles?

p.25595, l.18: I find it difficult to conclude from the model simulation that immersion freezing is the only ice nucleation process happening at the given conditions, since the heterogeneous ice nucleation rate coefficient implemented in the model only contains the formulation for immersion freezing.

p.25597, l.28: What is the reason why the numerical model might slightly overpredict the droplet volume?

p.25598, l.24: I am not very comfortable with the term "singular model" in connection with a model involving a nucleation rate. Although I am aware that Marcolli et al. (2007) also used the term "singular" for a model assuming a contact angle distribution, my understanding of the singular hypothesis is that freezing is deterministic. Any formulation involving a nucleation rate coefficient, however, remains stochastic in nature, irrespective of the assumptions made concerning the IN surface.

p.25599, l.24: Would not CNT already predict the temperature dependence of the nucleation rate to be different for homogeneous and heterogeneous freezing? The derivative of the nucleation rate coefficient with respect to temperature is proportional to the energy barrier, which is considerably lower in the heterogeneous case. Therefore one should expect the homogeneous nucleation rate to increase at a stronger rate with decreasing temperature than the heterogeneous nucleation rate.

p.25600, l.10: This is too general a statement. The agreement between the orange and the red curve in Fig. 7 justifies the assumption concerning the constant freezing temperature and the freezing time. However, I don't see how the method of determining the fitting coefficients can be justified with this.

Technical comments:

p.25580, l.23: At this point, the meaning of “nucleation time” is not really clear to the reader.

p.25582, l.13: “atmospherically” instead of “atmospherical”

p.25582, l.17: “electrical” instead of “electric”

p.25592, l.20: I guess the fitting parameter “a” should have the units per second per square meter.

p.25599, l.5: The structure of the sentence does not make sense.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 25577, 2010.