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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.

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MAJOR COMMENTS referee # 1:

Only after getting to near the end of the paper did I realize what the authors meant by the title of the paper and that I misunderstood it until then. As it is, the title suggests that the paper will address the physical concept of the LACIS instrument and provide details about its realization. To some extent that is true but it is not quite reflective of

what the paper is about. LACIS has been written about extensively before and the operating principle is not detailed in this paper much beyond what has been already written elsewhere. In fact, the description given here relies on references to previous papers. This paper describes a numerical model of processes within the instrument and compares model predictions with measured ice crystal formation within the instrument. If the paper really set the goal of proving the validity of the various aspects of the operating principle of LACIS one would have wanted to see empirical proof of instrument characteristics. Perhaps that was already given in earlier papers.

In order to avoid misunderstandings we changed the title to "Homogeneous and heterogeneous ice nucleation at LACIS: operating principle and theoretical studies." The three main foci of the present paper are the description of (1) the physical setup and operating principle of LACIS for investigating homogeneous and heterogeneous ice nucleation (especially immersion freezing in the latter case), (2) the introduction of the numerical model developed to design and interpret the experiments at LACIS, and (3) the interpretation of actual experimental results by comparison with ice nucleation theory (Classical Nucleation Theory and a CNT-based parameterization). This rather theoretical paper and the Niedermeier et al. (2010) are linked closely. In Niedermeier et al. (2010) mainly the experimental results are presented, whereas in this paper for the first time the numerical model FLUENT/FPM (Computational Fluid Dynamics (CFD) code FLUENT, Fluent Inc., 2001) combined with the Fine Particle Model (FPM, Particle Dynamics GmbH, Wilck et al., 2002; Whitby et al., 2003) is introduced, as extended to deal with ice nucletion. The coupled fluid and particle dynamical processes taking place in LACIS are illustrated including the presentation of the temperature, supersaturation, droplet-/ice particle mass fraction, and nucleation rate profiles. As extended, the numerical model accounts for both homogeneous and heterogeneous ice nucleation separately. Furthermore, the validity of assumptions made for the CNT-based parameterization of immersion freezing in Niedermeier et al. (2010) is discussed.

Considering your concerns we changed the title of section 2 to "Leipzig Aerosol Cloud Interaction Simulator for ice nucleation" and we also modified the last paragraph of the introduction to: "The three main foci of the present paper are the description of (1) the physical setup and operating principle of LACIS for investigating homogeneous and heterogeneous ice nucleation (especially immersion freezing in the latter case), (2) the introduction of the numerical model developed to design and interpret the experiments at LACIS, and (3) the interpretation of actual experimental results by comparison with ice nucleation theory (Classical Nucleation Theory and a CNT-based parameterization). This rather theoretical paper and that of Niedermeier et al. (2010) are linked closely. In Niedermeier et al. (2010) mainly the experimental results are presented, whereas in this paper for the first time, the numerical model FLUENT/FPM (Computational Fluid Dynamics (CFD) code FLUENT, Fluent Inc., 2001) combined with the Fine Particle Model (FPM, Particle Dynamics GmbH, Wilck et al., 2002; Whitby et al., 2003) is introduced, as extended to deal with ice nucletion. The coupled fluid and particle dynamical processes taking place in LACIS are illustrated including the presentation of the temperature, supersaturation, droplet/ice particle mass fraction, and nucleation rate profiles. As extended, the numerical model accounts for both, homogeneous and heterogeneous ice nucleation separately. Furthermore, the validity of assumptions made for the CNT-based parameterization of immersion freezing in Niedermeier et al. (2010) is discussed." (p.25581, l.7 et seq.)

Here, impressive precision is quoted for the temperature control but this is not translated into measured accuracies of air temperature as a function of time and how it may undergo transients changes. Flow rate, the thickness of the ice coating on the walls, deviations from laminar flow are some of the issues. What happens when vapor deposit on the ice walls grows in dendritic or other complex form?

It should be noted, that both model calculations and experiments are carried out assuming steady state conditions. In other words, boundary conditions are held constant in both model and experiment. However, it is correct that the building up of the ice layer on the tube walls may introduce an undesired transient behavior. The initial ice layer is generated before each experiment and grows during the experiment. Possible effects of the ice growth are (1) an increased flow velocity inside LACIS, (2) time dependent heat transfer to the tube walls, (3) a disturbance of the laminar flow profile, and (4) the splintering of small ice crystals from the ice layer. Effects (1) and (2) are observed for longer measurement times, and experiments are stopped as soon as they become noticeable. Furthermore, experiments are multiply repeated, performed in different sequences concerning different wall temperature settings, and show similar results. Disturbances of the laminar flow profile (3) influence the stability of the aerosol beam at the center of LACIS. This effect is directly observable in the optical particle counter underneath LACIS but occurs a lot later than effects (1) and (2), i.e. measurements are usually terminated before this effect occurs. Towards the end of a measurement it can appear that ice crystal parts break off of the ice layer covering the wall inside LACIS (4). For these differently sized and oriented ice particles, the scattering signals at the OPC could be discerned from those of desired particles so that an exact determination of the ice fraction is possible. However, as soon as this effect occurs the experiment is stopped. In summary, experiments are terminated as soon as transient effects start to occur.

Following explanations are added at the end of section 2.4: "It should be noted, that both model calculations (see below) and experiments are carried out assuming steady state conditions. In other words, boundary conditions are held constant in both model and experiment. However, the building up of the ice layer on the tube walls may introduce an undesired transient behavior. The initial ice layer is generated before each experiment and slowly grows during the experiment. Possible effects of the ice growth are (1) an increased flow velocity inside LACIS, (2) time dependent heat transfer to the tube walls, (3) a disturbance of the laminar flow profile, and (4) the splintering of small ice crystals from the ice layer. Effects (1) and (2) are observed for longer measurement times, and experiments are stopped as soon as they become noticeable. Furthermore, experiments are repeated multiple times, performed in different sequences of the different wall temperature settings, and show similar results. Disturbances of the laminar flow profile (3) influence the stability of the aerosol beam at the center of LACIS. This effect is directly observable in the optical particle counter underneath LACIS but occurs much later than effects (1) and (2), i.e. measurements are usually terminated before this effect occurs. Towards the end of a measurement it can appear that ice crystal parts break off the ice layer covering the wall inside LACIS (4). For these differently sized and oriented ice particles the scattering signals at the OPC could be discerned from those of desired particles so that an exact determination of the ice fraction is possible. However, as soon as this effect occurs the experiment is stopped. In summary, experiments are terminated as soon as transient effects start to occur."(p.25586, l.13 et seq.)

The authors tackled the difficult task of modeling processes within the instrument, both with regard to conditions and with regard to evolution of the three phases of water. This is a major achievement. However, a limitation of the theoretical description isn't specifically stated (or I missed it). At this stage, the model is formulated for monodisperse uniform chemical aerosol and for a single (selectable) mode of ice nucleation. Neither the theoretical formulation nor the practical implementation are described for dealing with multiple processes acting at the same time. These may follow in the future, one can surmise.

The model as described in the manuscript features two different ice nucleation modes. Specifically homogeneous and heterogeneous (immersion freezing) ice nucleation are considered. The following text is already in the paper, for example:
"The newly-developed phase transition model, which transfers particles from the

seed particle-droplet mode to either homogeneous or heterogeneous ice mode, is implemented in the moment dynamics equations via the respective sink/source terms $\vec{S}^k_{\mathrm{hom},i}$ and $\vec{S}^k_{\mathrm{het},i}$." (p. 25589, l. 11-14)
Consequently we are able to distinguish between ice formed via homogeneous and heterogeneous nucleation as shown e.g. in Fig. 2, Fig. 5 and Fig. 7. As a result, effects of competing processes (here homogeneous and heterogeneous ice nucleation at low temperatures) can be analyzed (see Fig. 5 and Fig. 7). Consideration of additional heterogeneous freezing modes is straightforward if a theoretical expression is given, however this is not within the scope of this paper. Choice of the nucleation rate coefficient is arbitrary so that different expressions can be tested and compared as done in Fig. 6 and Fig. 7. To make the advantages of the newly-developed phase transition model clear, we changed the sentence (p. 25590, l.6-8) to:
"Consequently, the concept outlined above facilitates the distinction between ice formed via homogeneous and heterogeneous ice nucleation and effects of competing processes can be analyzed. The concept does not depend on any specific homogeneous and/or heterogeneous nucleation rate coefficient, so different coefficients, e.g. those discussed below, can be implemented and tested."
Similar to the experimental investigations, we assume multicomponent (mineral dust core with an ammonium sulfate coating) and monodisperse seed particles. The following text passage is modified:
"In the calculations the seed particles are treated as multicomponent and monodisperse, consisting of an insoluble core (e.g. ATD) and a soluble coating (e.g. $(NH_4)_2SO_4$)." (p. 25589, l.22-23)
Extension of the model towards polydisperse internally- and/or externally-mixed particle populations is possible, however far beyond the scope of this paper, and beyond the current stage of experiments.

In dealing with heterogeneous ice nucleation, the authors take the careful approach of considering both CNT and the singular description. The latter was shown by

Niedermeier et al. (2010) to provide the better explanation of observations.

In the present paper, no singular model was applied (see second comment on 2 December 2010).

Their formulation takes the form of an empirical fit to results obtained and is here used in analyzing the same instrument for the same type of aerosol. Thus, as far as I can see, the results here presented are really summed up in lines 8-10 of page 25600, while the sentence following that in lines 10-12 is not fully justified.

We agree with the referee concerning the fact that validity of the parameterization concept itself is not verified in this study. Therefore, further investigations analyzing the immersion freezing behavior as function of temperature (wider temperature range than investigated here), IN surface (varying ice nucleus sizes), IN structure and chemical composition and ice nucleation are fundamentally necessary.
Consequently, we change following sentences:
1) "Finally, reviewing the assumptions made during the derivation of the CNT-based parameterization for immersion freezing, it was found that the assumption of constant temperature during ice nucleation and the chosen nucleation time were justified, underlining the applicability of the method to determine the fitting coefficients in the parameterization equation." (p.25578, l.25)
2) "Consequently, the method assuming constant temperature during ice nucleation and the chosen nucleation time for determining the fitting coefficients in the CNT-based parameterization equation are justified and valid. For verifying the parameterization concept itself, further investigations analyzing the immersion freezing behavior as function of temperature (wider temperature range than investigated in the present paper), IN surface (varying ice nucleus sizes), IN structure and chemical composition and ice nucleation time are fundamentally necessary." (p.25600, l. 10 et seq.)

3) "Finally, reviewing the assumptions concerning constant temperature and ice nucleation time made in Niedermeier et al. (2010) when deriving a CNT-based parameterization for the nucleation rate coefficient in the immersion freezing mode, the good agreement between parameterization and simulation results shows that both assumptions were justified. This underlines the applicability of the method to determine the fitting coefficients in the CNT-based parameterization equation." (p.25601, l.26 et seq.)

A fundamental issue of ice nucleation can also be raised. Equation (11) presents the singular model as a rate function, i.e. time dependent. This is in contradiction with the basic notion of the singular theory. A rate function can be applied here because temperature is changing at a fixed time rate, so f(T) can be substituted by g(t). However, a change in the transformation function T(t) would require the constants of (11) to be changed.

In the present paper, no singular model was applied (see second comment on 2 December 2010).

<u>MINOR POINTS:</u>

page/line 25579/0-8: Why are all-ice clouds excluded?

To avoid misunderstandings, we modified the sentence. "Ice containing clouds have an impact on the Earth's radiative balance by scattering and absorbing solar and terrestrial radiation (Zuberi et al., 2002; Hung et al., 2003)." (p.25579, l.1-3)

25579/18: Immersion nuclei do not have to be CCN; they can enter cloud droplets by (passive) scavenging
25579/20: What is the importance of quoting Megahed (2007) here? The statement is generally accepted as is - what does the reference add to it?.

It is not explicitly stated in this text that the ice nuclei have to act as CCN initially. This pathway is only one of several possibilities how an ice nucleus becomes immersed in a droplet. The text passage is change to:
"A partly insoluble aerosol particle acts initially as cloud condensation nucleus (CCN) or becomes immersed after collision in a droplet. Due to temperature decrease, ice nucleation takes place directly at the IN surface and induces the freezing of the supercooled droplet." (p. 25579, l.17-20)
Usually, the fact that IN/all seed particles act initially as CCN before freezing can occur, yields for immersion freezing investigations at LACIS. Concerning the quotation of Megahed (2007) you are completely right, we removed it.

25579/28: IN what sense are the IN "effective"?

To avoid unclarity the world "effective" is deleted, so the sentence runs:
"Various field observations of droplet freezing through heterogeneous ice nucleation show that insoluble substances, especially mineral dust particles, act as IN in the atmosphere (DeMott et al., 2003a,b; Sassen et al., 2003; Cziczo et al., 2004; Richardson et al., 2007; Seifert et al., 2010)"

25584/22: How certain are the authors about the efficacy of the water/ice discrimination? Couldn't the tail of the narrow distribution (assumed to be water) be due to ice? Couldn't some part of the broad distribution (ice) be due to water droplets? It would

be useful to have some quantitative assessment of this potential overlap, especially if experiments are to be conducted at higher temperatures or with polydisperse/mixed composition aerosol. The mention of future improvements in this regards adds to the feeling that the current approach has definite limitations in acuity.

The issue raised here is discussed in considerable detail in (Niedermeier et al., 2010) and in our opinion beyond the scope of the present paper. In (Niedermeier et al., 2010) (p.3607, third paragraph, right column) the following argumentation is given "Firstly, the distinction between seed particles (coated or uncoated ATD particles), supercooled water droplets and ice crystals is not straightforward. However, the optical signal which originates from the seed particles is smaller than signals resulting from the droplets/ice crystals and is clearly distinguishable from them. Under the given conditions inside LACIS, the spherical droplets activate and grow (or evaporate) to similar sizes resulting in a narrow size distribution. In contrast, the growth of the ice crystals results in nonspherical shapes, and leads to optically much broader size distributions in comparison to the droplets. This behavior is utilized to distinguish between droplets and ice particles."

25585/27 and following text: How could contact nucleation be identified? What if ice crystals formed in the first place, were evaporated and formed a second time by other mechanisms?

In principle, ice crystals cannot evaporate under the investigated conditions, because, after being formed, they never experience an ice-undersaturated environment.
Since we have not been operating LACIS analyzing contact nucleation yet and we do not have experience in this field, we decided to remove this part (p.25586, l.2-5) explaining contact freezing modes of operation from the current paper.

25586/6: It seems redundant to talk about ice supersaturation when water supersaturation is specified.

We deleted ice supersaturation in this sentence.

25593/17: Are two decimals justified?

There are not two decimals in line 17. Nothing is changed in the manuscript.

25593/11: "Section" is used both for parts of the apparatus and for parts of the paper. Not a source of major confusion but if possible, it should be avoided. Perhaps 'segments' or 'stages' could be used for the apparatus.

We would like to continue using the word "section". However, we made sure that the actual meaning can easily be deduced from the context (e.g. adding words such as tube, freezing or the specific number of the LACIS tube section).

25593/22: Again, is two decimal accuracy justified and needed?

We prefer to stick to the two decimals as this corresponds to the value used in the model. Nothing is changed in the manuscript.

25594/1: In what sense are the temperature profiles 'inhomogeneous'?

To make the meaning clear "inhomogenous" was changed to "spatially inhomogenous".

(p.25594, l.1)

25594/11: This definition of the temperature error seems highly arbitrary.

The wall temperature error of 0.3 K is derived from the temperature fluctuation of the water jacket refrigerant enveloping a tube section due to temperature regulation of the respective thermostat. The following sentence is added to explain the wall temperature error in more detail:

"The wall temperature error of 0.3 K is derived from the temperature fluctuation of the water jacket refrigerant enveloping a tube section due to temperature regulation of the respective thermostat." (p.25584, l.1)

25594/21: "... version b ..." is not used in section 3.2

In section 3.2 the following is stated: "For determining the homogeneous and heterogeneous ice nucleation rate coefficients to be used in FLUENT/FPM, two different model approaches are adopted: (a) Classical Nucleation Theory is applied for both homogeneous and heterogeneous ice nucleation, and (b) CNT is used for modeling homogeneous nucleation, but immersion freezing is described by implementing a CNT based parameterization derived from prior LACIS measurements (Niedermeier et al., 2010)."
To clarify what is meant with "... version b ...", the following bracket is included. "(CNT for modeling homogeneous ice nucleation and CNT-based parameterization for immersion freezing)" (p.25594, l.21)

References

Cziczo, D. J., Murphy, D. M., Hudson, P. K., and Thomson, D. S.: Single particle measurements of the chemical composition of cirrus ice residue during crystal-face, J. Geophys. Res.-Atmos., 109, D04201, doi:10.1029/2003JD004032, 2004.

DeMott, P. J., Cziczo, D. J., Prenni, A. J., Murphy, D. M., Kreidenweis, S. M., Thomson, D. S., Borys, R., and Rogers, D. C.: Measurements of the concentration and composition of nuclei for cirrus formation, Proc. Natl. Acad. Sci. USA, 100(25), 14655–14660, 2003a.

DeMott, P. J., Sassen, K., Poellot, M. R., Baumgardner, D., Rogers, D. C., Brooks, S. D., Prenni, A. J., and Kreidenweis, S. M.: African dust aerosols as atmospheric ice nuclei, Geophys. Res. Lett., 30(14), 1732, doi:10.1029/2003GL017410, 2003b.

Hung, H. M., Malinowski, A., and Martin, S. T.: Kinetics of heterogeneous ice nucleation on the surfaces of mineral dust cores inserted into aqueous ammonium sulfate particles, J. Phys. Chem. A, 107(9), 1296–1306, 2003.

Niedermeier, D., Hartmann, S., Shaw, R. A., Covert, D., Mentel, T. F., Schneider, J., Poulain, L., Reitz, P., Spindler, C., Clauss, T., Kiselev, A., Hallbauer, E., Wex, H., Mildenberger, K., and Stratmann, F.: Heterogeneous freezing of droplets with immersed mineral dust particles — measurements and parameterization, Atmos. Chem. Phys., 10, 3601–3614, doi:10.5194/acp-10-3601-2010, 2010.

Richardson, M. S., DeMott, P. J., Kreidenweis, S. M., Cziczo, D. J., Dunlea, E. J., Jimenez, J. L., Thomson, D. S., Ashbaugh, L. L., Borys, R. D., Westphal, D. L., Casuccio, G. S., and Lersch, T. L.: Measurements of heterogeneous ice nuclei in the western united states in springtime and their relation to aerosol characteristics, J. Geophys. Res.-Atmos., 112, D02209, doi:10.1029/2006JD007500, 2007.

Sassen, K., DeMott, P. J., Prospero, J. M., and Poellot, M. R.: Saharan dust storms and indirect aerosol effects on clouds: Crystal-face results, Geophys. Res. Lett., 30, 1633, doi:10.1029/2003GL017371, 2003.

Seifert, P., Ansmann, A., Mattis, I., Wandinger, U., Tesche, M., Engelmann, R., Muller, D., Perez, C. and Haustein, K.: Saharan dust and heterogeneous ice formation: Eleven years of cloud observations at a central European EARLINET site, J. Geophys. Res.-Atmos., 115, 13, doi:10.1029/2009JD013222, 2010.

Stratmann, F., Kiselev, A., Wurzler, S., Wendisch, M., Heintzenberg, J., Charlson, R. J., Diehl, K., Wex, H., and Schmidt, S.: Laboratory studies and numerical simulations of cloud droplet

formation under realistic supersaturation conditions, J. Atmos. Ocean. Tech., 21(6), 876–887, 2004.

Whitby, E., Stratmann, F., and Wilck, M.: Fine Particle model (FPM) for FLUENT, Manual, 2003.

Wilck, M., Stratmann, F., and Whitby, E. R.: A fine particle model for FLUENT: Description and application, Proc. Sixth Int. Aerosol Conf., Taipei, Taiwan, Chinese Association for Aerosol Research in Taiwan/International Aerosol Research Assembly, 1269–1270, 2002.

Zuberi, B., Bertram, A. K., Cassa, C. A., Molina, L. T., and Molina, M. J.: Heterogeneous nucleation of ice in $(NH_4)_2SO_4$-H_2O particles with mineral dust immersions, Geophys. Res. Lett., 29(10), 1504, doi:10.1029/2001GL014289, 2002.

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