

Interactive comment on “Kaolinite particles as ice nuclei: learning from the use of different types of kaolinite and different coatings” by H. Wex et al.

Anonymous Referee #2

Received and published: 15 January 2014

In this study, kaolinite samples from two different sources (Fluka and CMS) were examined with respect to their ability to act as ice nuclei. Kaolinite particles were either investigated uncoated or with levoglucosan, succinic acid or sulfuric acid coatings. Measurements were carried out with two different setups (CFDC and LACIS) in subsaturated and supersaturated conditions with respect to water. The observed heterogeneous freezing of the coated particles below water saturation is interpreted as immersion freezing of solution droplets. This study provides an important contribution to the understanding of heterogeneous ice nucleation of coated particles. The authors have performed a profound evaluation of the data. Nevertheless, there are some questions regarding the evaluation procedure that should be resolved prior to publication in ACP (see general and specific comments). Also, I think that the new definition of

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condensation freezing proposed by the authors is not needed and might lead to more confusion than a clarification.

General comments:

I have a mature concern with the way condensation freezing is first redefined in the introduction section and then – based on this new definition – rejected as a separate nucleation mode in the summary and conclusions section. The generally accepted view of condensation freezing refers to the sequence of events of condensation of liquid water on an IN directly followed by freezing (e.g. Fukuta and Schaller, J. Atmos. Sci., 39, 648-655, 1982). It is thought to occur when starting from subsaturated conditions, water saturation or supersaturation is reached. This is in contrast to the new definition suggested in this article, where condensation freezing is considered to occur below water saturation in haze droplets. It might be the case that condensation freezing is a special case of immersion freezing with a comparable nucleation efficiency, but this conclusion should not be reached by the artifice of a redefinition of condensation freezing.

Another major point is the application of a stochastic approach to the Fluka kaolinite. This approach might be well justified for the CMS kaolinite but not for the Fluka kaolinite sample. Pinti et al. (2012) observed more than one freezing peak for this kaolinite sample for emulsions investigated by DSC. Moreover, Atkinson et al. (2013) revealed that this sample contained 5% feldspar which might be responsible for a frozen fraction < 5%. All these facts are discussed in the manuscript. Nevertheless, a stochastic formulation of the nucleation rate is applied for the analysis of the data. This seems to be inconsistent.

The physical basis of Eq. 2 ($J_{het} = A \exp(BT)$) is unclear. It seems to be just a fitting function and does not show the temperature dependence expected for an activated process given by the Arrhenius equation ($J_{het} = A \exp(-B/kT)$). Nevertheless, this equation is inserted in Eq. 3 as if it described a physical correct temperature dependence of

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the nucleation rate. In view of this shortcoming, an analysis of the data as suggested in the comment from Alpert and Knopf is highly recommended.

Specific comments:

Page 30314, lines 2-14: This seems to be a new definition of condensation freezing (see general comments). I advise the authors to stick to the common notion of condensation freezing and to use for heterogeneous freezing of solution droplets containing an IN the term immersion freezing (of solution droplets) as has been done in other studies before (e.g. Hoose and Möhler (2012); Zobrist et al., 2006, *Atmos. Chem. Phys.*, 6, 3115–3129; Zobrist et al., 2008, *J. Phys. Chem. A*, 112, 3965-3975; Koop and Zobrist, 2009).

Page 30314, lines 13- 14: also refer to the recent review by Ladino et al. (*Atmos. Chem. Phys.*, 13, 9745–9769, 2013) about contact freezing.

Page 30317, lines 12-17: Can you comment on the effect of the discharger? This might be interesting information for other groups using similar setups.

Page 30327, lines 19-21: It should be explained in more detail how Koehler theory was used to convert from Sw to aw.

Page 30329, line 5: Inspecting Fig. 4, it does not seem to be justified to use the same A and B average values (ACMS and BCMS) for Fluka kaolinite treated with sulfuric acid and CMS kaolinite.

Page 30331, lines 15-17: The second part of this sentence should be removed (see general comments).

Page 30333, line 18 - page 30334, line 2: I do not agree with the statement that the term immersion freezing has so far been reserved for insoluble particles immersed in diluted droplets. Hoose and Möhler (2012), Zobrist et al. (*Atmos. Chem. Phys.*, 6, 3115–3129, 2006), Zobrist et al., (*J. Phys. Chem. A*, 112, 3965-3975, 2008) and Koop and Zobrist (2009) have used the term immersion freezing for dilute and concentrated

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solutions. I agree with the authors that there is indeed no need to distinguish between immersion freezing of dilute and concentrated solutions and that for both cases the term immersion freezing should be used. The term condensation freezing should therefore be kept for the process that implies the sequence of condensation of liquid water and freezing (see general comments).

Figure 2: The RH conditions for the data shown in this figure should be explicitly stated.

Figure 3: The color code should be improved. CMS particles should be given a different color from Fluka particles. Fluka 300 nm and Fluka 700 nm particles should be given different colors.

Figure 4: The average values AFluka, BFluka, ACMS, BCMS should be given as lines in Fig. 4.

Figure 5: it is difficult to assign freezing curves to the specific experimental conditions given in Table 1. Either the readability of the figure has to be improved or the data should be in addition given in a Table for selected nucleated fractions.

Technical comments:

Page 30314, line 15: second end bracket is missing.

Page 30330, line 8: “deliquesce” instead of “deliquescence”.

Figure 1: spelling error: “dilution” instead of “dillution”.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 13, 30311, 2013.

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