Atmos. Chem. Phys. Discuss., 9, S476–S480, 2009
www.atmos-chem-phys-discuss.net/9/S476/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Freezing of water droplets colliding with kaolinite particles" *by* E. A. Svensson et al.

Anonymous Referee #1

Received and published: 27 February 2009

General Comment

This study reports on some interesting, if somewhat poorly quantified, studies of contact freezing using an electrodynamic balance device. New experimental observations of contact freezing are needed and use of this method for this purpose is welcomed. The experiments indicate an important quantitative impact of qualitatively-defined changes in humidity on this freezing process. While these results are worth reporting, the conclusions and implications stated are not justified, in my opinion. Many additional details would be required to bolster conclusions that the "high" humidity results are most relevant to contact freezing in clouds in the atmosphere or that current model parameterizations of contact freezing by kaolinite are accurate. I gather that such additional detail as an estimate of humidity is not accessible. So, although the

authors attempt to qualify the results and the need for more quantitative data in their conclusions, I feel that they must be clear on the speculations involved in stating such. The interesting results should simply be reported and the needs for studies to further elucidate mechanisms and their relevance to clouds should be stated. Personally, I would only wonder if the observations might be artifacts of the methodology, a key factor being the nature of the conditions to which the particles are exposed when humid, dry air, and particle streams are mixed.

Specific Comments

1) Abstract: a. The abstract uses the terms "dry" and "high relative humidity" in a relative sense. Since these terms cannot be quantified, some statement is required here to indicate that these are qualitative or relative terms.

b. "The data recorded at high humidity should be most relevant to atmospheric conditions, and the results indicate that parameterizations currently used in modelling studies to describe freezing rates are appropriate for kaolinite aerosol particles." The results of this study do not appear to supporting no such conclusions. If the words "We assume that..." or "We speculate that..." preface both statements, then they become more acceptable. Knowledge of the mechanisms, role of particle size, and so on is lacking still. The application of a simple parameterization applied uniformly to all particles of a certain type is not necessarily supported by these new studies.

2. Experimental: Organization of this material could be improved. I gather that the investigation of humidity effects was not necessarily the focus of the studies originally, but was instead an outcome of attempting to operate at higher temperatures. This is stated several sentences into the Results section. It is fine is this if how things happened, but those statements belong here in the description of experiments, not later. On a related matter, it bears bringing out more clearly that the RH variation was not controlled (only qualitatively low, medium, high, as stated in the Results instead of here), but that the authors believe it was below water saturation, at least near the

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drops. This is vitally important to the entire presentation because the inability to define RH limits any insights into understanding the mechanism involved.

a. Page 2420, lines 9-10 - The mixing of "dry" air with "humid" air "increases the RH substantially." What does "substantially" mean here? Do the authors have any estimate of the chamber RH before and after mixing? Furthermore, is there any reason to believe that the mixing of the different streams leads to a monotonic increase in RH versus possible transients? Is it assured that such does not in itself potentially lead to ice embryo formation in the seed particles under some set of conditions?

b. Is the number concentration of particles from the generator that is said (page 2420, lines 20-21) to remain stable the "total" number concentration, as measured by the CPC (mentioned on page 2421)? What were the total number concentrations? Only the size distribution above 0.3 microns is shown in Fig. 2 and then there is an extended discussion on pages 2421-2422 about the total concentrations (again not shown) being unrepresentative of the concentrations in the vicinity of the drop. One of the reasons given is that "the probability for particle-droplet collisions depends on the flow conditions around the droplets during the experiments, which introduces a dependence on particle size." Then, in the last sentence of the experimental section, it is stated that "the observed particle number concentrations were assumed to be proportional to the probability of collision with a droplet per time unit." This is all quite confusing. Please revise this discussion to state clearly what number concentrations were typically used and why some explicit determination of collision rates using a number size distribution estimate, drop size, and flow rate cannot apparently be made or estimated. Only then can one understand and evaluate the alternate method used for estimating collision rates.

3. Results: a. Page 2422 - "The last column in the table gives the ratio between the number of frozen droplets and the total number of droplets at each temperature." Does this mean that in some cases the drop did not freeze before evaporating? Please clarify.

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b. Page 2423 - Is the single C value used from the CPC or from the Grimm data?

c. Page 2423 - "...at 240 K, C is calculated by setting E to 1 for the droplets at this temperature." Why does C have to be calculated at 240K if it is measured? Did the authors mean to say F? If the first collision freezes the drop at this temperature, would not the freezing be nearly instantaneous with turning on the aerosol flow? I do not understand.

d. Page 2423 - "Effects of droplet size were not included in this analysis." Effects of aerosol size were also inherently not included, correct?

e. Page 2423 - To repeat a general comment, I wonder if the authors can rule out impacts of the humid air on particles prior to their collision with drops? In other words, one might imagine a process being induced akin to the one Cooper (1974) describes, but that the ice embryos are produced in the mixing region and then impact the droplets. This of course would not be relevant to the real atmosphere.

4. Discussion: I wonder if the results do not potentially support Cooper's hypothesis? He noted that the time involved in growing an ice embryo near a drop was less than a millisecond, but he did not investigate the situation for very low humidity and strongly evaporating particles.

5. Conclusions: a. Page 2426, line 17 - "Assuming that collisions at the lowest temperature employed had a probability of unity, one or a few collisions were usually sufficient to produce contact freezing." I cannot understand this sentence. To me it says that assuming a collision is highly effective for freezing, then a few collisions produce freezing. It is the same. Whatever the meaning, does it apply to all temperatures or just the lowest temperature? Please explain the intent here or remove the sentence.

b. Page 2426, line 19 - "In clouds, the RH is close to 100 percent. Hence, the data recorded with water vapor added is the most realistic case for atmospheric applications." There is no knowledge of the exact RH (or its proximity to water saturation) in

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this study or its relevance to atmospheric conditions favoring contact freezing. The higher RH data may be most relevant. It is speculation.

c. Page 2426, line 21 - "The present results support the parameterization used in the modelling study by Lohmann and Diehl (2005)." How so? What do these experiments have to do at all with the simplified assumptions used in a modelling study. These types of proclamations tend to be abused.

Editorial Comments

Abstract, sentence 1: To emphasize which particles were primarily flowing in these experiments, I suggest that it should read, "Contact freezing of kaolinite dust particles colliding with single supercooled water droplets has been investigated."

Page 2424, lines 17-19 - "The results from the present study are consistent with the previous results by Pitter and Pruppacher (1973) when water vapour is added." The meaning of this sentence is unclear. I assume it intends to state that the results are consistent if one compares PP73 with the results obtained when water vapour was added in the present study.

Page 2426, line 3 - Surface layer more easily "adapts".

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 2417, 2009.