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Atmospheric
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Discussions

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

E. A. Svensson et al.

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We would like to thank the anonymous referees and Dr. Kulkarni for their helpful comments on our manuscript. We believe that this will help improve the manuscript substantially, and we have followed the recommendations and clarified where it was necessary.

Responses to specific comments by Anonymous Referee 1:

1. a) We have changed and clarified this.

b) This sentence has been changed in accordance to the comments.

2. We have rearranged the manuscript according to the comments.

a) This has been clarified. According to Salam et al.(2006) the onset of deposition ice nucleation is far away from the conditions in the present experiment.

b) We have revised this section and hopefully clarified the message.

3 a) Yes, or at least we did not observe them freezing. This is now explained in the text.

b) It180;s from the CPC.

c) Yes, we meant F. Corrected.

d) Yes, that is correct; we did not include particle size either.

e) We believe, based on the reasoning which we have improved the presentation of, that we can exclude this possibility.

4) We have added a short note on the consistency with Cooper (1974).

5) a) This sentence has been clarified.

b) ’the most realistic’ has been changed to ’likely to be a more realistic’.

c) The commented sentence has been removed.

We have followed the editorial recommendations.

Responses to specific comments by Anonymous Referee 2:

Why is the freezing efficiency greater than 1 for experiments performed at 242 K? (Greater than 2, in fact.) The only way I could explain this would be for droplets to freeze without having collided with kaolinite, but the authors have ruled this out as a possibility (lines 1-4 on page 2421). (Brief digression: You state that no droplets froze when droplets were not injected into the chamber. Did you also test to see that no droplets froze when no particles were injected but when the air was humidified?) The freezing efficiency for droplets at 244 K is also greater than 1. Please explain.

The uncertainties are very large when only a few droplets were investigated. In no case the freezing efficiency was above 1 within one standard deviation. We did regularly check whether droplets froze at every temperature when particle flow was turned off,

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and this not not happen. At the high temperature cases, the flow of humidified air was turned on the whole time.

I find Equation 3 to be quite confusing...

The issue is how to estimate time the unfrozen droplets stayed 'active' in the trap, since they evaporate rapidly. We did this by using the average time of the frozen droplets. This may cause a bias towards higher E values, but on the other hand the droplets may have frozen without us being able to detect it.

Also, please take a look at the caption for Equation 3.

We have rearranged the equation to make it clearer.

In line 7 on page 2423, you state that C is calculated by setting E to 1. I think this should be F.

That is perfectly correct and we now have changed it.

I think it would be worthwhile to tabulate a few more of the quantities you use. For instance, a table of F at 240 would show the kind of variation you have in the experiment. Similarly, a statement of tf for each temperature (along with a standard deviation or some other measure of the variation) would be interesting to see. (I would be particularly interested to see if tf is exponentially distributed.)

We have added the means and standard deviations to the table. A comment on the statistical distributions of the freezing times has been added to the manuscript.

pg. 2418, line 19: 8221;Other influential factors are droplet volume and surface area...8221; For heterogeneous nucleation, droplet volume is usually considered to be irrelevant because the nucleation rate will be dependent on the surface area of the water-catalyst interface, but the rest of the volume of the droplet doesn8217;t matter. The surface area of the heterogeneous catalyst is certainly relevant, and for contact nucleation, the surface area of the supercooled droplet is as well. (I suppose you could

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argue that, therefore, the volume is relevant because greater volume implies greater surface area.)

This sentence has been rewritten.

The premise of the paper is that high relative humidity makes kaolinite a better ice nucleus in the contact mode. From Figure 3, it seems that assertion is based primarily on the data point at 263 K. The data clearly show that kaolinite becomes less effective with increasing T for the low relative humidity series. I understand that increasing relative humidity at higher temperature was a way to make the droplets evaporate more slowly, but why wasn8217;t high relative humidity tested for the lower temperatures. The case would certainly be more convincing if E was higher for, say 250 K, for high RH than for the low RH.

We have revised the abstract and conclusions to make the paper less definitive.

pg. 2422, lines 3-4: I would reverse the sense of this sentence. The probability of collision with a droplet per unit time is assumed to be proportional to the observed particle number concentration.

We have followed this recommendation.

Responses to comments by Dr. Kulkarni:

After reading the paper I could not find any satisfactory thermodynamical reason for how contact freezing occurs. To explain this freezing mechanism authors have highlighted new references like Suzuki et al. (2007a and b), ruling out earlier hypotheses, and indicated solid-liquid-air interface theory could favor the contact freezing nucleation. I wonder if the authors would like to discuss more about solid-liquid-air interface theory. Would be nice work to develop ice cloud parameterizations using this theory and experimental results presented in the paper. I suggest authors to have a look at Djikaev and Ruckenstein, 2008 (J. Phys. Chem. A 2008, 112(46), 11677) paper for more theoretical discussion on solid-liquid-interface theory applied to contact freezing *nucleation mechanism.*

The main subject of this article is to present the experimental results and to stimulate more research into the subject. We will not go into the detailed thermodynamic calculations that exist, but we have added a reference to the Djikaev and Ruckenstein paper.

In the present paper it is observed that probability of freezing decreases with increase in temperature, whereas it increases with increase in humidity. I was wondering if authors have any detailed explanation for this observation.

No, we do not have any satisfactionary explanation to the RH dependency. It however appears consistent with the hypothesis in Cooper (1974) as we note in the revised manuscript.

This could be important as to know whether temperature or humidity or both variables are important for the contact freezing.

It is clear from this and several previous studies that the temperature has a major role in contact freezing. Our experiments indicates that the RH also plays a role.

Also I could not find at what relative humidity values the experiments are carried out, and how do you measure it.

We are not able to determine the RH in the vicinity of the droplets since the mixture of the air ’masses’ takes place within the chamber.

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