Atmos. Chem. Phys. Discuss., 14, C390–C393, 2014
www.atmos-chem-phys-discuss.net/14/C390/2014/

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Representing time-dependent freezing behaviour in immersion mode ice nucleation" *by* R. J. Herbert et al.

Anonymous Referee #1

Received and published: 11 March 2014

Herbert et al. present an interesting framework which can be used to reconcile laboratory immersion freezing data determined through various measurement devices including different measurement techniques and nucleation time scales. The presented work contributes to finding a comprehensive tool for consistently describing (laboratory) heterogeneous freezing data. The FROST framework is extensively and comprehensibly described. However, I have some comments concerning the laboratory database which should be addressed before the manuscript can be published.

Specific comments

Page 1401: You write that contact ice nucleation occur "through the collision between an IN and the air–liquid interface of a supercooled droplet". You may include that there

are two different possibilities: outside-in and inside-out collision. To my understanding the latter mechanism is the important one for freezing of evaporating supercooled droplets (as you mention in line 22-23)

Page 1402: Ervens and Feingold (2013) show in their study (single component system, based on CNT i.e., stochastic approach) that a change in temperature has a larger effect on the frozen fraction compared to a change in time (one to several orders of magnitude depending on the chosen contact angle). But this finding is not in contrast to the findings of Kulkarni and Dobbie (2010), Murray et al. (2010) and Welti et al. (2012). An isothermal experiment only shows the stochastic nature of freezing, i.e., the time dependence. But the interesting question is how does a change in temperature relate to a change in time. For example, Welti et al. (2012) showed for their kaolinite sample that a change of $\Delta T = 1K$ in temperature relates to a time ratio change (t2/t1) of a factor 10, i.e., temperature is more important than time. Please rephrase the paragraph accordingly.

Page 1403: The probability for critical cluster formation depends on droplet volume and time, but most important on temperature! The lower T, the higher is the freezing probability.

Page 1406, Eq. 1: Do you consider that after each time step the liquid droplet number has to be adjusted so that the total number (frozen plus liquid droplets) keeps constant?

Maybe I missed that piece of information: How many droplets do you examine in your cold stage cell studies for kaolinite and feldspar. What are the measurement uncertainties for the determined frozen fractions due to temperature uncertainty (0.4 K for isothermal experiments), considered droplet number (Poisson statistics), etc.?

Chapter 4.1 and Fig. 4 and 5: Regarding the microlitre data in Fig. 4a, I would think it shows a less steep slope compared to the picolitre data. Is there maybe an obvious difference between the two samples? And why does the frozen fraction in Fig. 5a for 0.5 K/min and 1K/min cooling rate does not reach 1?

As you investigated droplets featuring more than one particle you should avoid saying that kaolinite is a uniform species since you can not prove it. Due to a high amount of particles per droplet it might be possible that there is a high probability for droplets featuring particles with similar ice nucleation activity. Welti et al. (2012) also used kaolinite, indeed they used a sample from Fluka which differs from your sample. However, they show that there is a particle-to-particle variability for their kaolinite sample. In my opinion you should only state that for your investigations the kaolinite sample can be treated as a single component system but there could be multiple IN populations leading to different results if droplets with single particles are regarded (you have this kind of interpretation on page 1417/1418).

Fig. 8: Please include the measurement uncertainties also in the raw data. For me it looks like that the raw data already agrees within the uncertainties if the error bars from Fig. 8b) and 8d) are used. Can you also plot a figure for the Welti data as you did in Fig. 4b and Fig. 7, i.e., calculating μ and σ for this kaolinite sample?

Looking separately on Fig. 9a, I would think that both data sets are clearly distinguishable through different slopes or cannot completely be represented by a single straight line curve (i.e., your straight line fit cannot be extrapolated to higher T). So, I am not completely convinced that the volcanic ash sample behaves like a single-component system (Can you please plot the nucleation rates here? If it behaves like single-component, the rates for both experiments should fall onto one line without fitting.). As already stated in the text, more isothermal or cooling rate experiments would be needed to prove this assumption.

Minor comments

Page 1401, line 9: Replace warmer temperature through higher temperature.

Page 1409, line 1: It should read n_frozen/n_liquid. However, to my understanding the frozen fraction is defined as the frozen droplet number divided through the total number of particles (i.e, sum of frozen and liquid droplets). I am just wondering about

the definition of Eq. (13), is it valid with your fraction frozen definition?

Page 1410, line 16: I am not sure about the sign for the cooling rate: Is it positive or negative? Intuitively I would think r_cool > 0 K/min?

Page 1419, line 1: Please add that T' = T_exp – β(r)

References

Ervens, B. and Feingold, G.: Sensitivities of immersion freezing: reconciling classical nucleation theory and deterministic expressions, Geophys. Res. Lett., 40, 3320–3324, doi:10.1002/grl.50580, 2013.

Kulkarni, G. and Dobbie, S.: Ice nucleation properties of mineral dust particles: determination of onset RHi , IN active fraction, nucleation time-lag, and the effect of active sites on contact angles, Atmos. Chem. Phys., 10, 95–105, doi:10.5194/acp-10-95-2010, 2010.

Murray, B. J., Broadley, S. L., Wilson, T. W., Atkinson, J. D., and Wills, R. H.: Heterogeneous freezing of water droplets containing kaolinite particles, Atmos. Chem. Phys., 11, 4191–4207, doi:10.5194/acp-11-4191-2011, 2011.

Welti, A., Lüönd, F., Kanji, Z. A., Stetzer, O., and Lohmann, U.: Time dependence of immersion freezing: an experimental study on size selected kaolinite particles, Atmos. Chem. Phys., 12, 9893–9907, doi:10.5194/acp-12-9893-2012, 2012.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 1399, 2014.