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Interactive Comment

Interactive comment on "Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments" *by* C. Hoose and O. Möhler

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We thank the reviewer for his/her constructive and detailled review of the manuscript. The reviewer's comments are included below in italics.

I rate this manuscript as excellent on all accounts, and in need of the following minor revisions prior to acceptance in ACP. This is a serious undertaking and a carefully constructed summary of many ice nucleation measurements in the atmospheric The manuscript represents a major and useful contribution to be used by many other researchers. The only serious omission I see is a discussion of contact freezing. I am

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surprised by this omission, since the lead author has stated in previous work that contact freezing is in fact, the least understood, mechanism in heterogeneous freezing at this time.

We have added a new section (4.1.3) on contact nucleation summarizing historic and recent experiments. Most of these were already listed in the tables included in the discussion version. Because the number of collisions is very difficult to constrain, this information is unfortunately not available for most contact nucleation experiments, making quantitative comparisons very difficult. Therefore we were unable to summarize the results of contact freezing experiments in an overview figure such as those for immersion freezing and deposition nucleation.

A second omission is that surface area vs. volume based nucleation rates occupied a lot of literature discussion and should also be included, at least, in brief.

As we have limited our article strictly to heterogeneous ice nucleation, this discussion (which referred to homogeneous ice nucleation) was not included on purpose. We think that including a discussion of homogeneous ice nucleation would go beyond the scope of this already lengthy manuscript.

Third, aerosol aging processing which are not represented by coating should be at least mentioned. While these are almost non-existent in the literate, this type of experiment needs to be undertaken in a serous way in future experiments It would be nice to that mentioned briefly.

We have added mention of aging by exposure to reactive gases such as ozone and to light (photochemical aging) in section 3.4. Table 6 now includes new references (Attard et al., 2012; Chou et al., 2012) which employed these artificial aging methods. As mentioned by the reviewer, there is only a small number of such studies. We think that producing an overview plot would be too premature at this point.

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Minor comments follow: Pg. 12. The manuscript states that - According to Murray, commercially available minerals may have underdone acid washing or other chemical processing. It should also be mentioned that commercial processing can include physical processing, such as milling which may change particle size , morphology, and surface roughness.

We fully agree with the reviewer. However, it was surprisingly difficult to find a peer-reviewed reference describing the processing of the clay mineral samples studied in ice nucleation experiments. The reference which we finally included is an online publication describing of the treatment of the Clay Mineral Society Source Clays. The newly added sentence reads: "Powder samples have usually undergone mechanical processing such as mechanical desaggregation and milling (Moll, 2002)."

Pg. 14. In the discussion of impacts of coatings added to aerosols in laboratory, it would be useful to mention that methods of coating have been poorly constrained in most studies and that the resulting coating vary in thickness and also may vary in the percentage of the aerosol population which is coated vs. uncoated. This makes comparison of results from various experiments difficult, and also makes appropriate use and interpretation of these for atmospheric modeling difficult.

This is a good point. We have inserted the following sentences: "Intercomparison between different experiments is difficult because various coating methods have been applied. The resulting coating thicknesses are often poorly constrained and not necessarily evenly distributed over the particle size distribution." The difficulties in applying the results of these experiments in models are briefly discussed in the last paragraph of this section.

Pg. 14. Line 9 The manuscript defines a norm, referencing only 2 publications and *then states that 3 manuscripts are exceptions to this norm. This needs revision.*

More references for the “norm” (i.e. for an increase in saturation ratio by sulfuric acid and ammonium sulfate treatments) are added to the text. These are the majority of the experiments shown in Fig. 9. Only a subset of the data in the 3 other manuscripts are “exceptions”.

Pg. 15. Line 11. Word choice: ..."(in reality always" is out of place.

These words have been removed in the revised manuscript.

Pg. 17 line 17 "High nucleation onset temperatures are consistent with most results at low T" This does not make sense as written.

This sentence has been reformulated as follows: “the high nucleation onset temperatures for relatively large particles ... are actually consistent with most of the results at lower temperatures, which were obtained with smaller particles.”

Pg. 21-22. Section on time dependence. This fairly lengthy section really only discusses the work of Murray 2011. A literature survey would reveal that there are quite a large number of other published works addressing the issue of time dependence. Omitted them and focusing exhaustively on Murray is inappropriate.

This paragraph has been extended by the discussion of both older and more recent studies: “In extensive early studies (Vali and Stansbury, 1966; Vali, 1994, 2008), it was shown that immersion freezing exhibits time-dependence, but that the observed time dependence is weaker than expected from classical nucleation theory (see Appendix A1). Recently, further in-depth analysis of the influence of time on the activated fraction were conducted by Murray et al. (2011), Broadley et al. (2012) and Welti et al. (2012). In their experiments, the cooling rate or residence times could be varied by more than a factor of 10. Their results are summarized in Fig. 16, along with two experiments by Niedermeier et al. (2011) and DeMott (1990). While Murray et al. (2011)'s and Welti et al. (2012)'s results indicate significantly more ice nucleation in experiments with slower cooling rates, the other studies do not confirm this behaviour and instead show sometimes more, sometimes less ice nucleation when more time is available for freezing. It should also be noted that the spread in the data entering these calculations is large. For none of the experiments, the ratio of ice fraction or INAS densities in the slow versus fast experiments converges towards the ratio of the cooling rates or residence times, as would be expected if nucleation was described well by a temperature-dependent nucleation rate with one value for all particles. Nevertheless, Murray et al. (2011) found that such a single component stochastic model fits their data best, in particular if constant-temperature experiments are considered. Welti et al. (2012) could best fit their results with a distribution of contact angles."

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