

## ***Interactive comment on “Immersion freezing of birch pollen washing water” by S. Augustin et al.***

### **Anonymous Referee #2**

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Augustin et al. report new data on the immersion freezing of birch pollen extracts. Experiments were performed on two pollen samples (Northern and Southern birch) for mobility diameters ranging from 150 to 800 nm using the LACIS cloud simulator. The authors find that the majority of particles that were atomized from the extracts nucleated ice between -20 and -24°C. From these data the authors derive heterogeneous nucleation rates for IN active molecules present in the sample.

The topic of the manuscript is of great current interest to the community. The IN measurements performed with the LACIS system are sound and the data are new. Similar pollen samples have been studied before by Pummer et al. This study is different from Pummer et al. because it uses water droplets suspended in air instead of water droplets located on a solid substrate. It also enables the authors to study the dependence of freezing on particle size (or surface area or volume) which are more difficult to control in the cryo-microscopy setup used previously. Given the difficulties

in IN measurements and the great uncertainties surrounding the topic I consider these differences sufficient that the manuscript may be publishable. Notwithstanding this recommendation however, I find that the manuscript has serious weaknesses that need to be addressed before it should advance to ACP.

Major concerns:

The word macromolecule is used a total of 61 times in the manuscript and assertions are made about their abundance and nature in different species. This paper however can hardly make assertions about macromolecules. The notion that macromolecules cause freezing is by reference to past work and in most places the authors simply speculate. All the authors studied were atomized birch pollen extract that was aged overnight inside a refrigerator. The IN activity may emanate from bacteria that are associated with the pollen, bacteria that grew in the extract overnight, contaminants that came with the purchased samples, or even dust that has settled on the pollen. It is interesting to mention what the authors believe to be true based on earlier work but in absence of clear evidence on the nature of the IN obtained via chemical analysis of the particle residues the discussion of the results and conclusions should limit itself to the simple fact on what was done (atomizing washed pollen extract).

The authors claim that their model can explain the IN behavior of the samples (e.g. pg. 32920). However, all the authors are doing is fitting their data with a model. When the Northern Pine did not confirm to the model, they simply changed it to include another population of particles. A more accurate description of the methodology would be “we are able to fit the data assuming . . .”.

It was pointed out by referee #1 in the initial unpublished review that it is unclear that Figure 8 shows indeed two different slopes and hence two different IN populations. Perhaps there are two populations, but I find that the authors have too much faith in their data. For example DeMott et al. (2011, BAMS) report data from an ice nucleation workshop where all participants sampled the same aerosol using different methodologies.

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Examining their Figure on Sahara Dust should make it clear that small changes in  $f_{ice}$  or freezing temperature are unlikely to have meaningful physical interpretations until the community can substantially improve the accuracy of IN measurements. Perhaps LACIS is indeed superior to all of the other techniques, but the onus is on the authors to present a convincing argument why this would be the case. The observed second population may simply be due to a different size distribution and multiply charged particles (not accounted for here) in the sample, or different densities of IN active substances in the particle. It could be due to more unaccounted frost falling from the walls or the presence of more IN inhibiting substances in the matrix of the washing water. To my eyes the difference in the freezing temperatures are within a couple of degrees which seems unresolvable given the combined uncertainties in the experiment.

In summary, the authors should remove the discussion and revisit the explanatory constructs made with nucleation rates, number and types of INA macromolecules washed of, and partial dissolution of samples. This study demonstrates that washing birch pollen in water and atomizing the extract produces IN active particles that nucleate ice at temperatures -20 to -24C. A set of CHES parameters can be found to fit the data for each species. The IN activity is only a weak function of particle diameter and pollen species. Drawing further conclusions seems to be speculative and thus not warranted.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 32911, 2012.

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