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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice-nucleating efficiency of aerosol particles and possible sources at three coastal marine sites" *by* Meng Si et al.

Anonymous Referee #1

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Review of "Ice-nucleating efficiency of aerosol particles and possible sources at three coastal marine sites" by Si et al., submitted to ACPD:

The study described in this manuscript is an interesting addition to similar work by the group in which M. Si is working. It interprets measurements of atmospheric aerosol wrt. concentrations of ice nucleation particles (INP) and their possible sources, as well as effects of particle size on INP activity. The work is interesting and timely. However, I have a few major comments (besides for a number of smaller ones) that need to be addressed before the work can be published. The major comments mainly concern the amount of data (which is rather low) and a possible malfunctioning of one of the size spectrometers used and related consequences on the results.

But altogether, the study merits publication once again my comments will have been

considered and changes will have been implemented adequately.

Major comments:

The first comment concerns the amount of data used for the study. For two locations, only one measurement was made, while for the third location, the data used has already been used in a different study on INP before. This is properly stated in the text. And the results obtained herein evaluate these data in a new way, yielding more results. But the abstract had raised high expectations, and I was quite disappointed when I realized that the abundance of data included in this study is rather low. It should be made clear already in the abstract and again in the conclusions that the data base is not very strong. This might also influence the results, as one measurement does not deliver good statistics, and this has to be dealt with offensively and should be discussed.

I am also concerned about the misfit in the particle number size distributions between those number concentrations measured by the SMPS and by the APS. There is a large gap at $\sim$ 500nm which implies that one of the two instruments might have not worked well. As the majority of the particles is in the SMPS size range, as usual, and as these seem to fit well with literature (at least that is what is said on page 8, line 14), it might have been the APS, measuring roughly one order of magnitude too low concentrations (that's roughly the size of the gap, larger for Lancaster Sound, a little less for Amphitrite Point and only $\sim$*2 in the Labrador Sea). This would translate to the same magnitude of error (i.e., overestimation in this case) in INP concentrations and surface site densities, affecting many statements/results reported in the text. The observed strong increase in n_s for particles < 500nm to particles > 500nm likely is (at least in part) related to this gap between number concentrations as measured by SMPS and APS. Is there a way to find out what the problem might have been? Was there a total particle number counter deployed that could shed light onto this? In any case, this problem has to be critically discussed and related changes in the interpretation of the data has to be included in the revised version of the manuscript.

page 9, line 9: It is interesting that you find n_s depends on size. But why could that be? - The larger particles would have to consist of a material that is more ice active (per surface area) than the smaller ones. How should this come about? (A mechanism would have to be that e.g., clay minerals make smaller particles, and then, the larger the mineral dust particles get, the higher becomes their feldspar content, and isn't this unlikely?) - This observed increase could be a measurement bias (as mentioned above and again in my comment concerning Fig. 4). - This needs to be discussed!

page 6, line 30: Was the model really run for 2001? If yes, why not for the respective month of 2013 and 2014, i.e., when the sampling was done? Can it really be assumed that the average monthly INP concentration is the same every year? How much variation could there be expected, and where within this variation are your data? Could this influence your results?

Figure 4: Again, as this is where I first noticed it: There is a VERY pronounced gap between number concentrations measured with the SMPS and the APS. Do you have any explanation? This could potentially influence the derived n_s values and the fraction of INP per particle as well as your comparison with the model, particularly if it was the APS that did not operate well.

Minor and technical comments:

page 1, line 17: Add "particle“ before "size“.

chapter 2.1.2: Also mention the temperature and RH at which droplet formation was done (the text has to be complete, i.e., readers should not have to look this up in another paper)

equation (1): There seems to be a typo in the formular: N_0 should not be there as a factor. The value resulting from this equation has a wrong dimension (assuming the corrections factors are dimensionless, which is how they are given in the Supplement). Please check this carefully – see also e.g., Hader et al., 2014.

page 5, line 24: Reaching an RH below 2% by a silica gel diffusion dryer is quite low (even when the silica gel is changed every 24 hours), unless the dew point of the outside air is quite low, anyway. – Did you estimate this value or check it?

page 6, line 6: Please give wind-speed in SI units – knots is a unit many (including myself) may not be familiar with.

page 6, line 10: As the MOUDI was inside at least at Amphitrite Point, drying will automatically have occurred, as it will have been warmer in the container than it was outside. This typically leads to a quick drying. The way you formulated this here is correct, however, it may be good to hint at the fact that the RH will also not have been the outside one.

page 7, line 6 and page 10, line 12: The “-“ is missing for Vergara-Temprado.

page 7, line 17-18: It might be better to formulate it in a different way. Basically you are saying that you don't know where the INP came from (marine or terrestrial and maybe even from further away), so please say something like: “Therefore it is not possible to determine if the INP are of marine or terrestrial origin and they may even have been long-range transported from sources more than three days away.”

page 7, line 24: Check with chapter 2.3.1 – you give different dates for the sampling period at Amphitrite Point.

page 7, line 26: Clearly state how many samples you used for the present study.

page 8, line 14: Marine sites may differ depending on the sea spray production typical for the area. Were the two studies you cite here done at locations that are similar to yours in this regard? Please mention in the text if they are.

page 8, line 25 ff: This effect was already reported by Mertes et al. (2007), which therefore should be cited here or in the following paragraph.

page 9, line 26-27: Niemand et al. (2012) report n_s for dust samples in which all

particles were dust. But your way to determine n_s relates the ice activity to the total particle number concentrations that were measured, hence, this is not the exact same parameter. This needs to be clearly stated here. BTW: In DeMott et al. (2016), INP concentrations for an assumed total particle number concentration of 150 cm-3 were reported (at least that's the value the laboratory data are normalized to - this is said in the caption of Fig. 1 to which you are referring), which is a factor of 2 to 4 below your values – this should also at least be mentioned, although, admittedly, this will not change your results.

page 9, line 29-30: It should be motivated a bit stronger why you make these statements here. My comment above this one might be one reason, but I am sure you had more in mind when writing these lines.

figure 1: The insets of the pictures of the ship and measurement container might not be visible any more in a printed version, so if you care for them, you might want to make them larger (there is enough "empty" space on the map).

chapter S1: Add values for the growth factors used (and / or for values for kappa).

figure S3: In the caption you say that "Each data point was calculated by adding together the numbers from Fig. 4." Did you really simply add the data points? Looking at the number, this does not seem to have been the case, and it would have been totally wrong.

Literature:

Hader, J. D., T. P. Wright, and M. D. Petters (2014), Contribution of pollen to atmospheric ice nuclei concentrations, Atmos. Chem. Phys., 14(11), 5433-5449, doi:10.5194/acp-14-5433-2014.

Mertes, S., B. Verheggen, S. Walter, P. Connolly, M. Ebert, J. Schneider, K. N. Bower, J. Cozic, S. Weinbruch, U. Baltensperger, and E. Weingartner (2007), Counterflow virtual impactor based collection of small ice particles in mixed-phase clouds for the physico-

chemical characterization of tropospheric ice nuclei: sampler description and first case study, Aerosol Sci. Technol., 41, 848-864.

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