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Comment

Interactive comment on “Ground based measurements of immersion-freezing in the eastern Mediterranean” by K. Ardon-Dryer and Z. Levin

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General Comment

This manuscript provides interesting and very useful new results on ice nucleating particle concentrations for a locale that has not been well documented previously, and one that is particularly heavily influenced by desert dust. It is an appropriate, and will ultimately make a nice contribution to ACP. I have a few critical comments and suggestions on the paper, which I list here. I have not considered comments by other reviewers in this assessment. One primary concern is to note the expectation upfront

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that the results distinguished as dusty versus non-dusty are really degrees of dustiness for the location, since the PM values alone indicate that the site is never truly clean in the sense of sites away from deserts or other strong aerosol or pollution sources. Secondly, I wonder if the categorization by PM could not be quantified in a manner besides the median freezing temperature. What I mean is to show the impact of PM across the temperature spectrum of ice nucleating particle concentrations. Absent some estimate of surface area or particle number concentrations in different size categories, there is no ability to normalize the results to see if they fit the sense now understood from laboratory studies of mineral dusts. This may require some reorganization and use of fewer figures to focus on ones that present data already processed for volumetric concentrations versus simple frozen fractions of drops. I also list below a number of specific comments on the section discussing biomass burning that I will not summarize here. Regarding comparison of the slope of the ice nucleating particle number concentration temperature spectrum to other published data is interesting, but one of the points of the DeMott et al. (2010) paper was that number concentration alone has no particular meaning or expected slope when assessed at a number of different places in the free troposphere where sources and losses are integrated into the observations rather than being characterized by a single dominant regional source. It may well be that the slope inferred in the present studies is in disagreement with the observations made by the method used in that paper, but showing the data together in this manner is not a very good diagnostic of such an issue. This is exacerbated by the fact that the immersion freezing spectra do not extend to the lower temperature range to prove if the simple exponential function fits across the full mixed phase cloud regime, a point that should also be mentioned. Finally, why is no attempt made to integrate previous measurements in the region into such a plot? I suppose it is deemed that the previous measurements were not necessarily for immersion freezing, but I think that no attempt has been made to assess if the condensation freezing methods applied in those papers might actually be quite consistent with the immersion freezing data assembled in this paper.

Specific Comments

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1. Page 474, line 1: Here it is suggested that the data collected in this paper is distinguished by mode, but as I mention in my last general comment, I do not feel that it has yet been considered if the results of a specific immersion freezing measurement are or should be distinct from processing particles on a surface under humidity that is at or forced to exceed water saturation. At least it should be evaluated if the prior measurements appear to be part of a similar data set. I think they do, with the Levi and Rosenfeld (1996) data fitting fully within the range found at the warm temperature limit of the present measurements, and matching the quantitative impact of dust loading, and the Gagin (1975) measurements appearing as one might perhaps expect for air mixed to sub-cloud levels.

2. Page 475, line 3: Could the definition of dust “episodes” in Ganor (1994) be mentioned here? As a reader, I would already have the idea that dust is omnipresent in the region, so the concept of an episode needs to be made clear.

3. Page 475, line 20: I think the absolute lower size limit of the CPC type mentioned is 0.011 microns, or 11 nm, not 0.11 microns. See also line 20 on page 480 if this is the case.

4. Page 476, line 28: For the sake of consolidating experimental protocol, could it be mentioned here how efforts were made to define and limit any artifacts? For example, were “pure” droplets distributed using the same pre-cleaned tools onto the Vaseline to obtain negative checks, and were these subtracted from the polluted drops in a manner consistent with the freezing data analysis? This topic is not really introduced until the discussion section on page 478.

5. Page 479, lines 5-6: You may have misinterpreted the data shown in Figure 5 of the DeMott et al. (2006) extended meeting abstract as freezing spectra. They are not really that. Plotted there are cumulative frequency distributions of 1 minute observations of CFDC instrument processing temperature, ice supersaturation, and IN particle concentration in four research projects. What the plots show for the two Arctic studies are

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the range of temperatures and water vapor supersaturations covered in each project, and the fraction of time a given IN particle concentration was exceeded for the entire data set. The 50% processing temperature and supersaturation for each study is not necessarily the value associated with the 50% IN particle number concentration. There is in fact no distinct association of the IN data with temperature in the figure, except that the concentration distribution goes with the temperature range examined.

6. Page 479: The discussion of the biomass burning case here raised more questions for me than nearly any other section of this paper. I feel that the present conclusions exceed the bounds of what can readily be discerned from the data. First, it is not clear what the basis is for comparing the case to others. I presume at this point in the paper it is simply the freezing fraction spectrum rather than volumetric concentrations or volumetric particle fractions. I think that the best basis for making comparisons are the fully processed volumetric concentration data, so I suggest that those plots are introduced rather than focusing any discussion around median freezing temperatures alone. Now I will list my questions and comments concerning the presentation and conclusions regarding ice nucleating particles from biomass burning.

a) There were two filters on May 1, but only one is discussed. Is there a relation between them and a reason the one labeled “15” was so different than the one labeled “23”. Was there a regional change in the background air mass at the time? For example, how did PM levels change in different size fractions around this time? A related question is if there is really a way to distinguish the background on which the smoke is being placed, except by comparison to all other spectrum obtained? There were no data collected on the following day, so it is hard to place the festival data in context of before and after.

b) Is there a reason for relating the present data to the Amazon data of Prenni et al. (2009b)? That is a completely different and perhaps unique location, and median freezing temperatures are not at all discussed in that paper, only volumetric concentrations.

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c) It may be that the wood type burned and the composition of the subsequent nuclei is important, and this is useful information, but please be careful in comparing to other studies. The temperatures at which an impact of biomass burning was noted in most of the studies you reference were lower than most of the range you examine in the present study. Thus, you do not have information on the potential impact of smoke at temperatures below about -25°C . Also, please be clear that your conclusion is that particles from “this type” of biomass burning are not effective ice nucleating particles.

d) I was somewhat surprised that the ice nucleating particle fraction of this particular sample was not much lower than the other samples if there was in fact so much additional pollution from the fires. Yet there is no apparent separation of the spectral results from the other days when the results are placed on the basis of total particle numbers in Figure 7. This led me to realize that the total particle numbers are listed in Table 1 (but not mentioned here), which demonstrates that indeed they were not greatly enhanced during the burning period. Hence, the question is if a true perturbation on any particle type already present was made due to burning? What other evidence indicates that this time was heavily influenced by smoke at the site?

e) Having some experience with filters collected under smoke conditions, I wondered if the filter clearly indicated smoke particle deposition by appearance and if any difficulty was experienced in assuring that all particles were being effectively rinsed from the filter?

7. Page 480, lines 11-12: Does one get an exponential equation as the best fit to all of the data, or do you mean instead that an exponential fit was assumed?

8. Page 480, lines 25-27: Again, the fractions are not with respect to 0.1 micron, but I think 0.011 microns. Mainly though, since some might be tempted to consider using the results in Fig. 7 for parameterization purposes, it might be useful to point out that referencing the IN particle number concentrations to total particle numbers adds no apparent power for predicting ice nucleating particle concentrations. In fact, the data

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spread is increased.

9. Page 481, Section 4.1: As someone living in an area where $50 \mu\text{g m}^{-3}$ is more representative of a day characterized by long range transport of dust or of regional smoke or pollution, I feel that it might be useful to point out from the start of this discussion that dusty versus non-dusty in this case is a subjective and qualitative assessment intended only to roughly segregate the data into dusty and less dusty for the surface boundary layer at the site. This would frame your discussion of results at the bottom of page 482. However, it seems like this section as a whole begs for some more in depth analyses to quantify and display the impact of PM values on ice nucleating particle number concentrations. For example, did the highest PM10 days contain the most effective IN? Perhaps those points could be highlighted. Also, what many readers may be interested in is variations with surface area. I realize that such a measurement was not obtained, but is there any historic data from dust episodes in the area relating total mass and surface area distributions such that an estimate of surface active site density could be made for comparison to laboratory dust studies such as Niemand et al. (2012)? This could give special insight into the utility of published laboratory assessments in describing real world dust cases.

10. Page 482, lines 3 to 8: Figure 9 is not extremely useful in my opinion. You could, if you desire, summarize drop median freezing temperature conditions for all experiments and segregations of such in a table. The reference to Pitter and Pruppacher seems like the only reason to mention the frozen fraction curves. Freezing spectra of IN particle number concentrations are the most important to report here.

11. Page 482, lines 13-16: Is the reference to Hoffer (1961) regarding the impact of solute concentration on heterogeneous freezing exceeding that expected on the basis of freezing point depression alone really applicable to the studies reported here? Particles were diluted into 10 ml of water. Given the collected mass concentration, could you not bound the expected solute concentration to know if you expect any such effect in your study? I suspect that you should assume no such influence on your

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freezing results, and that this effect is only applicable to studies of more concentrated solutions such as would exist naturally in the slightly water subsaturated regime. If the reference is instead to the fact that the particles may have been processed already through some conditions that may have led to degradation of active sites, then that is perhaps worthy of a simple mention, but there really is no evidence for such.

12. Page 482, lines 16-19: Similarly, I do not feel that reference to the impact of acidic sulfates (not ammonia) is relevant here either. Your drops are likely too dilute. Furthermore, a few of the referenced studies support that simple solutes may lead to no degradation of immersion freezing, and Sullivan et al. (2010b) supports that chemical processing even by some acidic species does not always lead to degradation of freezing nucleation activity. Hence, I suggest that this entire paragraph may contribute little to the paper.

13. Page 483: I mentioned my concerns with Figure 11 in my general comments, suggesting that it needed some qualification despite my understanding why it would be shown. There is only so much that can be interpreted from such comparisons using a simple ice nucleating particle number concentration plot. That was the basis for the extended analysis reported in the 2010 paper. Regarding the Meyers et al. formulation, it was a point of that 2010 paper that the data set used by Meyers et al. was extremely limited and entirely based on surface sites.

14. Page 485: Surface area is mentioned again as a possible parameter in interpreting results, but no such estimate is made here to evaluate any consistency with the data.

15. Figure 2: Already published and so you can simply refer to your earlier paper for the methods.

Technical Corrections

Page 472, line 20: Hanging thought here. Quantifying what?

Page 473, line 1: Capitalize M in DeMott.

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Page 479, lines 14-16: Currently associates the “filter” with “effective.” I suggest, “Therefore we expected that the filter sampled during Lag Ba Omer (an Israeli festival with lots of bonfires and thus a highly polluted day) on 1 May 2010 23, might contain larger numbers of effective ice nucleating particles.”

References:

Niemand, M., O. Moehler, B. Vogel, H. Vogel, C. Hoose, P. Connolly, H. Klein, H. Bingemer, P. DeMott, J. Skrotzki, and T. Leisner, 2012: A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles *J. Atmos. Sci.*, **69**, 3077-3092.

Sullivan, R. C., L. Miñambres, P. J. DeMott, A. J. Prenni, C. M. Carrico, E. J.T. Levin, and S. M. Kreidenweis, 2010b: Chemical processing does not always impair heterogeneous ice nucleation of mineral dust particles. *Geophys. Res. Lett.*, **37**, L24805, doi:10.1029/2010GL045540.

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