

K. Ardon-Dryer and Z. Levin present the ice nucleation efficiency, via immersion freezing, of ambient aerosol particles collected in Israel. Ambient aerosol particles were collected on filters and their ice nucleation abilities were studied in the FRIDGE-TAU chamber. The methods and the data analyses are the same as those presented by Ardon-Dryer et al. (2011). The authors found that the collected ambient particles nucleated ice via immersion freezing at temperatures as high as -12C with an average median freezing temperature of -21C. Ice nuclei concentrations were found to range from 0.32L^{-1} to 211L^{-1} . Additionally, aerosol particles collected under polluted conditions (daily average PM_{10} ranging from 254 to $867\mu\text{g}\text{m}^{-3}$) were found to be slightly more efficient than aerosol particles collected under clean conditions (daily average PM_{10} ranging from 30 to $39\mu\text{g}\text{m}^{-3}$). This is a nice data set and a valuable contribution to the ice nucleation community; however, major corrections are needed before the manuscript can be accepted.

General comments:

The introduction is very general and far from being complete. Important information regarding heterogeneous ice nucleation (especially immersion freezing) and the atmospheric relevance of this study is missing. Additionally, literature data is not properly presented. A detailed error analysis should be added to the Methods. The uncertainties in calculating FN and hence AF are necessary. Furthermore, the uncertainties in the reported temperatures are essential.

There are many imprecise statements in the results section. The authors move back and forth between frozen fractions, onset freezing temperatures, IN concentrations and activated fractions. This section can be divided into subsections with a better structure. Figures 4, 6 and 7 are not necessary. The same results are clearly reflected in Figures 9 and 10.

The manuscript will substantially benefit if it is edited by a native English speaker. Lastly, in the abstract, introduction, methods and results sections it is mentioned that the ambient particles were studied under different meteorological conditions. However, these conditions (besides the wind direction presented in the back trajectories) and their influence on the IN abilities of the sampled particles are unclear. I fully agree with the key points (i.e., the transfer efficiency, the use of median freezing temperatures, the comparison of the current results with literature data and the spread of the spectra at high temperatures) highlighted by Dr. Vali.

Specific comments

Abstract

P. 472, l. 10-11: It is unclear to which droplets are the authors referring to. I suggest changing it to: Droplets containing aerosol particles from dusty days froze at warmer temperatures than droplets containing aerosol particles from clean days.

Introduction

The paragraphs are not clearly structured. The heterogeneous ice nucleation modes are mentioned but they are not explained/defined in the text. Especially, immersion freezing, which is the ice nucleation mechanism of this study, was never defined. Where and under what conditions is immersion freezing important? The motivation of this study is very general. The importance of the IN concentration and its atmospheric relevance is not clearly stated in this section. Why is it important to measure the IN concentration at the ground-level? What is the contribution of the present study to our current knowledge? Literature data regarding IN concentrations is cited/discussed; however, a distinction

between ground-based and aircraft-based measurements is not provided. Without this information, readers who are not familiar with ice nucleation will easily get confused.

P. 472, l. 14 to 18: There are several sentences that should be cited. Add the corresponding references.

P. 472, l. 23 to 26: I think the authors can replace the cited references by the review done by Hoose and Möhler (2012).

P. 473, l. 9: Why is it dust important? Do the authors mean that its atmospheric relevance is greater than other types of aerosols?

P. 473, l. 9: Is it true that dust is the most effective IN? What about bioaerosols (e.g., bacteria). Recent reviews have shown that bioaerosols are more efficient IN than mineral dust particles via different heterogeneous ice nucleation mechanisms (e.g., Hoose and Möhler (2012), Murray et al. (2012) and Ladino et al. (2013))

P. 473, l. 13: The ice nucleating efficiency of an aerosol particle via deposition nucleation is not directly correlated with temperature. The relative humidity with respect to ice (RH_{ice}) at which ice nucleation is observed is commonly used to infer particle's efficiency. I suggest separating the literature studies and the conditions at which mineral dust was found to nucleate ice as function of the different nucleation modes (e.g., immersion freezing, condensation freezing, contact freezing and deposition nucleation).

P. 473, l. 26-28: These sentences are grammatically incorrect. Re-phrase it.

P. 473, l. 29: Condensation freezing is not defined.

P. 474, l. 3-4: Re-phrase it: "...to characterize the efficiency of the eastern Mediterranean aerosol particles to act as IN via immersion-freezing under different meteorological..."

P. 474, l. 4-5: Add the meteorological conditions that were tested.

The characteristics of the research area

P. 474, l. 6: Remove "the": Characteristics of the research area.

P. 474, l. 7: Add months. "...during 01.2009 and 12.2010".

P. 474, l. 13-16: This part is confusing. It reads as if the aerosol particles from the Sahara desert and from marine environments were anthropogenic, even though they are clearly biogenic sources of aerosol particles. Please re-phrase it.

P. 474, l. 19: "...and anthropogenic aerosols with a relatively..."

P. 474, l. 24: What do the authors mean by intense?

P. 475, l. 3: What do the authors mean by episodes? Can the authors be more explicit? (e.g., "dust storms or days with dust concentration larger than...")

P. 475, l. 4: Is a dust-depositing storm the aforementioned episode of dust? If yes, be consistent with nomenclature.

P. 475, l. 4-7: This long sentence can be divided in two sentences.

P. 475, l. 7-9: Is it possible to re-phrase this sentence. It is a bit confusing.

Method of analysis

I suggest adding a subsection about the uncertainties. The uncertainties should be added to the corresponding figures.

P. 475, l. 14: Why is the sample flow 20 LPM and not 8 LPM as in Ardon-Dryer et al. (2011)?

P. 475, l. 16: Meteorological conditions such as what?

P. 475, l. 18: Is it possible to add to Table 1 such conditions (i.e., polluted, clean, and the most relevant meteorological conditions for each sample)?

P. 475, l. 19: The sampling time is not clearly mentioned. Was it constant for each sample? If not, it can be added to Table 1.

P. 475, l. 20: Is it the size range in radius or diameter? Please clarify it.

P. 475 and 480, l. 20: The lower limit detection of the 3010 CPC is 10 nm which means 0.01 μm and not 0.1 μm .

P. 475, l. 21: "operating" is not appropriate.

P. 475, l. 22: "...concentration (Nt) of the aerosol particles..."

P. 475, l. 22: Was it Nt determined or measured?

P. 476, l. 7: I suggest to re-phrase "for measuring ice nucleation". I think that it the following would be more clear: "to investigate/study ice formation by deposition nucleation and by condensation freezing".

P. 476, l. 7-10: Which modifications were needed to study Immersion freezing with the FRIDGE-TAU?

P. 476, l. 13: Provide the revolutions per minute used in the shaker.

P. 476, l. 13: How accurate is this assumption? Did the authors further shake the same filter for another 30 minutes (or longer) to measure the resulting particle's concentration? Was it zero? Or, did the droplets from the new solution (i.e., the solution resulting of extra 30 minutes of shaking) freeze at the same temperature as pure water drops?

P. 476, l. 16: How were the droplets placed on the stage? Did the authors use a syringe? Please clarify it and provide the needed missing information.

P. 477, l. 25-26: What do the authors mean by freezing mode? There are several freezing modes, please clarify it.

P. 477, l. 26: (total of 10 mL without extra shaking)...

P. 478, l. 6: "...because of the decrease in the number of immersed particles per droplet, and hence a decrease in the available surface area (Pruppacher and Klett, 1997)".

Results and discussion

Why is there not any data for the 2009 and 2010 summer seasons? Although the frequency of dust storms is very low during the summer season, it may be important as a background measurement. The sampling time for each filter needs to be provided (Add to Table 1).

Using Equation 1 and assuming a sampling time of 2 hours, a droplet volume of 1 μ L and a pump flow of 20 lpm, resulted in FN values which are one order of magnitude smaller than the reported values in Figure 6. A FN concentration of 0.03/L was found when 1 drop freezes and FN is 20/L when 139 drops freeze. Is there anything wrong in this calculation?

This section can be divided into subsections (e.g., Onset freezing temperatures, Clean versus polluted days, Median freezing temperature, Ambient IN concentrations). I think that a new Figure where the IN concentrations are plotted as a function of time can be added. It will be interesting to see how the IN concentrations change every month, by seasons, and between the 2009 and 2010.

P. 479, l. 3: "The drops containing the collected ambient particles began to..."

P. 479, l. 4: Bioaerosols is too broad. It is better to be specific." ...some bioaerosols such as bacteria and leaf litters (e.g..."

P. 479, l. 3-9: The authors move back and forth between onsets and spectra. Along the paper, the authors mentioned/discussed three variables to address the IN efficiencies of their ambient particles: onset freezing temperatures, freezing at which 50% of the droplets freeze (It could be called "median freezing temperature") and also the number of immersion freezing IN. In the discussion, the above mentioned variables are combined, even in the same paragraph, making the manuscripts confusing to read.

P. 479, l. 5-6: Do the authors expect that the chemical composition of the particles measured by DeMott et al. (2006) in the Arctic are similar to the aerosol particles measured in this study? What could be the reason of the similarity between the freezing spectra from both studies?

P. 479, l. 5-12: The elemental composition of your samples will fit nicely here.

P. 479, l. 8: What is the author's definition of onset?

P. 479, l. 10: Replace "the immersion-freezing" with "ambient aerosols". The authors are investigating ambient particles from Israel and they are compared them with soot. Re-phrase it.

P. 479, l. 19-21: This is confusing. Prenni et al. (2009b) did not observed biomass burning on their TEM grids.

P. 480, l. 19 (and throughout the paper): I think it is better to use "activated fraction" instead of "activation fraction".

P. 480, l. 19-27: Why an activated fraction of one was not reach?

P. 480, l. 29: 0.6? Are the authors referring to FN>0.6. Be explicit.

P. 481, l. 4: "...role in ice formation".

P. 481, l. 15: "these" refers to clean days? It needs to be clarified.

P. 481, l. 15-16 and 21-22: This is redundant. The same point is repeated in these two paragraphs.

P. 481, l. 17-24: What happened with the other six days? Can the authors comment on it?

P. 482, l. 5: Is it 1.8C within the temperature uncertainty?

P. 482, l. 6-7: Is there any experimental evidence that montmorillonite was present in the measured ambient particles during the dusty days?

P. 482, l. 11: "that some". Is it possible to be more quantitative?

P. 482, l. 12-13: Why is it the elemental composition not shown? This is very important. The elemental composition could help the authors to interpret their data. Which was the measured NaCl mass compared to the total mass?

P. 482, l. 16-19: This is not completely true and needs to be corrected. For example, Gallavardin et al. (2008) and Wex et al. (2013) did not use neither ammonia nor ammonium sulfate. Additionally, the reduction of the IN efficiency discussed in the aforementioned studies is more related to deposition nucleation and not to immersion freezing. IN deactivation cannot be generalized. It strongly depends on the particles' composition, coating material, coating thickness, and the ice nucleation mode.

P. 483, l. 14-16: This is a very strong conclusion to make. I am not sure about the accuracy of this assumption. The IN concentrations from DeMott et al. (2010) were obtained using a continuous flow diffusion chamber (CFDC) which operates on a single particle basis. This is in contrast with the FRIDGE-TAU chamber which measures the IN concentrations from the bulk. It could be that the observed difference in the IN counts between the CFDC and the FRIDGE-TAU are due to different sensitivities in both instruments for detecting ice. It is notable that DeMott's data from the AMAZE-08 campaign (Prenni et al. (2009b)) are one order or magnitude smaller than the present observations. The IN measured during the AMAZE-08 were highly influenced by biological aerosols which are known to be very efficient ice nuclei.

Conclusions

P. 483, l. 19: "For the entire sampling period the ambient aerosol particles were found..."

P. 483, l. 19: what do the authors mean by effective?

P. 483, l. 19-20: "...-29C, with an average temperature at which 50% of the drops froze of -21C."

P. 483, l. 20-21: "FN concentrations of..."

P. 483, l. 23: what are the authors referring to by "case"?

P. 483, l. 26: "...more effective as FN. This is..."

P. 484, l. 4: "Droplet containing ambient particles from dusty..."

P. 484, l. 5: "...warmer temperatures than droplet containing particles from clean days."

P. 484, l. 10: What is the meaning of background level in this context?

Table 1: Indicate that the CPC counts are the average.

Add the standard deviations of CPC, PM₁₀ and PM_{2.5}.

Add a column where the sampling time is indicated.

Add a column where each samples is categorized as polluted or clean.

Figure 2: This figure was already published in Ardon-Dryer (2011). It must be cited. Is it possible to provide more details about this figure? A figure should be self explanatory. What is in the left and right part of the figure?

Figure 3: Can the authors add error bars in both axis of the right Figure? Figure caption needs to be re-phrased because it does not read well. It must be indicated that the spectra vary as a function of temperature.

Figure 4: I think that this figure is not necessary. I suggest replacing it with Figure 9.

Figure 5: Based on my suggestion for Figure 4, I think that it makes more sense to plot the PM10, PM2.5 and PM10-2.5 average values under clean and dusty conditions and not for the whole data set as shown in Figure 5.

Figure 6: I think that this figure is unnecessary. I suggest replacing it with Figure 10a.

Figure 7: I think that this figure is unnecessary. I suggest replacing it with Figure 10b.

Figure 9: Add error bars in the x-axis.

Figure 10: Add error bars in both axes.

Technical corrections

P. 472, l. 3: Spell out FRIDGE-TAU.

P. 472, l. 5, 8: Be consistent with decimals (i.e., -12C, 29C and -15.5C).

P. 472, l. 5-6: This is grammatically incorrect. Please re-phrase it.

P. 472, l. 7-8: Be consistent with decimals (0.32L^{-1} , 1L^{-1} and 211L^{-1})

P. 472, l. 9: "maps" is not necessary.

P. 472, l. 10: remove the word "the" between "from" and "dusty"

P. 472, l. 11: "D" in dusty should not be capitalized.

P. 472, l. 20: This is grammatically incorrect.

P. 473, l. 12: I suggest replacing the word "works" by "studies"

P. 473, l. 22: change "years" by "decades". There are several field studies that measured the IN concentration in the seventies and eighties (e.g., Bigg E. (1973), Price and Pales (1964), among others).

P. 473, l. 25: Add "temperature", i.e. "... over the temperature range of..."

P. 474, l. 3: Change "was" with "is".

P. 474, l. 23: It should be southwest.

P. 474, l. 26: Delete the word "direction".

P. 475, l. 11: I think it is better "01.2009-12.2010" than "2009-2010".

P. 475, l. 22: Replace "were" with "was".

P. 475, l. 23: Replace the word "activation" with "activated". The word "values" is not necessary.

P. 476, l. 7: Cite Ardon-Dryer et al. (2011).

P. 476, l. 12: The units are wrong. It should be megaohm · cm.

P. 476, l. 23: Spell out CCD.

P. 477, l. 20: "...found to be a reliable..."

P. 477, l. 21 (and throughout the manuscript): the "M" in montmorillonite should not be capitalized.

P. 478, l. 2: Remove the word "results".

P. 478, l. 9: The volume should be 50 mL, not 20 mL.

P. 482, l. 5: Remove the word "as".

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