Reply to Reviewer 1

We would like to thank the anonymous reviewer 1 for his helpful comments and suggestions. In line with the comments and suggestions we have revised the manuscript and made significant additions and changes. Below are all the comments (in bold) followed by the replies. The parts that are in italic are corrections that are included in the revised version of the paper:

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.

We modified the introduction based on the reviewers' comments and suggestions. Explanations on the different nucleation mechanism were added. Errors bar has been added to all the figures with explanations in the text.

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.

Figures with freezing fraction (Figures 4 and 9 from the original manuscript) have been removed from the new revised manuscript. Instead, this information will be presented in table 2 in the new revised manuscript. However we don't agree with the reviewer regarding figures 6 and 7 (in the original manuscript). These figures present the results of IN concentrations and activated fraction for all the sampling days (19 in total) and thus are valuable for comparison with the days we identified as dust storm days and as clean days. The relatively small spread in the results helps to strengthen our argument that dust particles are always present in the atmosphere in this region.

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.

We decided not to use the term meteorological conditions but rather to specify that the measurements were done during days which were classified as dust storm days and those without dust storms. As was mentioned above, the median freezing temperatures will only be presented in a table. Our reply to the question about the removal efficiency is given further down in our reply.

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.

Thanks. The sentence has been changed

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.

We have modified the whole introduction based on the comments and have added an explanation about the different nucleation mechanisms.

Ground measurements have the advantage of allowing to monitor aerosol and ice nuclei characteristics on a regular basis. In fact we have monitored ice nuclei concentrations by condensation freezing on a daily basis for over two years (to be submitted), something that is impossible to do using airborne platforms. Furthermore, it is important to note that during dust storms the atmosphere is well mixed almost all the way up to cloud levels. And indeed our ice nuclei measurements are in general agreement with measurements of ice crystals concentrations in clouds in Israel (Levin et al., 1996) and with LIDAR measurements. This does not mean that the ice nuclei measurements have one to one relations to ice crystals in clouds but it does indicate that the ground measurements are valid as indicators of the potential for ice formation.

It is interesting to point out that Kanitz et al. (2011) and Seifert et al. (2010) observed a relatively high fraction of ice in mid-level startiform clouds with cloud top temperatures as warm as -10^{0}C or even warmer when dust particles were present. This is very similar to the results of Levin et al. (1996) who reported on ice concentrations in Eastern Mediterranean convective clouds. It is also in good agreement with the present results showing the effectiveness of the mineral dust particles as freezing nuclei at such warm temperatures.

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

We have changed the sentences and added the citations.

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

We have changed the sentences.

In the last decade much attention has been given to laboratory studies on heterogeneous ice nucleation (e.g. Hoose and Möhler, 2012 and references therein)

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

Dust is important because it contributes to the atmospheric aerosol loading more than any other type of particles. It is especially important in the Mediterranean region due to the proximity to the deserts of North Africa (Lelieveld et al., 2002). It is also important because along their trajectory from the deserts, many of the particles undergo changes due to chemical processes (e.g. sulfate coating; Levin et al., 1996) and/or attachment to other particles (e.g. sea salt; Levin et al., 2005).

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))

Although biological particles are the most efficient IN, their concentrations in the atmosphere are relatively low as compared to dust aerosols.

Although biological particles have been found to be among the most efficient IN (e.g. Schnell and Vali, 1976; Levin and Yankofsky, 1983; Levin et al., 1987; Diehl et al., 2002), their concentrations in the atmosphere are relatively low. This makes them less likely to dominate the ice processes in clouds (Hoose et al., 2010). On the other hand, mineral dust aerosols are among the largest contributors to atmospheric aerosols (Goudie and Middleton, 2006). The presence of dust particles inside many ice crystals suggests that ice nucleation is often initiated by mineral dust aerosols in the atmosphere (Isono, 1955; Isono et al., 1971; Kumai, 1961, 1976; Twohy and Poellot, 2005; Cziczo 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 (RHice) 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).

We added a list with different examples of mineral dust that were found to nucleate ice as a function of the different nucleation modes

Dust particles have been observed to nucleate ice at different heterogeneous nucleation modes: deposition freezing (e.g. Möhler et al., 2006; Kulkarni and Dobbie, 2010; Kanji et al., 2013), condensation freezing (e.g. Roberts and Hallett, 1968; Levi and Rosenfeld, 1996; Zimmermann et al., 2008; DeMott et al., 2011), contact freezing (e.g. Pitter and Pruppacher, 1973; Ladino et al., 2011) and immersion freezing modes (e.g. Pitter and Pruppacher, 1973; Marcolli et al., 2007; Lüönd et al., 2010; Broadley et al., 2012; Pinti et al., 2012; Welti et al., 2012; Kanji et al., 2013).

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

We corrected the sentences

Ground measurements of IN concentration by Levi and Rosenfeld (1996) using a thermal diffusion chamber at -15°C reported similar IN concentrations to those reported by Gagin (1975). Levi and Rosenfeld (1996) found that the concentration of IN during dusty periods was more than double than those found during non-dusty periods.

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

We added a definition for condensation in the introduction

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…"

We Re-phrased the sentence

The aim of the present research is to characterize the efficiency of IN in the Eastern Mediterranean area in immersion freezing mode in dust storm days and during days without dust storms.

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

We decided not to use the term meteorological conditions but rather to specify that the measurements were done during days which were classified as dust storm days and days in which dust storms did not occur.

The characteristics of the research area

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

We removed the word.

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

We added Jan 2009 to Dec 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.

We Re-phrased the sentence.

The Eastern Mediterranean region is characterized by air masses arriving from different sources (Lelieveld et al., 2002). Many of these air masses often contain aerosols from distant and local anthropogenic sources (Levin and Lindberg, 1979; Graham et al., 2004). Some contain dust particles from the Sahara desert (Ganor, 1994; Levin et al., 2005) while others contain marine and biogenic aerosols from the Mediterranean Sea (Levin et al., 1990) and from land sources (Ganor et al., 2000).

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

We corrected this sentence.

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

This was changed to: *more common with much lower visibility and much higher aerosol loading*

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…")

The term episode was replaced with dust storms.

According to Ganor (1994) the word episode represents events with high mass loading of suspended dust. In the text we define what we mean by this in terms of the PM_{10} values and the back trajectory.

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

We replaced the dust depositing storms with dust storms

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

We divided the sentence

Dust storms are most common between December and April (Katznelson, 1970) with maxima occurrences in spring time, mainly during April (Ganor, 1994). During the summer very few dust storms occur (Ganor et al., 1991).

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

We Re-phrased the sentence

Although during dust storms mineral dust particles are present in high concentrations, such dust aerosols are almost always present in the atmosphere in this region (Levin and Lindberg, 1979).

<u>Method of analysis</u>

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

A standard deviation values were added to all the figures

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

In the Antarctica campaign reported by Ardon-Dryer et al. (2011), we were required to limit the electrical power and to minimize the weight, thus a smaller pump was used.

Previous work from our group (not published) found that a collection of 400 liters is needed to make a proper analysis. This is why we used a pump of 20LPM for 20 min for each sample.

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)?

A column was added to table 1 which describes the conditions under which the samples were collected.

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.

The sampling time was 20 min for all the filters. We clarified it in the revised manuscript.

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

The particle size is the diameter. This was clarified in the revised manuscript

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.

We thank the reviewer for this comment. We forgot to mention that on the CPC we used a Particle Size Selector (TSI, Model 376060) with a few screens that selectively remove small particles while passing through particles larger than 0.112 microns.

The aerosol total concentration was measured by TSI Condensation Particle Counter (CPC) Model 3010, which was located next to the filter sampler. In order to measure the concentration of particles in the size range of 0.11-3µm, a TSI Particle Size Selector Model 376060 with a number of screens placed in the front, were used to remove particles smaller than 0.112 microns.

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

Instead of operating we changed the word to located.

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

We corrected it.

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

Nt was 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".

We re-phrased it

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

We had to change the program controlling the temperature in order to modify the cooling rate to allow the temperature to decrease at 1°C per minute. In addition, the program controlling the camera had to be modified to allow an increased rate of picture taking.

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

The ultrasonic cleaner was Sonicor SC-52T, which operates at 60Hz.

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?

The method used in this paper assumes that the efficiency of removal of particles from the filter is close to unity. It is based on experiments (not published) that were carried out about 15 years ago. Following the comment by Vali, we decided to repeat this experiment in order to re-check our assumption. Unfortunately, since the laboratory of Prof. Levin at Tel Aviv University has been shut down a few years ago we used instead the facility made available to us at Dr. Bingemer lab at the University of Frankfurt. Although the facility is not identical to the one used in this paper, the general characteristics are similar, namely, the stage in the FRIDGE, the temperature controller and the camera are similar. On the other hand the double distilled and deionized (DDI) water is of slightly poorer quality than the one used at Tel Aviv University. Nevertheless, we decided to run a number of tests as described below with each test composed of more than 130 drops.

A volume of 384L of air containing Arizona Test Dust (ATD) particles were deposited on Nitrocellulose Membrane Black filters of 47mm diameter with 0.45μm pore size (the same one used in the paper). The filter was put into the DDI water (resistivity of 15.87 MΩ·cm) and placed for 15 min in an ultrasonic shaker, thus exposing the samples to similar conditions used in the paper (this was done following consultation with the manufacturer of the ultrasonic shaker).

Drops containing aerosols from the water were placed on the FRIDGE’s temperature controlled stage. The temperature was lowered at a similar rate to the one we used before and the temperature at which the drops froze was recorded (named ATD - after 1 ultrasonic shaker cycle). Then the filter was placed in a new test tube with fresh water and put in the ultrasonic shaker, repeating the procedure above. The freezing temperature of the drops was recorded (named ATD - after 2 ultrasonic shaker cycles). The experiment was repeated again for a third time (named ATD - after 3 ultrasonic shaker cycles). In addition to the above tests, similar procedure was carried out using clean water with no filter and a clean filter without aerosols.

In these experiments we assume that the nucleation is deterministic, namely it is enough that one nucleus is active at a certain temperature for the drop to freeze. The results of the freezing fraction after the three shaking procedures are presented in Fig.1.

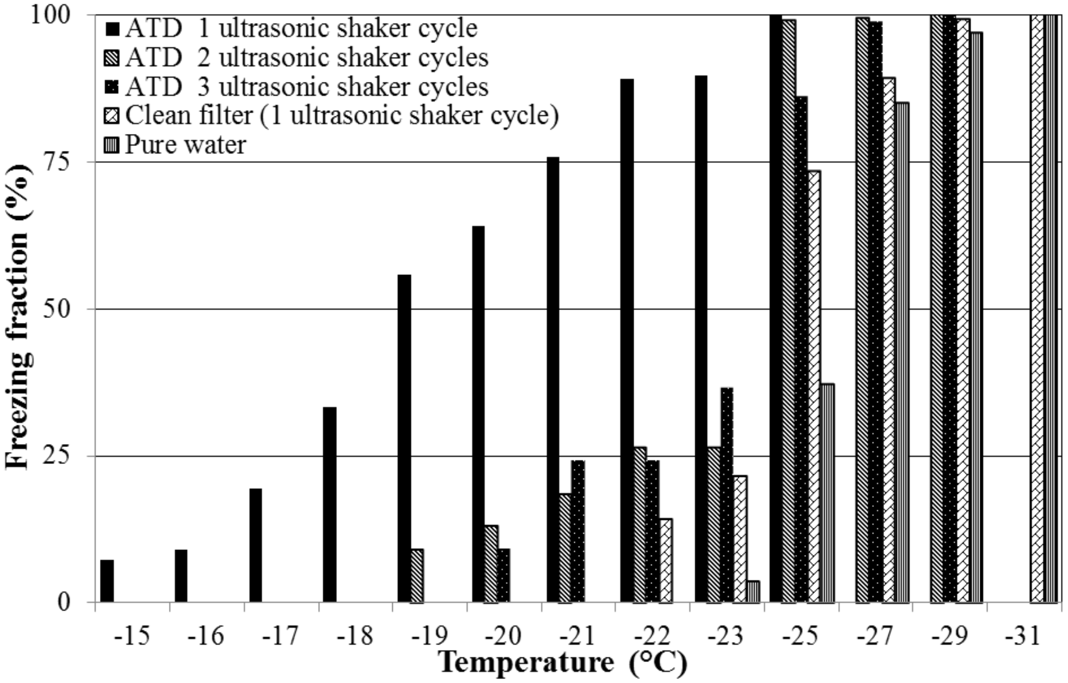

Fig 1: The freezing fraction as a function of temperature in the experiment testing the removal efficiency of the particles from the filter. In black, ATD particles after 1 ultrasonic shaker cycle, in dark downward diagonals ATD particles after 2 ultrasonic shaker cycles, ATD particles after 3 ultrasonic shaker cycle white points on the black background. Clean filter experiment (after 1 ultrasonic shaker cycle) in diagonal bricks and pure water in the narrow vertical lines.

From Fig 1 one can see that down to a temperature of about -19°C there were no freezing drops after the second and third shaking. At around -20°C about 10% of the drops froze after the second shaking and this number increased to almost 25% at -22 to -23°C. However, we can also see that the clean filter and the clean water started freezing also at the latter temperatures. This indicates that it is very likely that some of the drops frozen after the second and third procedure at these lower temperatures were actually nucleated due to the contamination in the water. This was not the case in the original experiment reported in the paper where drops started to freeze at lower temperatures (see Fig 4 in the original paper).Thus, based on the present tests it is difficult to evaluate the efficiency of removing the aerosols active at these relatively low temperatures. However, it seems that most particles active at the higher temperatures have been removed in the first run. Of course more careful experiments using different aerosols, different ultrasonic shaking times and different ultrasonic power are needed. But this will have to be done as a separate experiment.

In the revised manuscript we added a few sentences addressing this issue:

Each filter containing the collected aerosols was placed in 10ml of double distilled water (resistivity of 18.2 MΩ·cm). The aerosols were then removed from the filter by an ultrasonic shaker. The use of the ultrasonic shaker was found to be effective for particle removal into the water solution. This method which is more aggressive then the removal method used by Vali (1968) was found to be effective in removing all of the most effective particles after only one cycle of shaking in the ultrasonic bath.

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.

The droplets were put on the stage using a pipette; we have added this information to the method section.

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

We added the word immersion-freezing.

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

We added it to the sentence.

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)".

We modified the sentence as suggested by the reviewer. We do not think that the reference of Pruppacher and Klett is appropriate here.

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).

The filters were sampled during conditions of dust storms, clean and polluted days (e.g. Lag BaOmer) or just randomly to represent other “regular-background” conditions. Five samples were collected on days that do not fit our strict definitions of dust storms, clean or polluted days. Thus, the results from the dusty conditions could be compared to days without dust, which resemble summer time conditions.

It is worth mentioning that another paper is in preparation in which a two year campaign of daily IN measurements in the deposition and condensation modes using the FRIFDGE-TAU will be presented. Together with the present paper it will allow us to compare different seasons and the ice nucleating efficiency in different nucleation mode.

Regarding the sampling time, all filters were sampled for 20minutes; we added this information in the revised manuscript.

Using Equation 1 and assuming a sampling time of 2 hours, a droplet volume of 1µL 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?

In our experiments each sample was collected for 20min, not for 2 hours. Using the correct sampling time, if one drop freezes at -15°C, K’ in equation (1) is $0.175L^{-1}$. If 139 drops freeze at about -26°C than K’ is $120L^{-1}$. Both values are similar to the numbers one gets from Fig 6 (in the original manuscript). If, on the other hand, the sampling time is longer and thus the total collected aerosol mass is larger, each drop in the sample will contain a larger number of particles capable of freezing at the higher temperature. This means that when the temperature is lowered to say -15°C, not one but many drops will freeze almost simultaneously. Of course in such a case, the sample should be diluted in more than 10ml of water and probably many more drops would be needed to improve the resolution.

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.

We made many changes in the revised manuscript. There will not be a section that describes in detail the onset freezing temperatures and median freezing temperature. Instead, this information will be presented in table 2 in the revised manuscript. In addition, following the comments by the reviewers we have added a new section that covers the Lag Ba Omer event. Furthermore, an additional explanation on the connection between ice nucleation and particle’s surface area has been added

Since we had only 19 days, we do not think that having a plot of IN concentrations as a function of time is informative enough.

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

The sentence was changed.

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

We accept the change and added it.

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.

We have changed the new revised manuscript, the onset freezing temperatures and median freezing temperature will only be presented in table 2 in the revised manuscript. We focus our attention on the IN concentrations and the activated fraction.

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?

We have taken this part out of the paper based on the comment and suggestion in the review of Paul DeMott (reviewer 2).

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

A section with elemental composition analysis that we had for two days, was added to the revised manuscript.

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

Onset represents the temperature at which the first drop freezes. We will clarify it in the new revised manuscript.

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

The sentence was changed.

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

We agree with the reviewer, this citation of Prenni et al. (2009b) was taken out of the new revised manuscript.

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

We agree with the reviewer. This correction will be made in the new revised manuscript.

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

In laboratory experiments using known particle composition and size, one may be able to get activated fraction close to unity. However, ambient aerosol samples that are composed of particles of different composition and size, with only a few that are very good IN, cannot give an activated fraction of one.

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

The FN concentration and AF values were found to increase with decreasing temperatures at a relatively high correlation coefficient of >0.6.

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

The word “the” was deleted.

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

The sentence was changed.

Samples were defined as clean days when PM_{10} *daily average values and the value measured during the aerosol sampling were below 50 µg* m^{-3} *(Ganor et al., 2009) and the air mass trajectory in the previous 72 hours did not pass over a source of dust.*

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

These paragraphs were changed.

The filter samples were separated into dust storms and clean conditions based on PM_{10} *values and the air mass back trajectory. The Back Trajectories (BT) were calculated for each measurement using the HYSPLIT method (Hybrid Single Particle Lagrangian Integrated Trajectory Model). Dust storm days were defined as days when the* PM_{10} *daily average values and the value measured during the aerosol sampling time exceeded 100µg* m^{-3} *(Ganor et al., 2009). In addition, the air mass trajectory in the previous 72 hours had to have originated over a dust source or passed over one. Samples were defined as clean days when* PM_{10} *daily average values and the value measured during the aerosol sampling were below 50 µg* m^{-3} *(Ganor et al., 2009) and the air mass trajectory in the previous 72 hours did not pass over a source of dust. It should be noted that in the research area the yearly average standard values of* PM_{10} *is 60µg* m^{-3} *(Israel Ministry of Environmental Protection, 2013).*

Out of all the days that were sampled, eight days were defined as dust storm days with daily average values of PM_{10} *from 254 to 867µg* m^{-3}*, with an overall average of 527±236. Five days were defined as clean days, with* PM_{10} *daily averages ranging from 30 to 39µg* m^{-3} *with an overall average of 34±3.8, as can be seen in Table 3. In the clean cases the air mass arrived* *from the west or northwest, while on the dust storms days the air mass arrived from the south or southwest, as can be seen in Fig. 6.*

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

The other days were not considered clean or dust storm days because they did not fit the strict criteria of PM values with air trajectory.

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

The difference of 1.8°C is larger than the uncertainly in temperature. In fact T test calculations show that the onset of freezing and the median freezing temperature between the clean days and dust storms days were significantly different from one another.

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

This is based on measurements of Ganor (et al., 2009), who found a high frequency of montmorillonite particles in dust storms in our area.

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

The whole paragraph will be deleted in the new revised manuscript.

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?

Although this paragraph will be deleted in the revised manuscript, we added a paragraph discussing the elemental composition analysis that was carried out.

P. 482, l. 16-19: This is not completely true and needs to be corrected. For example, Gallavardin et al. (2008) and Wex et el. (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.

We decided to take out this paragraph from the new revised manuscript.

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 FRIDGETAU 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.

Paul DeMott also requested that we change this paragraph. In the revised manuscript we removed figure 11 (in the original manuscript) and thus removed the comparison with some of the references mentioned in this paragraph.

Conclusions

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

We changed the sentence.

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

By effective we mean that the FN froze the drops in this temperature range.

Drops containing ambient aerosols were found to freeze between -11.8ºC down to -28.9ºC, with median freezing temperature that varied from -17.8ºC down to -24.4ºC.

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

See our correction in the comment above.

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

We changed the sentence.

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

We changed the sentence.

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

We changed the sentence

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

We changed the sentence.

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

We changed the sentence.

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

The sentence has been modified:

This observation agrees with previous studies showing that some dust particles are almost always present in the atmosphere in this region.

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

Add the standard deviations of CPC, PM10 and PM2.5.

We added standard deviations for CPC, PM_{10} and $PM_{2.5}$ values.

Add a column where the sampling time is indicated.

The sampling time was the same for all filters. We added this information in the Method section of the new revised manuscript.

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

A column that described the classification of each filter samples was added to table 1.

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 or the figure?

Following the recommendation of the reviewers, we removed this figure and only made reference to Ardon-Dryer et al. (2011).

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

Error bars have been added to the figure with an explanation in the text.

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

The figure was deleted from the new revised manuscript.

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.

Since not all the filter samples were classified as clean or dust storm days, we believe that showing this figure could be important. This figure shows that when PM_{10} increases there is a higher chance of finding more effective IN particles.

In addition, the average values of PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ under clean and dusty conditions were added to table 3 in the new revised manuscript.

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

Not all the days were dust storms or clean days, therefore we thought it would be interesting to show the range of IN concentration and activated fraction on the different days.

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

See our reply to the comment above.

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

The figure was deleted from the new revised manuscript but error bars were added to all the other figures.

Figure 10: Add error bars in both axes.

Error bars were added to the figure.

Technical corrections

All the technical corrections have been adapted and inserted into the new revised paper.

REFERENCES

All the references mentioned in this reply have been added to the revised version of the paper.

Reply to Paul DeMott

We would like to thank Paul DeMott for his helpful comments and suggestions. In line with the comments, we significantly revised the manuscript. Below are all the comments (in bold) followed by the replies. The parts that are in italic are corrections that are included in the revised version of the paper.

General Comment

One primary concern is to note the expectation upfront 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.

We agree with the comment that in this research area dust particles are commonly present in the atmosphere, although their concentrations depend mostly on the meteorological conditions and on the wind direction. We do not think it would be correct to compare our clean days with those existing in other areas, which are far away from dust sources (as Europe or the US). In addition, it is important to mention that in contrast to many other locations dust particles in our region are often coated with soluble material such as sulfate or sea salt. This is why it is important to study the ice nucleating properties of these particles even on “clean” days.

In the revised manuscript we added a section (4.1.1) describing the method we used to estimate the dependence of the ice nuclei active surface (INAS) as a function of temperature and compared the results to previous publications.

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.

In light of these constructive comments we replaced the plot from DeMott et al. (2010) with a comparison of our results from immersion freezing and the measurements of ice nucleation by condensation freezing taken in the same research area (Levi and Rosenfeld ,1996; Gagin, 1975).

We replaced Fig 11 in the original manuscript with the figure below, which is appears as Fig 7A.

Fig. 7A also shows a comparison of FN concentration from dust storms and clean conditions and the measurements of ice nucleation by condensation freezing taken in the same research area by Levi and Rosenfeld (1996) and Gagin (1975). The figure shows some agreement between the two modes of nucleation at the higher temperatures while there are differences of about one order of magnitude at lower temperatures. One possible explanation for this difference is that immersion freezing is more effective than condensation freezing. On the other hand, it is also possible that the difference is a result of the measuring method.

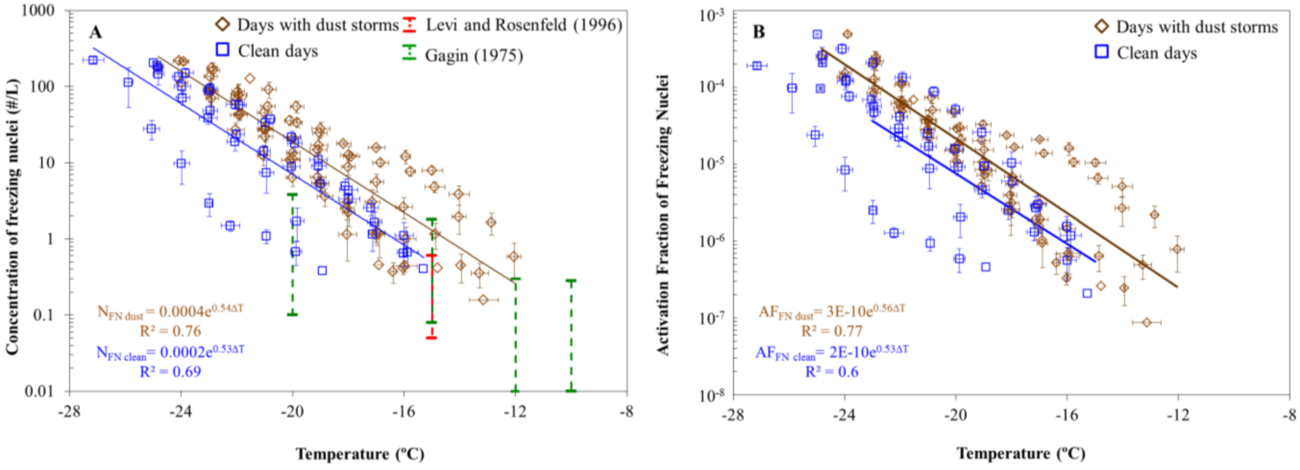

Fig. 7: Freezing nuclei concentration (A) and activated fraction values (B) with standard deviation, calculated for clean (blue) and dusty (brown) days. Best-fit lines and the equations that represent them are also shown. For comparison in (A) the ice nuclei concentrations measured in Israel near cloud base by Gagin (1975) (green bars) and ground measurements by Levi and Rosenfeld (1996) (red bars) are also shown.

Specific Comments

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.

See our reply to the last comment.

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.

The term episode was replaced by“dust storms“.

According to Ganor (1994) the word episode represents an event with high mass loading of suspended dust. In the text we defined what we mean by this in terms of the PM_{10} values and the back trajectory.

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.

We thank the reviewer for this comment. We forgot to mention that on the CPC we used a Particle Size Selector (TSI, Model 376060) this device selectively removes small particles while passing through larger particles. In this work a number of screens were used in the Particle Size Selector in order to count only particles above 0.112micron.

The aerosol total concentration was measured by TSI Condensation Particle Counter (CPC) Model 3010, which was located next to the filter sampler. In order to measure the concentration of particles in the size range of 0.11-3µm, a TSI Particle Size Selector Model 376060 with a number of screens placed in the front, were used to remove particles smaller than 0.112 microns.

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.

All experiments were conducted in the same way. Each part of the experiments was done using the same methods and with the same tools regardless whether the test was on sampled aerosols, clean filters or clean water. The same DDI water was used for all the experiments. Each test tube was washed with DDI before adding the DDI and the filter to it. The filters were cut with the same scissors, after the latter was cleaned with ethanol. The use of the ultrasonic bath for all filters was done in the same way. FRIDGE plate was cleaned with ethanol and the same amount of Vaseline was placed on the chamber’s plate before the droplets were put on it. In addition, the same clean pipette was used to put the droplets on the chamber’s plate. The droplets were placed in the chamber by the same person.

The main reason this was mentioned only on page 478 is due to the fact that this is the first time in this paper that we present the results. Whenever drops in the samples froze at the same temperature as the drops from the clean filter (usually below -23°C) these drops were removed from the calculation. This is because we could not be sure if those droplets froze due to the presence of effective ice nuclei in the sample or due to the presence of material that was released from the filter during the ultrasonic shaking.

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 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.

We thank the reviewer for the clarification and this comparison will be deleted from the new version.

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.

The intention of presenting the Lag BaOmer data was to illustrate that particles produced by burning of very dry Finish Pine wood in low temperature fires, are relatively poor freezing nuclei.

Following the comments by all three reviewers we have added a separate section (4.2) with more information on the Lag BaOmer event. The section includes information about aerosol concentration and the type of wood that is commonly used.

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.

On May 1, 2010, samples were collected at 15:00 and 23:00 local time. The sample at 15:00 was collected before the start of the bonfires and the sample from 23:00 was taken during the Lag BaOmer event itself. The Lag BaOmer event started around 19:00 and by 23:00 the atmosphere contained many biomass burning particles. The reason no filter samples were taken on the day after Lag BaOmer (May 02 2010) was because on May 2 at around 02:30 am local time rain began to fall clearing the atmosphere.

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.

Following this comment, we modified the revised manuscript as can be seen in section 4.2 of the new revised manuscript.

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.

The wood type that is most commonly used in these fires is very dry pine wood from Finland, mostly used in construction. The temperature of the fire is indeed much lower than big forest fires and it is classified as Type A. This point is discussed in section 4.2 in the new revised manuscript.

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?

Most Lag BaOmer events are characterized with a sharp increase in the aerosol concentration as compared to the concentration measured prior to the event itself (see the figures 10 in the new revised manuscript). This Lag BaOmer event did not have high particle concentrations as in previous years, because during this event the atmosphere was unstable leading to rain a few hours after the beginning of the bonfires.

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?

The exposed filter at 23:00 was definitely darker than the one from 15:00. It should be mentioned that the filters collected on dusty days sometimes appeared yellow and sometimes appeared dark brown due to the mixture of mineral dust and pollution.

Following this question we conducted a number of tests to evaluate the efficiency of removing particles form the filters. Although the water we used this time was much poorer than the ones we used in this paper, we observed that the ultrasonic shaking method is very effective in removing the particles from the filters. In fact, the number of immersion freezing nuclei that were active down to about -20C was reduced to almost zero after the first shaking procedure. Below this temperature it was difficult to separate the role of the particles in the sample from those of the clean water. See our reply to reviewer 1.

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?

An exponential equation represents the best fit for all the data. This fit was found to best represent all the data.

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 spread is increased.

As was mentioned before, the minimum size is 0.11 micron. A correction is added to the paper (see comment 3). Although we hope that caution will be used in using the best fit line, it is important to note that activtied fraction is a good indicator of ice nuclei concentrations. We are aware that some publications, including many by the reviewer himself, found a good correlation with active sites on the particles' surface, especially those larger than 0.5 microns. However, this empirical correlation does not necessarily exclude the possible connection with total concentration of particles larger than 0.1 microns. In any case, as discussed in the reply to the general comment above, we did calculate the INAS for sizes larger than 0.5 microns and also found a good agreement with other published works.

9. Page 481, Section 4.1: As someone living in an area where 50µgm^{-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.

The Israeli annual average standard of PM_{10} is higher than other locations ($60 \mu g/m^3$ $year^{-1}$) due to the present of dust particles. Although days with dust storms contains higher concentration of PM_{10} particles compared to clean cases, and even higher concentration of large particles ($PM_{10-2.5}$), we could not find a direct correlation between the PM_{10} and IN concentrations or even between the PM_{10} values and the activated fraction. However a good correlation was found between PM_{10} and the temperature in which the first freezing occurred and with the temperature in which 50% of the droplets froze. This may suggest that as the PM_{10} increases there is a higher chance of finding more effective IN particles.

The connection of the surface area with particles larger than 0.5 was discussed above (see reply to the first general comment)

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.

In line with the comments by the reviewers we modified the paper and included only a very small summary on the freezing fraction and focused more on the IN concentration and activated fraction.

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 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.

We agree with the reviewer that the effect of the solute is unimportant in the present case where a few micrograms of material are diluted in 10ml of water. This part has been deleted form the manuscript.

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.

This paragraph will be deleted from 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.

This figure was modified based on the reviewer's comments; see the reply to the second general comment for the new figure.

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.

See our reply to the first general comment.

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

The figure will be taken out of the paper.

Technical Corrections

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

The sentence has been modified

Difficulties arise in quantifying the mechanisms of ice nucleation because of the varied composition, surface characteristics and size distributions of the IN (Kanji et al., 2011).

Page 473, line 1: Capitalize M in DeMott.

Will be changed

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."

The paragraph has been modified as can be seen in comment 6

References:

All the references mentioned above have been included in the revised manuscript.

Reply to reviewer 3

We would like to thank the anonymous reviewer 3 for his helpful comments and suggestions. In line with the comments and suggestions, we revised the manuscript and made significant additions and changes. Below are all the comments (in bold) followed by the replies. The parts that are in italic are corrections that are included in the revised version of the paper:

General Comments:

[1] Several instances in the paper compare either initial freezing, 50% frozen fraction, or overall activation fraction curves in the FRIDGE-TAU experiments to previous studies on ice nucleation of montmorillonite, biological particles, soot, and biomass burning. While FN concentrations have been normalized per liter of air, special care must be taken when comparing freezing temperatures between experimental methods, especially when comparing freezing temperatures in laboratory experiments. For example, the reviews by Hoose and Mohler (2012) and Murray et al. (2012) have shown that normalization by particle surface area does lead to a convergence between different methods for immersion freezing. Thus, if possible, an estimation of the surface area loading would be appreciated. In theory, this should be possible by using particle number mass concentrations (Maynard, 2003). Furthermore, this analysis would allow direct comparison of these results to previous laboratory studies on well characterized dust sources in the immersion mode. This last step would allow the authors' better evidence to support their claim that dust particles are the IN responsible for ice nucleation on both "dusty" and "clean" days.

We took into account the reviewer's comment regarding surface area for comparison with literature; therefore, we calculated the size distributions for all our dust storm cases and calculated the ice nucleation active surface site (INAS) densities. This new material appears in the revised manuscript, section 4.1.1.

[2] The Lag Ba Omer festival that provides an interesting case study for a type of biomass burning aerosol; however, the authors may be overgeneralizing their results. As noted in the text, the work by Petters et al (2009) indicates that both the fuel type and burning conditions alter FN concentrations from biomass burning aerosol. Although the type of

wood and fire intensity was mentioned, it might be instructive to provide more details about the fuel type and the normal combustion conditions of the bonfires. Also, interestingly, initial freezing temperatures for the 1 May 2010_23 Lag Ba Omer experiments were lower than the average "clean" day initial freezing temperatures, which could point to a coating mechanism at these warmer temperatures. This should also be discussed.

Based on the comments from all three reviewers we have decided to add more information on the Lag BaOmer event including the aerosol concentration and information about the type of wood that is commonly used. The material appears in the revised manuscript, section 4.2.

Regarding the comparison with the clean days as was suggested by the reviewer, we cannot determine if the particles were coated since we did not analyze the chemical composition of these particles.

3] Finally, the FN concentration discussion could be greatly enhanced by an analysis of the temperature error associated with the FRIDGE-TAU chamber for immersion freezing. One easy way to do this would be to report the experimental error determined by the days where two frozen fraction curves were obtained by cutting the filter in half. This would give more credence to the montmorillonite data as well as the "dusty" vs "clean" days.

We added the temperature standard deviation values to each of the figures. As was mentioned in the paper, each experiment contains about 130 droplets. Since it is not possible to put all the droplets at the same time on the FRIDGE-TAU plate the experiment was split into 4-6 experiments. In each experiment about 20-30 droplets were tested. The final spectra are the combined results from each sample. The standard deviation was computed from these combined results.

Specific Comments:

Page 472, line 23: It may be more useful to cite the review of Hoose and Möehler (2012) here instead of listing these citations. If the citations are kept, it may be more useful to state what type of IN were examined in each study (i.e., dust, soot, etc.).

We have changed the introduction and we cite the paper of Hoose and Möhler (2012).

Page 473, line 4: Perhaps the author could be more specific about why these studies have "contributed a lot to our understanding of IN distributions in different parts of the world?"

Most of the scientific papers focus on laboratory work or field work in regions other than the Mediterranean. It turns out that the eastern Mediterranean is a crossroad for the transport of pollution and dust (Lelieveld et al., 2002). In addition, it has been shown that many of the dust particles are coated with soluble material such as sulfate and sea salt (Levin et al., 1996, 2005). These internally mixed particles could have a significant effect on the ice nucleation properties of the particles. This is the reason it was valuable to conduct such measurements in the eastern Mediterranean.

Page 475, line 20: Could you provide an estimate how much the total number concentration is underestimated for ignoring particles smaller than 110 nm and greater than 3μm in this region. Alternatively, you could be more explicit that you are likely calculating activated fractions for particles ≥110 nm?

Although we do not have measurements that include sizes larger than 3 microns, we can use the size distributions that were published in the past to estimate the ratio of particles in the 5-10 microns to those of 0.1-3 microns. The figure in Levin et al. (1980) can be used for this purpose. In it one can see that the concentrations of the 10 micron dust particles are more than two orders of magnitude smaller than the 3 micron particles. Thus, the surface area of the former is one order of magnitude smaller than the latter.

Similarly, from the same figure we can only assume that the concentration of the 0.01 microns particles is one order of magnitude greater than that of 0.1 microns. This implies that the surface area of the former is also one order of magnitude greater than the latter. Since many publications point to the fact that high correlation exists between ice nuclei concentrations and surface area of particles larger than 0.5 microns, the contribution of the smaller particles is very small. Similarly, although the surface area of each particle larger than 3 microns is large, their concentrations is much smaller and thus their contribution to the IN concentration is much smaller than the particles of sizes between 0.11 and 3 microns.

Page 476, line 26: The consistency of these results, however, was not reported. See general comment [3].

The main reason only half of the filters were used in most of the experiments was in order to allow us to use the other half as backup to the drop freezing experiment in case something went wrong. In some cases the other half was used for elemental analysis using the Environmental scanning electron microscope (ESEM).

In most cases the filters were cut in half before placing them in 10 ml of double distilled water. This was done in order to be able to duplicate the measurements if needed. In some cases the unused half of the filter was used for elemental analysis of individual particles with the Environmental Scanning Electron Microscope (ESEM) with an attached X-ray energy dispersive system (EDS).

Page 478, line 9: Here is one instance where surface area estimations would be useful to compare between experimental methods. See general comment [1].

See reply to comment [1].

Page 478, line 26: Is it valid to remove these points from your analyses? While some temperatures in your experiments overlap with the temperatures at which some particles froze during blank/pure water experiments, the frozen fractions are much different. As you

mention, the average shift is small, only 0.18°C, but it will greatly affect the results for some of the colder frozen fraction curves.

We believe that the procedure we used is appropriate to eliminate the possibility that some of the drops froze due to “contamination” from the blank filter and/or the water. Therefore, the same number of drops from the samples that froze at the same temperature as the clean filters, were deleted from the calculation. As was pointed out, the shift due to the removal of these drops was very small.

Page 479, line 3: This entire paragraph is another instance where surface area estimations would be useful compare between experiment methods. See general comment [1].

See reply to comment [1].

Page 479, line 19: As mentioned in general comment [2], the type of burning fuel will influence FN concentrations. Thus, the comparison to the study in the Amazon by Prenni et al. (2009) may not be valid and the conclusion “particles from biomass burning are not a likely source of effective ice nuclei” may be overstated.

This comparison was taken out of the revised manuscript, See section 4.2 in the revised manuscript.

Page 479, line 25: As mentioned in general comment [2], it would be helpful to expand upon the type of construction wood and the bonfire combustion conditions.

This information was added to the revised manuscript in section 4.2.

Page 482, line 5: Here is another example of why an estimation of the temperature error associated with FRIDGE-TAU immersion freezing experiments may be important.

The temperature error had been added to the figure and the text

Page 482, line 7: Again, surface area estimations would be useful to compare between experimental methods. See general comment [1].

See reply to general comment [1].

Page 482, line 12: How were these elemental compositions determined?

The elemental compositions were determined by single particle analyses using the Environmental Scanning Electron Microscope (ESEM) with an attached X-ray energy dispersive system (EDS). This point is elaborated on in the revised manuscript and the results are presented in table 4.

Page 483, line 2: This statement could be greatly enhanced by an estimation of the surface area. If an estimate can be provided, then the freezing results from both dusty and clean days can be directly compared to previous laboratory studies on dust proxies (Murray et al., 2012).

See reply to comment [1].

Technical Corrections:

All the technical corrections have been added to the revised manuscript.

References:

All the references mentioned above have been included in the revised manuscript.