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.

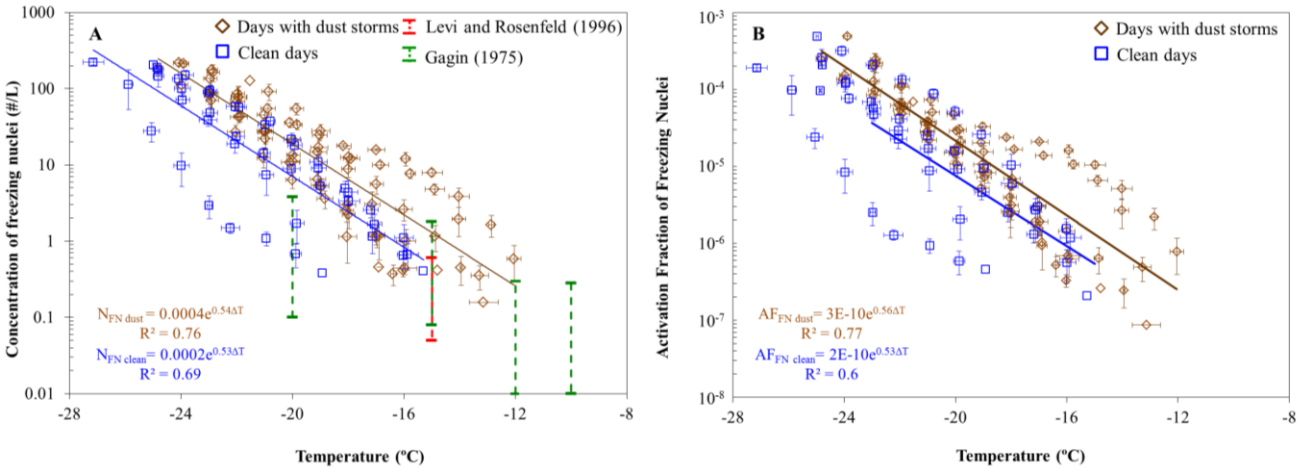

Fig. 7: Freezing nuclei concentration (A) and activated fraction values (B) with standard deviation, calculated for clean (blue) and dusty (brown) days. Best-fit lines and the equations that represent them are also shown. For comparison in (A) the ice nuclei concentrations measured in Israel near cloud 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 (60µg/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.