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.