Reply to the comments by Gabor Vali

The authors would like to thank Prof. Gabor Vali for his comments, which help improve our paper. 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:

2. The crucial question is how effective is the removal of INPs from the filter. There are other lesser issues as well. The removal efficiency was assumed to be unity in this paper, based on the vigorous and lengthy application of mechanical liberation of particles from the filter. How did the authors settle on the degree of agitation to be used? Vali (1968) used reverse flushing of the filter and determined of the efficiency of removal by repeating the process several times with the same filter and thus observing the decrease of transfer rate with time. A time integral of the total transfer (exponential with time) compared with the first rinse yielded a correction factor that could be applied to the measurement obtained with the first sample. This is a valid approach to any method of particle liberation used and it would be useful to make such tests in order to eliminate reliance on the assumption of full transfer efficiency. Of course, the removal efficiency can be expected to be a function of the size and type of aerosol being tested. The decay rate could be checked for a number of tests with different aerosols and a range of rates established. Then, it would be desirable to make at least two extractions for every sample and thereby obtain an indication of the decay rate for the particular sample. This would reduce significantly the uncertainty associated with the method. The accuracy so achieved could be stated clearly in terms of probable errors.

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 recheck 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.

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.

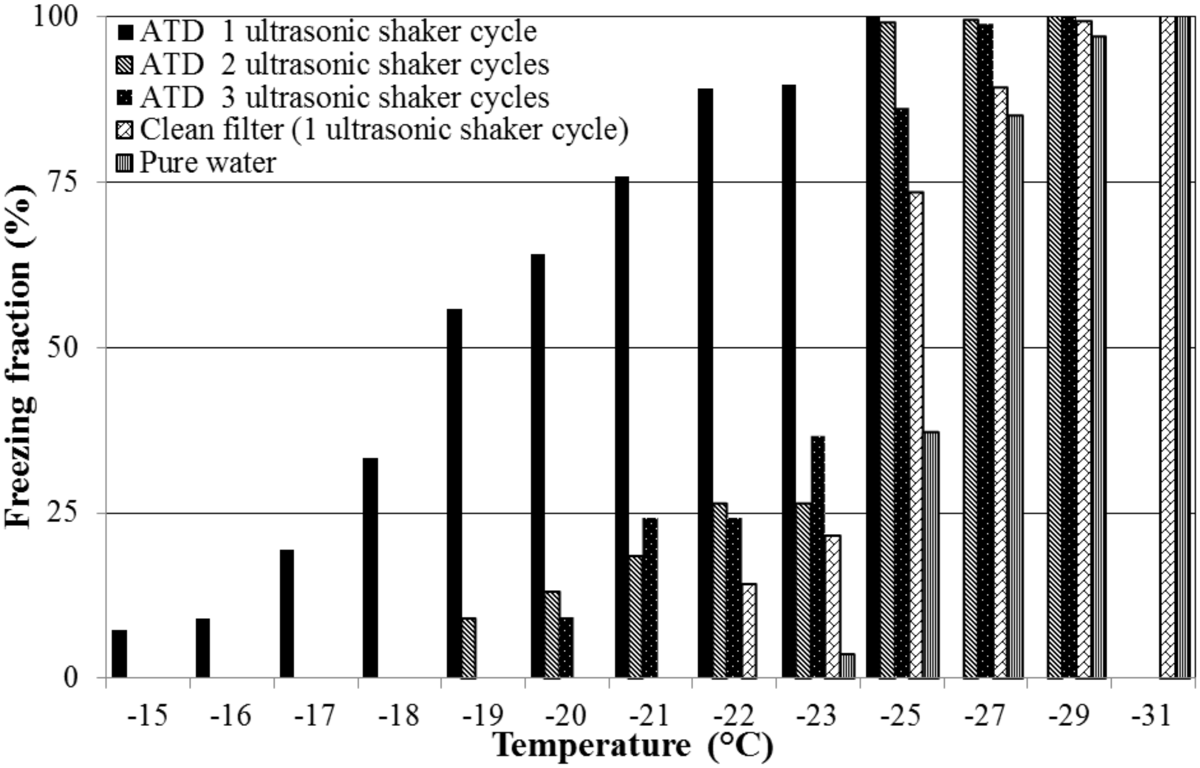

Fig 1: The freezing fraction as a function of temperature in the experiment testing the removal efficiency of the particles from the filter. In black, ATD particles after 1 ultrasonic shaker cycle, in dark downward diagonals ATD particles after 2 ultrasonic shaker cycles, ATD particles after 3 ultrasonic 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.

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

Each filter containing the collected aerosols was placed in 10 ml of double distilled water (resistivity of 18.2 MΩ·cm). The aerosols were then removed from the filter by ultrasonic shaker. The use of the ultrasonic shaker was found to be effective for removal particle 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 the most effective particles after only one cycle use of the ultrasonic shaker bath.*

3. As a minor point, it is worth emphasizing that values as that temperature at which 50% of the drops were frozen is not the best way to quantitate the results. Drop size and dilution influence these values. Concentrations at given temperatures are more definitive measures. Both types of data are given in this paper; deleting the former would not detract from the information presented.

We thought that using the 50% of drops frozen could be a way to compare with others. But based on the Vali's comment and the similar suggestion by one of the reviewers, we eliminate this point from the revised manuscript.

4. The comparison of the data in this paper with those of Bigg and Stevenson (1970) need to be viewed bearing in mind the differences in techniques, The difference cannot be associated with a true trend due to location or timing of the samples.

We agree that the method used in Bigg and Stevenson (1970) is different from the one used in this work. In the revised manuscript we indicated that Bigg and Stevenson measured ice nuclei concentrations by condensation freezing under water saturated conditions. However, it is still interesting to note the difference in ice nuclei concentrations with temperature.

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

Equation 2 (in the new manuscript) gives a concentration of $1L^{-1}$ at -15.3ºC.

This temperature is higher than the average temperature obtained by Bigg and Stevenson's (1970) measurements of condensation freezing that were taken around the world. These measurements are also higher than the condensation freezing measurements of Gagin (1975) that were observed in Israel (~$1L^{-1}$ at -18.4ºC).

5. The summary graph, Fig. 6, prompts the question why the spectra from the various samples show lesser spread at the lower end of the range than at higher values. A rough estimate is that 90% of the spectra fall within about a factor 5 at -22°C, while the spread is more like a factor 50 at -18°C. Similar trends are seen in many other data sets. Yet, on the average, the dusty and clean days differ to about the same degree all through the temperature range.

The spread at the higher temperature is probably a result of the lower concentrations of effective immersion freezing nuclei at these temperatures. In other words, due to their lower concentrations, their distribution is not homogeneous in the drops. This is the reason we needed a fairly large number of drops in each experiment. Since this study was part of monitoring ice nuclei spectra almost on a daily basis, we had to limit the number to about 130-150 per run. At lower temperatures, higher concentrations of particles are active as freezing nuclei, thus the probability that each drop contains at least one such nuclei increases, leading to a smaller spread in the results. We added a short explanation about it in the text