



Ground based  
measurements of  
immersion-freezing

K. Ardon-Dryer and  
Z. Levin

# Ground based measurements of immersion-freezing in the eastern Mediterranean

K. Ardon-Dryer<sup>1,2,\*</sup> and Z. Levin<sup>1,3</sup>

<sup>1</sup>Department of Geophysical, Atmospheric and Planetary Sciences, Tel Aviv University, Israel

<sup>2</sup>The Porter School of Environmental Studies, Tel Aviv University, Israel

<sup>3</sup>Energy, Environment and Water Research Center, The Cyprus Institute, Nicosia, Cyprus

\*now at: Department of Earth, Atmospheric and Planetary Sciences, MIT, Cambridge, M.A, USA

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Correspondence to: K. Ardon-Dryer (karinard@mit.edu)

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## Abstract

Ice Nuclei were measured in immersion-freezing mode on the Tel Aviv University campus using the FRIDGE-TAU chamber. The sampling was done under different meteorological conditions and air pollution levels and then analyzed in the laboratory using a drop freezing method. The droplets froze from  $-12^{\circ}\text{C}$  down to  $-29^{\circ}\text{C}$ , the average temperature at which 50 % of the drops froze occurred at  $-21^{\circ}\text{C}$ . Immersion-Freezing Nuclei (FN) concentrations between  $0.32\text{ L}^{-1}$  to  $211\text{ L}^{-1}$  were found with a concentration of  $1\text{ L}^{-1}$  observed at  $-15.5^{\circ}\text{C}$ . The measurements were divided into dusty and clean conditions based on their back trajectory maps and aerosol mass concentrations ( $\text{PM}_{10}$ ). Droplets from the dusty days froze at warmer temperatures than from clean days. The Dusty days had higher FN concentrations and active fraction (AF) values than clean days.

## 1 Introduction

Ice in clouds plays an important role in the development of precipitation and in affecting the planet's albedo. However, in spite of its importance the mechanisms that are responsible for its formation are not yet clear. Furthermore, the concentrations and physical properties of the particles known as ice nuclei, IN, vary greatly from place to place and with weather conditions. Part of the gap in knowledge comes from the lack of understanding or quantification of ice formation processes in the atmosphere (Cantrell and Heymsfield, 2005). Difficulties arise in quantifying because of the varied composition, surface characteristics and size distributions of the IN (Kanji et al., 2011). As a consequence, in the last decade much attention has been given to laboratory studies on heterogeneous ice nucleation (e.g. Gorbunov et al., 2001; Zuberi et al., 2002; Diehl and Wurzler, 2004; Archuleta et al., 2005; Shaw et al., 2005; Möhler et al., 2006; Marcolli et al., 2007; Salam et al., 2007; Zimmerman et al., 2007, 2008; Bundke et al., 2008; Vali, 2008; Petters et al., 2009; Welti et al., 2009, 2012; Lüönd et al., 2010; Niedermeier

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et al., 2010; Demott et al., 2011; Kanji et al., 2011; Jones et al., 2011; Pinti et al., 2012; Broadley et al., 2012). Field studies (e.g. DeMott et al., 2003a, b; Prenni et al., 2009a, b; Santachiara et al., 2010; Klein et al., 2010a; Ardon-Dryer et al., 2011; Conen et al., 2012) have contributed a lot to our understanding of IN distribution in different parts of the world. In addition, analysis of the chemical composition of IN residuals from air-borne measurements using electron microscopy and mass spectroscopy (e.g. Seifert et al., 2003; Cziczo et al., 2003, 2006, 2013; Froyd et al., 2010) have added a deeper understanding about the nature of some of the IN in the atmosphere.

Dust particles are among the most effective and important natural ice nuclei (DeMott et al., 2003b; Field et al., 2006; Knopf and Koop, 2006; Zimmermann et al., 2008; Niedermeier et al., 2010; Chou et al., 2011; Murray et al., 2011). Many field and laboratory works have shown that dust particles are effective IN at relatively warm temperatures (nucleation can start at  $\sim -10^{\circ}\text{C}$ ) for different heterogeneous nucleation modes (e.g. Roberts and Hallett, 1968; Levi and Rosenfeld, 1996; Marcolli et al., 2007; Kulkarni and Dobbie, 2010; Lüönd et al., 2010; DeMott et al., 2011; Pinti et al., 2012; Welti et al., 2012; Broadley et al., 2012; Kanji et al., 2013). Most articles show an increase of IN concentration during dust storms (e.g. Bowdle et al., 1985; DeMott et al., 2003a; Van den Heever et al., 2006), with increases as high as double (e.g. Levi and Rosenfeld, 1996) or even five times higher (e.g. Klein et al., 2010a) as compared to dust free conditions.

Results from extensive IN measurements around the world have been published in the past few years however, only very few were reported from the eastern Mediterranean (e.g. Gagin, 1975; Levi and Rosenfeld, 1996). Gagin (1975) sampled IN concentration at the bases of cumulus clouds using filter methods which were exposed to water saturation in a thermal diffusion chamber over the range of  $-5^{\circ}\text{C}$  to  $-25^{\circ}\text{C}$ . Ground measurements of IN concentration by Levi and Rosenfeld (1996) using the same instrument as above but at  $-15^{\circ}\text{C}$  and water saturation reported on similar IN concentrations, which were more than double in the dusty period. The two measurements reported above were for condensation freezing condition. So far no measure-

ment of IN in immersion-freezing mode have been reported from the eastern Mediterranean area.

The aim of the present research was to characterize the efficiency of IN in the eastern Mediterranean area in immersion-freezing mode under different meteorological conditions and air pollution levels.

## 2 The characteristics of the research area

The measurements were conducted during 2009–2010 in the eastern Mediterranean region on the Tel Aviv University campus, located at the north part of Tel Aviv, Israel (Fig. 1). The sampling was conducted on the roof top of the Department of Geophysical, Atmospheric and Planetary Sciences (32°6′46.7″ N 34°48′22.9″ E), about 20 m above ground, 60 m a.s.l. and about 2.5 km from the seashore.

The eastern Mediterranean region is characterized by air masses arriving from different sources and containing a variety of aerosol types, such as anthropogenic aerosols originating from local industrial pollution or from distant sources such as Europe (Levin and Lindberg, 1979; Graham et al., 2004), dust from the Sahara desert (Ganor, 1994; Levin et al., 2005), marine aerosol (Levin et al., 1990) and even biogenic material from land or marine sources (Ganor et al., 2000).

Air masses reaching Tel Aviv from the northwest carry a larger fraction of marine aerosols, mainly from sea spray and anthropogenic aerosols and a relatively small fraction of desert particles. Air masses reaching Tel Aviv from the southwest (Sahara and North African deserts), carry a larger fraction of desert and marine aerosols with a smaller fraction of anthropogenic particles (Levin et al., 1990, 2005). Dust also reaches Israel from the east, although the dust from the North African (south west) deserts is more common and more intense (Ganor et al., 1991). Ganor and Foner (1996) showed that the chemical composition of dust transported from North Africa is similar to the dust transported from the east direction. Both dust sources contain soluble and insoluble inorganic material as well as organic matter, but they are dis-

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located about 700 m north of the sampling station. The  $PM_{2.5}$  data were taken from a station at the Municipal High School Ironi D Station ( $32^{\circ}5'34.9''$  N  $34^{\circ}47'27.5''$  E) located about 2.5 km south west of our sampling site. Aerosol number concentration and aerosol mass concentration ( $PM_{10}$  and  $PM_{2.5}$ ) are also listed in Table 1.

The immersion-freezing measurements were conducted using the FRIDGE-TAU (FRankfurt Ice-nuclei Deposition freezinG Experiment, the Tel Aviv University version) chamber (Fig. 2). This chamber which is usually used for measuring ice nucleation by deposition or by condensation freezing (Bundke et al., 2008; Klein et al., 2010b) was used here to determine the temperature at which freezing of drops containing aerosols took place.

Each filter containing the collected aerosols was placed in 10 mL of double distilled water (resistivity of  $18.2 \text{ megohm cm}^{-1}$ ). The aerosols were then removed from the filter by ultrasonic shaker. It is assumed that the 30 min shaking removed all the collected aerosols from the filter. The resulting mixture of water and aerosols was the source of the drops tested for immersion-freezing. Each test consisted of about 140 drops ( $1 \mu\text{L}$ ; 0.8 mm diameter) that were placed on the temperature controlled stage in the FRIDGE-TAU. A thin layer of Vaseline was first put on the stage in order to prevent ice from forming on the surface during cooling. This is because sometimes very thin ice dendrites on the substrate start to grow by vapor deposition from the perimeter of some of the frozen drops, reaching and freezing some of their neighbors, thus affecting the measurements. The temperature of the cooling stage was lowered at a constant rate of  $1^{\circ}\text{C min}^{-1}$  and the number of drops that froze at each temperature was recorded by a CCD camera.

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 duplicate the measurements, thus assuring consistent results. The exceptions are the filters from the 24 January 2009, 19 February 2009 and 17 December 2009 which were not cut and were immersed in the distilled water and in the ultrasonic shaker.

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### 3.1 Calculation of immersion-Freezing Nuclei (FN) concentration

In order to estimate the concentrations of immersion-Freezing Nuclei (FN) in the air we converted Vali's (1971) equation taking into account the amount of air that had been sampled in each measurement. The equation is composed of two parts: the first is an integration of the differential probability that a drop will freeze at temperatures between  $T$  and  $T - \Delta T$  due to the presence of a single active nucleus in it over the temperature range from  $0^\circ\text{C}$  to  $T$ . The result of the integration is the cumulative nucleus concentration  $K'(T)$ , which represents the number of nuclei active at all temperatures warmer than  $T$ . In order to obtain the actual concentrations of ice nuclei in the sampled air, consideration must be given to the total air sampled. This is presented in the last part of the equation:

$$K'(T) = \frac{1}{V} \times [\ln(N_0) - \ln(N(T))] \times \frac{x}{y} \quad (1)$$

$K'(T)$  – Cumulative concentration of FN in the air active at temperature  $T$  ( $\text{L}^{-1}$ )

$V$  – Volume of drop (L)

$N_0$  – Total number of drops measured

$N(T)$  – Number of unfrozen drops at temperature  $T$

$x$  – The volume of water used to remove the aerosols from the filter (L)

$y$  – The volume of air sampled through the filter (L)

Equation (1) was verified and found as a reliable equation based on laboratory measurement of Montmorillonite particles (SWy-2 Na-Montmorillonite) on one filter. Three experiments were made with the same filter in which we diluted the aerosols in the sample three successive times and analyzed them each time in immersion-freezing mode. The dilution was done by placing the filter in the test tube and removing the aerosols into 5 mL of doubly distilled water. Drops from this sample were analyzed in the freezing mode. Then another 5 mL of water were added (total of 10 mL) and drops from this



in the experiment. The temperature at which 50 % of the drops froze after the reduction shifted by an average of 0.18 °C.

The drops from the sampled filters began to freeze at lower temperatures than those reported from some biological aerosols (e.g. Schnell and Vali, 1976; Maki and Willoughby, 1978; Schnell et al., 1982; Levin and Yankofsky, 1983). The freezing spectra were found to be similar to those reported by DeMott et al. (2006) for Arctic aerosols but warmer than those reported by Ardon-Dryer et al. (2011) for the South Pole. Most onset freezing temperatures in this work were lower than those measured by Conen et al. (2012) in the High Alpine Research Station Jungfraujoch in the Swiss Alps. Furthermore, the immersion-freezing in this report was much more efficient (occurred at warmer temperatures) than the immersion-freezing due to soot particles (Fornea et al., 2009).

Hobbs and Locatelli (1969) measurements downwind of a natural forest fire indicate that the smoke from the fire contained large number of ice nuclei. 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, will be very effective. However, our measurements show that the freezing spectrum from this day was not as effective as those from other filters. In addition, immersion-freezing started at a temperature of -18 °C, with 50 % of the drops freezing at -21.7 °C. This freezing temperature was similar to the findings of Prenni et al. (2009b), from the Amazon basins, suggesting that particles from biomass-burning are not a likely source of effective ice nuclei. Therefore we can claim that while some wildfire can be a significant source of IN (e.g. Pratt et al., 2011; Prenni et al., 2012), not all sources of biomass burning can generate effective IN particles (Petters et al., 2009). The main difference could be that in our case most of the wood that was burning was very dry (mostly construction wood) while in some of other reports the measured smoke was from freshly cut wood or from forest fires.

The temperatures at which 50 % of the droplets froze in each case were found to be correlated with the corresponding daily average of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>10</sub>-PM<sub>2.5</sub> (see Fig. 5). The correlation value of PM<sub>2.5</sub> ( $R^2 = 0.42$ ) was lower than those measured for

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$PM_{10}$  and  $PM_{10}-PM_{2.5}$  ( $R^2 = 0.47$ ). This suggests that the higher concentration of the larger particles (particles in the range  $PM_{10}-PM_{2.5}$ ) results in higher concentrations of effective freezing nuclei. Furthermore, as the aerosol concentration increases, the temperature at which 50% of the droplets freeze was higher. The presence of more particles and also larger ones increases the surface area on which nucleation can occur, leading to more FN per drop (e.g. Garten and Head, 1964; Philips et al., 2008; Welti et al., 2009; Klein et al., 2010b; Niedermeier et al., 2011).

The FN concentration of all the samples from Israel was calculated based on Eq. (1). The FN concentration for the different filters between  $-13^\circ\text{C}$  to  $-27^\circ\text{C}$  varied from  $0.32\text{ L}^{-1}$  to  $211\text{ L}^{-1}$ , as can be seen in Fig. 6. As expected, the concentration of active nuclei increases as the temperature decreases. Calculating the best fit line (black line in Fig. 6) from the entire data one gets an exponential equation;

$$N_{\text{FN}} = 4 \times 10^{-4} e^{0.51\Delta T}, \quad (2)$$

where  $N_{\text{FN}}$  represents the concentrations of FN ( $\text{L}^{-1}$ ) and  $\Delta T$  represents the supercooling in  $^\circ\text{C}$ . This equation gives a concentration of  $1\text{ L}^{-1}$  at  $-15.5^\circ\text{C}$ , which is higher than the average temperature obtained by Bigg and Stevenson (1970) from measurements around the world ( $\sim 1\text{ L}^{-1}$  at  $-20^\circ\text{C}$ ).

Activation fraction (AF) values of all filters were also calculated based on the total aerosol concentration (Nt) in the size range of  $0.11-3\mu\text{m}$ , which were measured simultaneously with the filters. The average AF value of all the filters was  $4.4 \times 10^{-5} \pm 7.5 \times 10^{-5}$  (average  $\pm$  standard deviation value) with variations from  $1.5 \times 10^{-7}$  up to  $4.9 \times 10^{-4}$ , as can be seen in Fig. 7. All the activation fraction values were combined and a best fit line was calculated (see black line in Fig. 7). The majority of AF values were in the ranges of activation fraction ( $10^{-3} - 10^{-6}$ , for particle  $> 0.1\mu\text{m}$ ) proposed by Pruppacher and Klett (1997), however the lowest AF value was much lower than in Pruppacher and Klett (1997).

The concentration of FN and the values of AF increased as the temperatures decreased; the relatively high correlation values ( $> 0.6$ ) imply a strong dependence of FN





agrees with the work of Levin and Lindberg (1979) who concluded that in this region the atmosphere always contains some dust particles even in the absence of dust storms.

In Fig. 11, DeMott et al. (2010) compiled data from measurements of ice nuclei concentrations as a function of temperature from around the world and added the prediction based on theoretical and empirical equations of Fletcher (1962), Cooper (1986) and Meyers et al. (1992). We added to this figure the FN concentrations as measured in the South Pole (Ardon-Dryer et al., 2011) and also the data and the best fit lines for dusty and clean conditions from the present work. The figure shows that the slope of the lines of the present data is similar to that measured at the South Pole and to the theoretical prediction of Cooper and Fletcher. Although at the high temperature the concentrations measured in the present work fall in the same region as those measured by others, our ice nuclei concentrations are higher than the data collected elsewhere. It is interesting to note that the parameterization by Meyers et al. (1992) predicts much higher ice nuclei concentrations at high temperatures. Based on Fig. 11, we can conclude that the FN concentrations in our area are higher than in other regions, probably due to the ever presence of mineral dust particles in the air.

## 5 Conclusions

Immersion Freezing Nuclei were measured from samples collected on different days in Israel during 2009–2010. FN was found to be effective from  $-12^{\circ}\text{C}$  down to  $-29^{\circ}\text{C}$ , the average temperature at which 50 % of the drops froze occurred at  $-21^{\circ}\text{C}$ . Concentration of  $1\text{ L}^{-1}$  was observed at  $-15.5^{\circ}\text{C}$ , with a range of concentrations from  $0.32\text{ L}^{-1}$  to  $211\text{ L}^{-1}$ .

The temperatures at which 50 % of the droplets froze in each case were found to be correlated with the corresponding daily average of  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_{10}-\text{PM}_{2.5}$ . The fact that the correlation value between FN concentrations and  $\text{PM}_{10}-\text{PM}_{2.5}$  was higher than  $\text{PM}_{2.5}$  suggests that the larger particles are generally more effective as FN and is

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in agreement with the notion that the activity of ice nucleation is correlated with surface area.

The measurements were divided into dust and clean conditions based on their back trajectory maps and aerosol mass concentrations ( $PM_{10}$ ). Droplets from the dusty days froze at warmer temperatures than the clean days. The dusty days had higher FN concentrations and AF values than clean days. These differences ( $> 2.5$  times) were smaller than what was found in other places (such as Germany and the US). This agrees with previous reports showing that the eastern Mediterranean atmosphere always contains some dust particles and therefore their effect on IN concentrations is to generally raise the background level.

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**Table 1.** List of filters that were sampled.

# Filter	Date	CPC <sub>3010</sub> number concentration (# cm <sup>-3</sup> )	PM <sub>10</sub> day average (µg m <sup>-3</sup> )	PM <sub>2.5</sub> day average (µg m <sup>-3</sup> )
1	24 Jan 2009	639.7	841.7	212.3
2	15 Feb 2009	726.8	245.4	44.4
3	19 Feb 2009	1251.2	441.9	100.1
4	8 Mar 2009	515.6	317.8	76.5
5	15 Mar 2009	1964.3	39.0	12.6
6	16 Jun 2009	1158.6	35.4	19.7
7	21 Sep 2009	841.0	29.9	22.3
8	22 Oct 2009	422.9	31.3	21.2
9	1 Nov 2009	1865.4	84.4	21.4
10	17 Dec 2009	1825.7	677.6	153.8
11	30 Jan 2010	718.4	383.4	59.7
12	9 Mar 2010	1600.2	408.1	83.5
13	11 Apr 2010	436.7	351.6	72.5
14	1 May 2010_15*	563.9	36.4	21.2
15	1 May 2010_23*	1344.8	36.4	21.2
16	27 May 2010	751.9	867.2	154.6
17	15 Nov 2010	2626.3	93.5	44.6
18	30 Dec 2010	1552.1	88.7	29.7
19	31 Dec 2010	612.2	49.2	21.0

\* Two samples were collected on the same day (1 May 2010) at 15:00 and at 23:00 LT (during Lag Ba Omer celebration).

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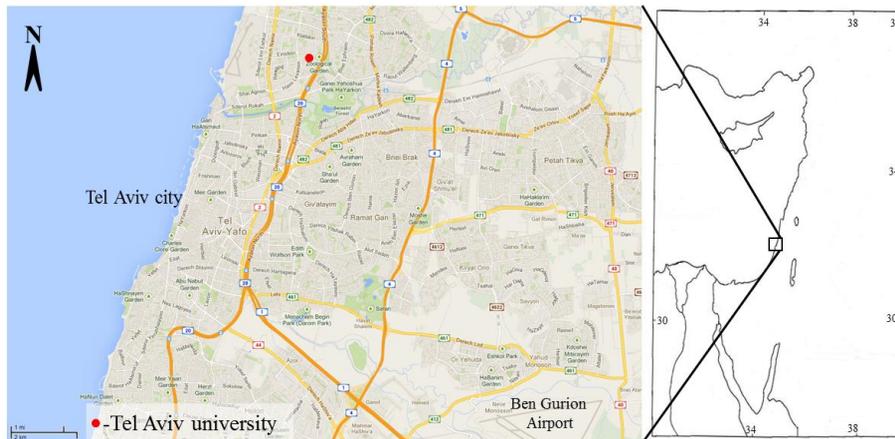
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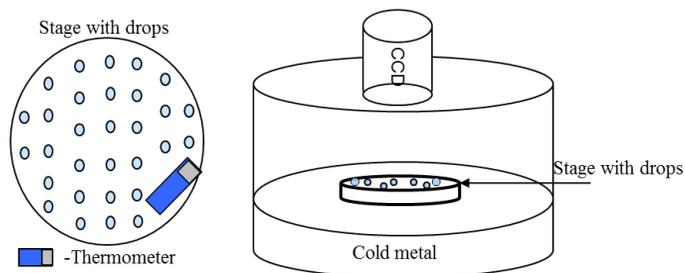
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**Fig. 1.** Location of the sampling station at Tel Aviv University marked in red (Google map, with modification, 2012).

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**Fig. 2.** Schematic diagram of the immersion-freezing method, in FRIDGE-TAU.

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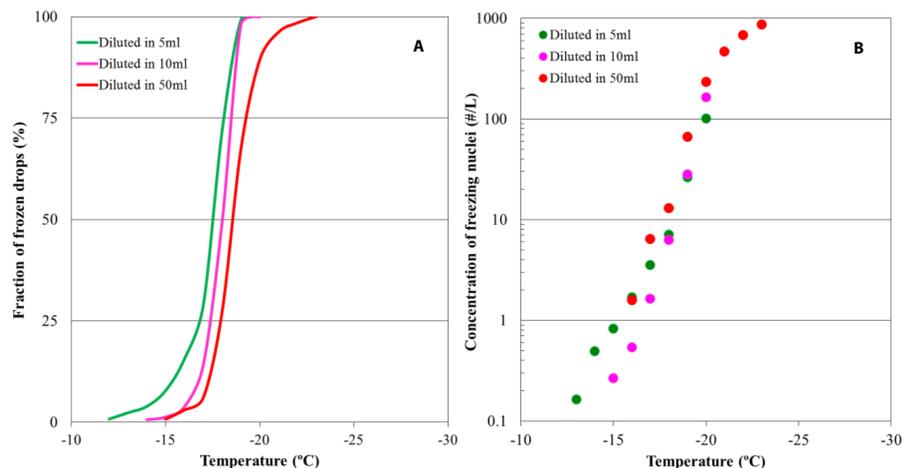
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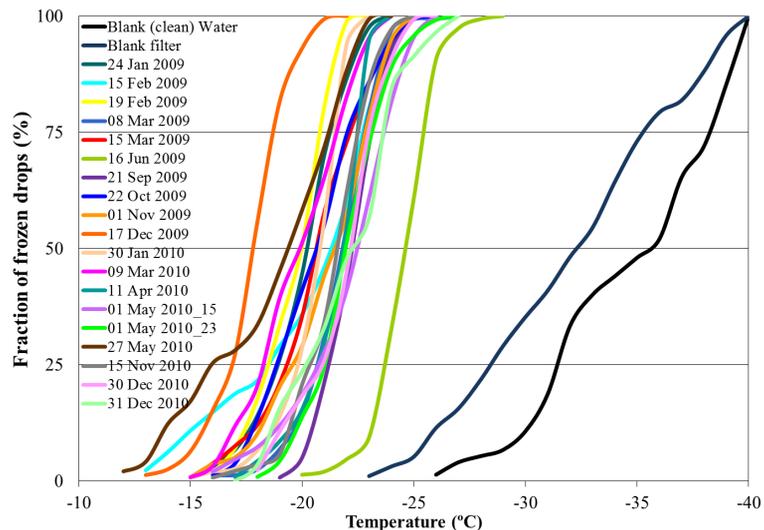


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**Fig. 3.** Cumulative freezing spectrum of all three experiments of Montmorillonite particles with different dilution (A) and in Concentration of Freezing Nuclei (FN) calculated for the different experiments (B), each color represents different dilution experiment.

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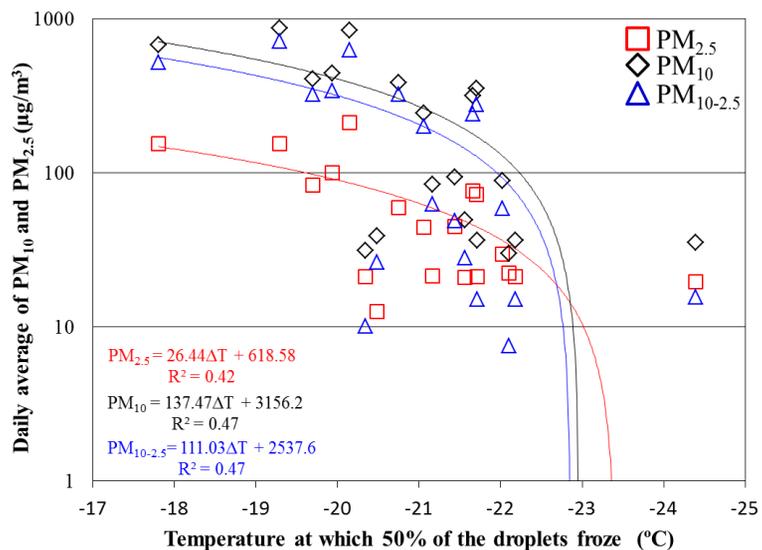
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**Fig. 4.** The original cumulative freezing spectrum of all measured filters and the average spectrum of freezing temperature from clean (blank) filters and blank (pure) water.

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**Fig. 5.** The connection between the temperatures at which 50% of the droplets froze to the daily average values of  $PM_{2.5}$  (in red),  $PM_{10}$  (in black), and  $PM_{10}-PM_{2.5}$  (in blue).

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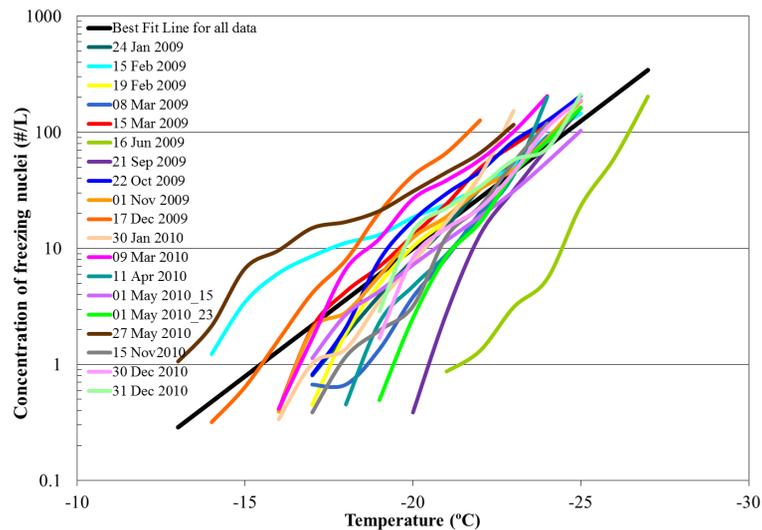
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**Fig. 6.** Concentration of immersion-Freezing Nuclei (FN) for the different filters, calculated from Eq. (1). The black line is the best-fit line representing all the data.

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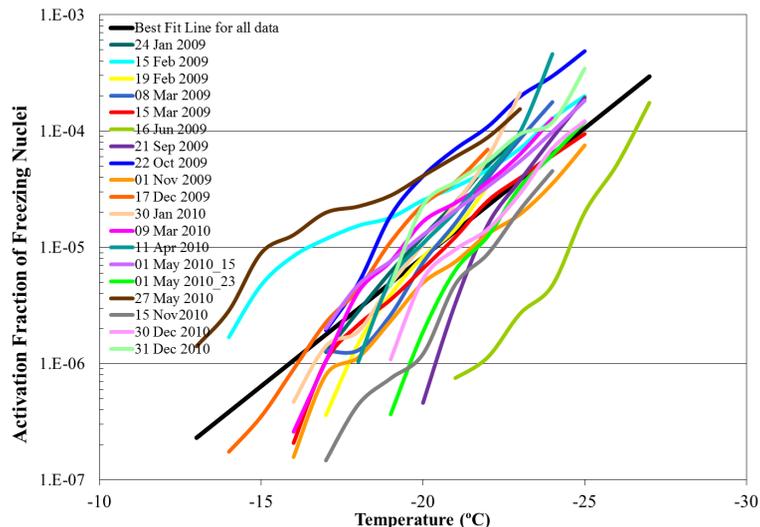
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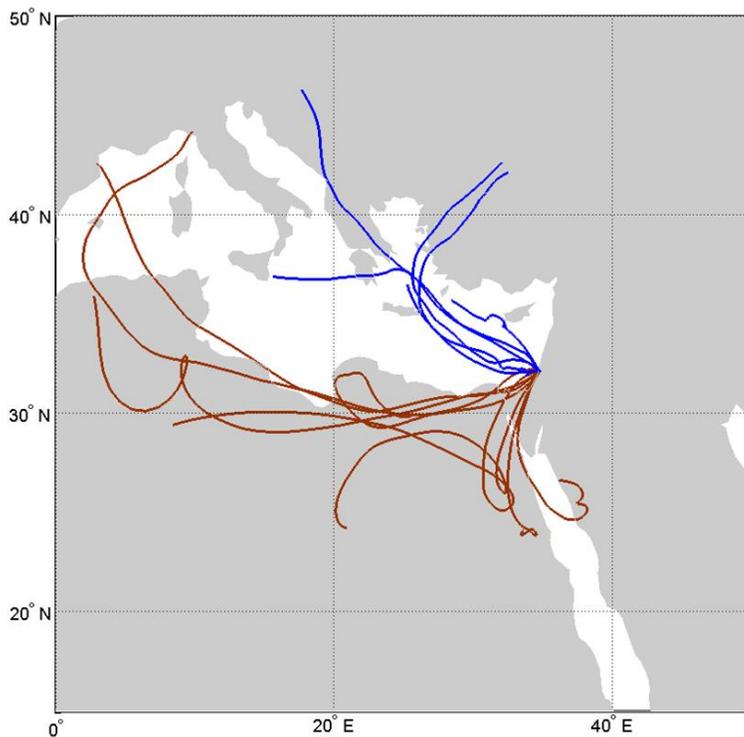
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**Fig. 7.** Activation fraction values of FN calculated for the different filters, each color represents different filter while the black line is the best-fit line.



**Fig. 8.** Back trajectory history of air masses showing passage of air prior to reaching the sampling station. Each line represents a 72 h trajectory at 500 m altitude. The back trajectories were taken from <http://www.arl.noaa.gov> website. The brown lines represent dust conditions and the blue lines represent clean conditions.

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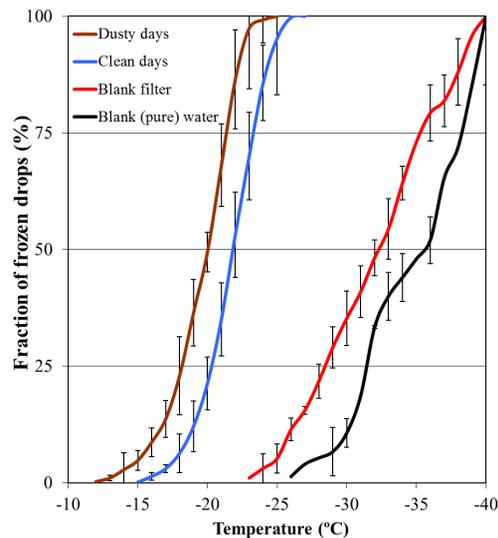
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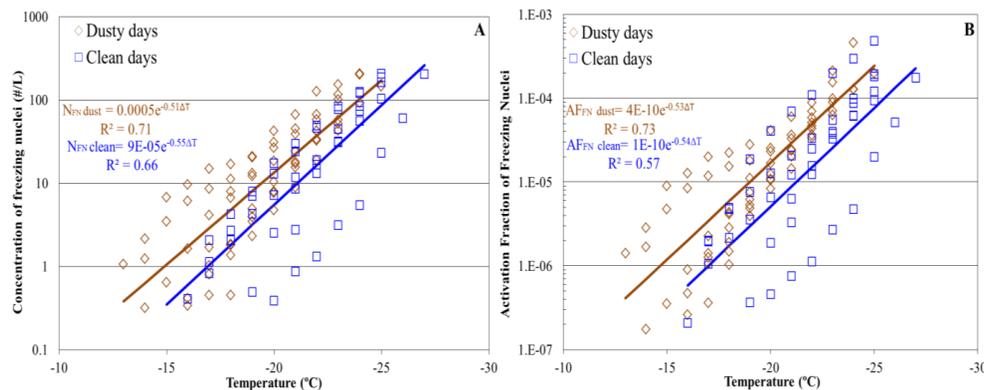


**Fig. 9.** The cumulative freezing spectrum of average clean and dusty samples and the average spectrum of freezing temperature for blank filters and blank (pure) water with standard error values for each temperature.

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**Fig. 10.** Freezing Nuclei Concentration **(A)** and Activation Fraction values **(B)** calculated for clean (blue) and dusty (brown) days. Best-fit lines and the equations that represent them are also shown.

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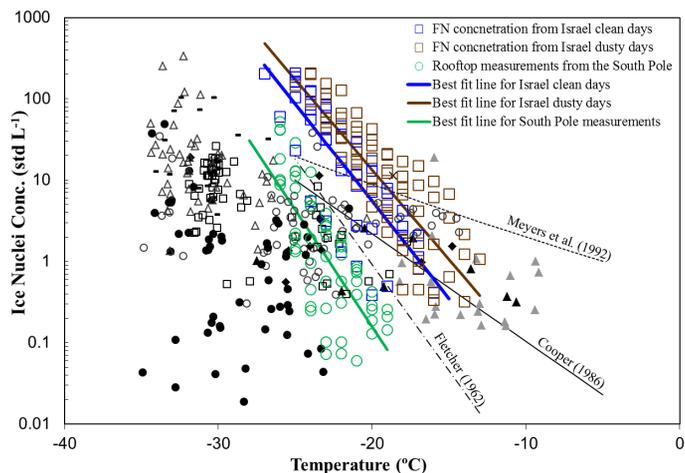
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**Fig. 11.** Ice nuclei concentrations as a function of temperature from DeMott et al. (2010) with FN calculated from filters measurements from Israel during dusty days (brown square) and clean days (blue square) and the measurements of Ardon-Dryer et al. (2011) from the South Pole (green circle) with their best fit lines. The parameterization curves of Fletcher (1962), Cooper (1986) and Meyers et al. (1992) are also shown.