

Characteristics of Immersion Freezing Nuclei at the South Pole Station in Antarctica

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Abstract

The effectiveness of aerosols as immersion freezing nuclei at the South Pole station was investigated during January and February 2009 using the FRIDGE-TAU. The analysis consisted of testing the freezing temperature of about 100-130 drops per sample containing aerosols collected at ground level and on a balloon lifted to different heights. All the drops froze between -18°C and -27°C . The temperature in which 50% of the drops froze occurred at -24°C , while nuclei concentration of 1 L^{-1} at -22°C was calculated. Meteorological conditions such as wind speed, ice precipitation as well as the trajectories of the air masses affected the ice nuclei concentrations. Higher concentrations were observed on days when the winds were stronger or when the air mass originated from the sea.

1. Introduction

Much attention has been given to condensation nuclei (CN) characteristics in the Antarctic continent (Saxena, 1983; Gras et al., 1985; DeFelice, 1996; DeFelice et al., 1997), yet the characteristics of ice-forming nuclei, their origin, composition and concentrations have only seldom been studied (Bird et al., 1961; Bigg and Hopwood, 1963; Kumai, 1976; Saxena and Weintraub, 1988; Junge and Swanson, 2008). Most of the above studies were conducted along the coast of Antarctica and only a very few reported on measurements at the South Pole (e.g. Kumai, 1976).

Different types of instruments and methods have been used to measure ice nuclei (IN) in Antarctica. Some measurements were conducted by sampling aerosols on filters and analyzing their ice nucleating properties by exposing them in the laboratory to

water saturation conditions at different sub-zero temperatures (Bigg, 1973). Others brought outside air into a mixing cold chamber and counted the number of ice crystals that fell into a sucrose solution (Bigg and Hopwood, 1963; Bigg, 1990). Some reported on ice nuclei concentrations by counting the number of supercooled drops that froze in a free-falling freezing tube (Junge and Swanson, 2008). Saxena and Weintraub (1988) tested the effectiveness of aerosols as ice nuclei by using the drop freezing technique. All the above mentioned methods analyzed the effectiveness of the aerosols as immersion freezing or as condensation-freezing nuclei.

Bigg and Hopwood (1963) reported on ice nuclei concentrations at McMurdo station located near the coast of Antarctica. They found that freezing occurred between -14° to -26°C , with concentrations of $0.5 - 13 \text{ L}^{-1}$ of active nuclei at -20°C . Most of the nuclei had a diameter of around $0.01\mu\text{m}$, while some had a diameter of $0.1\mu\text{m}$. Bigg (1973) measured ice nuclei concentrations at -10 , -15 and -20°C with mean concentrations of $1-5 \times 10^{-3}$, $10-20 \times 10^{-3}$, $0.1 \text{ (L}^{-1}\text{)}$ respectively. Saxena and Weintraub (1988) used drop freezing measurements at Palmer station over the temperature range of -5°C to -17°C . They reported on the presence of high concentrations of ice nuclei (0.01 to 10 L^{-1}) even at temperatures as high as -5 to -7°C . They also reported finding good correlation between ice nuclei concentrations and the presence of Potassium, Silicon and Zinc.

Carpenter et al (2000) and Warren and Hudson (2003) reported on the existence of bacteria in the South Polar snow. However, their effectiveness as IN is still unclear. Junge and Swanson (2008) suggested that immersion freezing of marine psychro-active bacteria and viruses would not be important for heterogeneous ice nucleation processes in polar clouds nor for the formation of sea ice, because they would nucleate ice at temperatures very close to the homogeneous nucleation temperature ($-42.2 \pm 0.3^{\circ}\text{C}$).

The relative contribution of mineral dust aerosols, which are known to be good ice nuclei, to the aerosol population in Antarctica, is still unclear. Bird et al (1961) claims that dust of land origin cannot generally contribute to nucleus concentration in the southern hemisphere. Iriondo (2000) suggested that mineral dust particles can be transported from Patagonia (South America) to western Antarctica, which seems to

agree with Kumai (1976) who reported that 85% of the snow crystals had nuclei in them which were mainly clay minerals and sodium chloride particles. The clay mineral nuclei consisted of 20% Illite, 8% Kaolin, 4% Halloysite, 3% Vermiculite, and 24% other related minerals. Similar concentration were found in Patagonia deserts (Johnson et al., 2010)

The objective of this paper is to report on ice nuclei measurements in a remote pristine region of Antarctica such as the South Pole station.

2. The characteristics of the research area

The measurements were conducted during January and February 2009 at the Amundsen-Scott station at the geographical South Pole (Latitude 89° 59.77' S, Longitude 92° 1' E, Fig. 1) on a polar plateau at an altitude of 2850 m (for more information about the campaign see Lawson et al., 2010). The weather is uniformly cold and dry (Hogan, 1979). Its average barometric pressure is about 680mb (Hogan and Barnard, 1978). The surface circulation on the polar plateau is dominated by a weak gradient wind, with slight katabatic reinforcement (Hogan et al., 1982). Surface winds generally swing from north to northwesterly (Bigg, 1980). Frontal type storms are very rare in the South Pole plateau (Hogan, 1975a). The summer time is characterized by clear skies with occasional occurrence of ice saturated layers just above the surface (Egan and Hogan, 1986). Most of the clouds are cirrus and altostratus; liquid precipitation is unknown but ice crystal precipitation is relatively common (Hogan, 1975b).

The South Pole is characterized mostly by very small aerosols (Bigg, 1980; Shaw, 1980; Park et al., 2004)). The submicron aerosol particles appear to be mainly sulfates (Deshpande and Kamra, 2004) in the form of sulfuric acid, ammonium bisulphate or ammonium sulfate (Bigg, 1980; Bigg et al., 1984). Bodhaine et al (1988) found black carbon at the South Pole, which they attribute to local pollution. Even sea salt particles have been found at the South Pole (Parungo et al., 1981; Hogan et al., 1984), although the station is located 1250 km away from the ocean (Kumai, 1976). Some of the common elements found in the Antarctic aerosol are iron, potassium, silicon,

calcium, aluminum, sulfur and titanium (Cadle et al., 1968; Parungo et al., 1981; Saxena and Weintraub, 1988).

According to Hogan et al (1982) a few times a year, large storm systems intrude far enough south to cause moist flow up the glacial slopes to the South Polar Plateau. This flow brings relatively warm moist aerosol laden air to the South Pole. As this air cools toward inland, ice crystals form, precipitate to the surface removing water and scavenge aerosols from the lowest layers.

3. Method of analysis

Twelve filter samples were collected; nine from the rooftop and three from a balloon (see Table 1) the inlet of the filter holder was positioned with the wind to its back. The aerosols were sampled on Nitrocellulose filters with diameter of 47 mm and 0.45 μm pore size. The effectiveness of the sampled aerosols as immersion freezing was measured using a drop freezing technique.

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., 2010) 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 megohm·cm). The aerosols were then removed from the filter by ultrasonic shaker. The resulting mixture of water and aerosols was the source of the drops tested for immersion freezing. Each test consisted of about 120 drops (2 μl ; 1.6 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, thus affecting the measurements).

The temperature of the cooling stage was lowered at a constant rate of 1°min^{-1} and the number of drops that froze at each temperature were recorded by a CCD camera.

In some cases (e.g. filter 4, 8-12), the filters were cut in half, due to the large volume of air that had been sampled through them. Some of the sections that had not been used to analyze ice nuclei were saved for elemental analysis of individual aerosols in an Environmental Scanning Electron Microscope, ESEM (filters: 4, 9 and 10).

4. Results and discussion

Twelve samples containing a total of 1465 drops were analyzed. The analysis was carried out between 0 to -27°C within which all the drops froze (Fig. 3). Most drops in most samples began to freeze only at -18°C. The freezing occurred at warmer temperatures as compared to water drops taken from pure water. Drop freezing experiments using water with blank filters were also conducted. They revealed that although 50% of the drops from the blank samples froze at lower temperatures (< -26.5⁰), there was some overlap with the cumulative spectra of a few of the filters. Fig. 3 shows that about < 24% of the warmest freezing events in the blank sample were similar to those from filters 7-12. This indicates that even the strict procedure of maintaining cleanliness could not rule out some contamination on the filters. In the subsequent analysis, therefore, the fraction of drops in the samples that froze at the same temperature as the blank have been removed. It is interesting to note that the elimination of the drops that froze at overlapping temperatures with the blank filters only modified our results (see Eq. 2 below) by 2.4%.

From Fig. 3 it is apparent that although freezing of drops started at -18°C most froze at temperatures lower than -20°C. 50% of the drops in all the samples froze between -23 to -24.5°C. The average temperature in which 50% of the drops froze occurred at -24°C, much warmer than reported by Junge and Swanson (2008) where near homogeneous freezing temperatures were observed for specific bacterial isolates.

In order to estimate the concentrations of immersion freezing nuclei (FN) in the air we converted Vali's (1971) equation, in order to take into account the amount of air that had been sampled in each measurement.

$$K'(\theta) = \frac{1}{V} \times [\ln(N_0) - \ln(N(\theta))] \times \frac{x}{y} \quad (1)$$

$K'(\theta)$ – Cumulative concentration of FN in the air active at temperature θ (L⁻¹)

V – Volume of drop (L)

N_o – Total number of drops measured

$N(\theta)$ – Number of unfrozen drops at temperature θ

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

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

The concentration of freezing nuclei in the air calculated for the different samples between -19 to -27°C varied from 0.1 L⁻¹ to 107 L⁻¹, respectively, as can be seen in Fig. 4. As expected, the number of active nuclei increases as the temperature decreases. A concentration of 1 L⁻¹ was observed in all the filters between -21°C to -25°C.

4.1 Measurements of freezing nuclei from the balloon

While most of the samples were collected from the rooftop (filters 1-4; 8-12), three samples were collected from a balloon. One sample (filter 5) was collected as the balloon ascended to 196 m and then descended to the surface. The second and third samples (filter 6 and 7) were collected when the balloon remained stationary at 20 m and 40 m above ground, respectively. Fig. 5 shows that the concentrations of ice nuclei collected at 20 m were higher than those measured during ascend and descend of the balloon. The lower FN concentrations at the higher altitude (filter 5) suggest that most of the effective FN originate from the surface or are transported from long distances at the boundary layer.

Since most of the measurements were carried out from the rooftop and since the difference between filters 5 and 6 is small, most of the rest of the analysis will deal only with the rooftop measurements.

4.2 Measurements of freezing nuclei from the rooftop

Fig. 6 represents the average concentrations of FN at a number of temperatures on each day of the campaign based on the measurements of the filters shown in Table 2. As can be seen, at the beginning of the campaign (28-29/01/09), larger concentrations of FN were found at all temperatures between ~ -20 °C to -27°C. With time, the concentration of FN decreased by about one order of magnitude at all temperatures. In order to understand these changes in FN concentration we decided to investigate the

possible connection with the meteorological conditions (local and historical) that existed at the time of the campaign.

Based on the meteorological data collected at the measuring site it appears that the aerosols samples were collected under different meteorological conditions. One local meteorological condition that could have had an effect on the FN concentration is wind speed. Each filter sample was collected under different wind speeds, as can be seen in Table 3. High correlation between wind speed and FN was found for all measured temperatures (Fig. 7a). Similarly, the correlation of wind speed with total aerosol concentrations was high also (Fig. 7b). It should be noted that the aerosol concentrations shown in Table 3 refer to total aerosols greater than $0.01 \mu\text{m}$ (measured by the TSI 3760). Most of these aerosols are not so relevant for the FN measurements because they are much smaller than the expected size of the ice nuclei (e.g. DeMott et al, 2010).

Our filter samples could be divided into two groups based on wind speed. Filters exposed to winds greater than 6 m s^{-1} and those exposed to lower wind speeds. For example, filters 1-4 were exposed to higher wind speed (average of 9 m s^{-1}) and had 16 L^{-1} at -25°C . Filters 8-12, which were exposed to lower average wind speeds of about 4.4 m s^{-1} had only 2 L^{-1} at -25°C . One explanation for this observation was suggested by Hogan (1979) who showed that strong winds in the South Pole increased the mixing of air near the surface, leading to higher concentration of larger particles at higher elevations, implying higher concentrations of FN. During the sampling time of filter 9-11, the station was affected by falling ice crystals and broken clouds that occurred from 30.01.09 to 01.02.09 (see decrease in FN concentration in Fig. 6). These falling ice crystals undoubtedly scavenged aerosols in the lower atmosphere, thus reducing the measured FN concentrations. The possibility that sublimated ice crystals in the air affected the ice nuclei measurements was ruled out because the relative humidity was far above ice saturation. The only possible effects could be the collection of some of the ice crystals on the filters, which sublimated later, leaving the ice nuclei on the filters. However, the chance for this to happen was considered low because of the orientation of the inlet.

In addition to the effects of the local meteorological conditions, the history of the air masses also had an effect on the effectiveness of the FN. Fig. 8 shows an example of two different air mass back trajectories calculated using the HYSPLIT method - Hybrid Single Particle Lagrangian Integrated Trajectory Model (<http://www.arl.noaa.gov>). Filters 1-8 had similar trajectories that originated from the ocean. These air masses named maritime because according to their back trajectories, they originated over the Ocean about 60 hours before reaching the South Pole station (e.g. note the back trajectory of the air sampled on filter 4 in Fig.8). In contrast, the air sampled on filters 9-12 had trajectories that spent at least 72 hours over the continent, much longer than the air that was sampled on filters 1-8. Therefore in this paper we refer to them as continental (e.g. note the back trajectory of the air sampled on filter 10 in Fig 8).

The differences in the history of the air mass, based on their back trajectory must have had some effect on the FN concentration and activity. As can be seen in Fig. 9, higher FN concentrations were observed for all the rooftop filters that sampled air that had originated from the ocean (1-4 and 8) as compared to samples of air that spent much longer time over the continent (filters 9-12). The higher concentrations of FN in maritime air could be caused by anthropogenic pollution emitted from ships and cruises that frequent the coast during the summer (Graf et al., 2009). We cannot rule out the role of bio-aerosols emitted from the coastal water as an important source of FN. It is also clear that the events with high concentrations of FN (filters 1-4, see also Table 3) were mostly associated with strong winds.

Although continental air contained lower concentrations of FN than the maritime one, the difference between the elemental compositions of the aerosol particles in these two air masses as measured with ESEM-EDX, was relatively small, with Al, Si, Fe, Ca, more abundance in the former and Cl and Na more abundance in the latter (Table 4). The possible explanation for these relatively small differences in elemental composition is the fact that the station is located over 1000 km away from the coast. Thus air mass that originates from the ocean spent a long time over the land, mixing with continental type aerosols. Furthermore, the elemental composition listed in Table 4 is similar to that found in the Patagonia desert (Gaiero et al., 2007), and is in

agreement with the measurements of Parungo et al (1981), suggesting the transport of mineral dust from this region in South America.

Although we found differences in FN concentrations for different air masses and meteorological conditions we think that it is valuable to obtain an average parametric equation for FN concentrations in such a remote site, something that could be helpful in numerical models. For this purpose all the rooftop measurements were combined and a best fit line was calculated (Fig. 10). The resulting best-fit line is:

$$N_{FN} = 5 \times 10^{-7} e^{0.66\Delta T} \quad (2)$$

N_{FN} - the number concentrations of FN (L^{-1})

ΔT - supercooling

From the figure one can see that ice nucleus concentration of $\sim 1 L^{-1}$ is found at $-22^{\circ}C$, which is a little lower than what Bigg and Stevenson (1970) reported as average concentrations at higher latitudes ($\sim 1 L^{-1}$ at $-20^{\circ}C$).

Comparing the FN concentrations obtained from Eq. 2 with previous measurements show that at $-15^{\circ}C$ the calculated concentrations are somewhat lower than what Bigg (1973) reported from his coastal measurements ($\sim 0.015 L^{-1}$). On the other hand, at Palmer Station, Saxena and Weintraub (1988) found ice nuclei concentrations of $\sim 1 L^{-1}$ at $-18^{\circ}C$, which are higher than those reported here and by Bigg (1973). There could be a number of reasons for this difference, the most reasonable one is that in the past 37 years the anthropogenic activity in and around Antarctica has increased dramatically, leading to more pollution from ships and other activities (Graf et al., 2009).

Overall the Ice nuclei concentrations that were found in the South Pole station are in agreement with other IN concentrations that were measured in other locations around the globe, as can be seen in Fig.11. However, the temperature dependence in these data is much stronger than generally found in the data from DeMott et al (2010). There may be many reasons for such differences that could only be revealed by further research. For example, data were not obtained to determine any association of IN with the presence of larger aerosols, as was inferred to be partly responsible for IN variability at one temperature by DeMott et al (2010).

5. Conclusions

Immersion Freezing Nuclei were measured from samples collected at the South Pole station during late January early February, 2009. FN was found to be effective from -18°C down to -27°C , with concentrations ranging from 0.1 to 107 L^{-1} . The temperature at which 50% of the drops froze occurred at -24°C . Concentration of 1 L^{-1} was observed at -22°C .

The meteorological conditions, such as wind speed or precipitation in addition to the trajectory of the air mass appear to have affected the Freezing Nuclei concentrations at the South Pole station. Higher concentrations were found in cases when the air originated from the ocean (maritime). The strongest correlation of FN concentration was found with wind speed. FN concentrations were almost eight to nine times higher when the wind increased from 4.4 to 9 m s^{-1}

Using the data, a parametric equation was calculated for the mean concentrations of FN as a function of temperature for a remote location such as the South Pole station. Due to the fact that measurements were conducted under different meteorological conditions, application of the computed parametric equation requires more measurements under different meteorological conditions.

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Table 1: Description of sampled filters

Filter #	Time of measurement (UTC)	Volume of air sampled (L)	Filter Location
1	28.01.09	344	Rooftop
2	28.01.09	327	Rooftop
3	29.01.09	319	Rooftop
4	29.01.09	450	Rooftop
5	29.01.09	327	Balloon
6	29.01.09	245	Balloon
7	29.01.09	245	Balloon
8	30.01.09	499	Rooftop
9	30.01.09	1317	Rooftop
10	31.01.09	1579	Rooftop
11	01.02.09	1873	Rooftop
12	02.02.09	2495	Rooftop

Table 2: The filters used for calculating the average FN concentrations based on the day of measurement

UTC time	28.01.09	29.01.09	30.01.09	31.01.09	01.02.09	02.02.09
Filter #	1, 2	3, 4	8, 9	10	11	12

Table 3: Average surface meteorological conditions during sampling.

filter number	Temperature (deg C)	Ambient pressure (hPa)	Wind speed (m/s)	Wind direction (deg)	RH in the air (%)	Aerosol number concentration >0.01 μ m (cm ⁻³)
1	-25.0	680.2	9.7	5.5	98.507	1864.3
2	-25.2	680.0	10.3	5.0	98.877	1996.3
3	-26.5	680.0	8.6	10.4	97.879	1978.0
4	-27.0	679.9	7.5	16.2	98.245	1748.5
5	-27.2	679.5	6.5	21.0	98.268	1464.7
6	-26.6	679.0	8.0	23.3	97.932	1757.7
7	-26.8	678.8	6.4	28.1	96.258	1704.2
8	-31.3	680.4	4.7	353.4	97.139	335.4
9	-32.6	683.4	5.4	76.0	98.045	545.3
10	-33.1	682.9	4.1	92.5	97.353	520.6
11	-27.5	685.0	5.6	59.3	97.367	1152.2
12	-30.6	689.1	2.4	298.9	82.720	1308.5

Table 4: Frequency of occurrence (%) of elements in individual particles in maritime and continental air masses.

Elements	Continental	Maritime
Al	44.4	37.4
Ca	35.3	27.5
Si	32.0	22.0
Fe	23.5	18.7
K	22.2	12.1
Cu	21.6	11.0
Mg	18.3	16.5
F	15.7	8.8
S	14.4	14.3
Cl	11.1	24.2
Na	8.5	22.0
Ti	3.3	1.1
Zn	2.0	9.9
Cr	2.0	8.8
Au		4.4
Ni		2.2
Sn		2.2
Mo		1.1
Cd	3.3	
P	0.7	
Br	0.7	
Mn	0.7	
number of particle	153	91

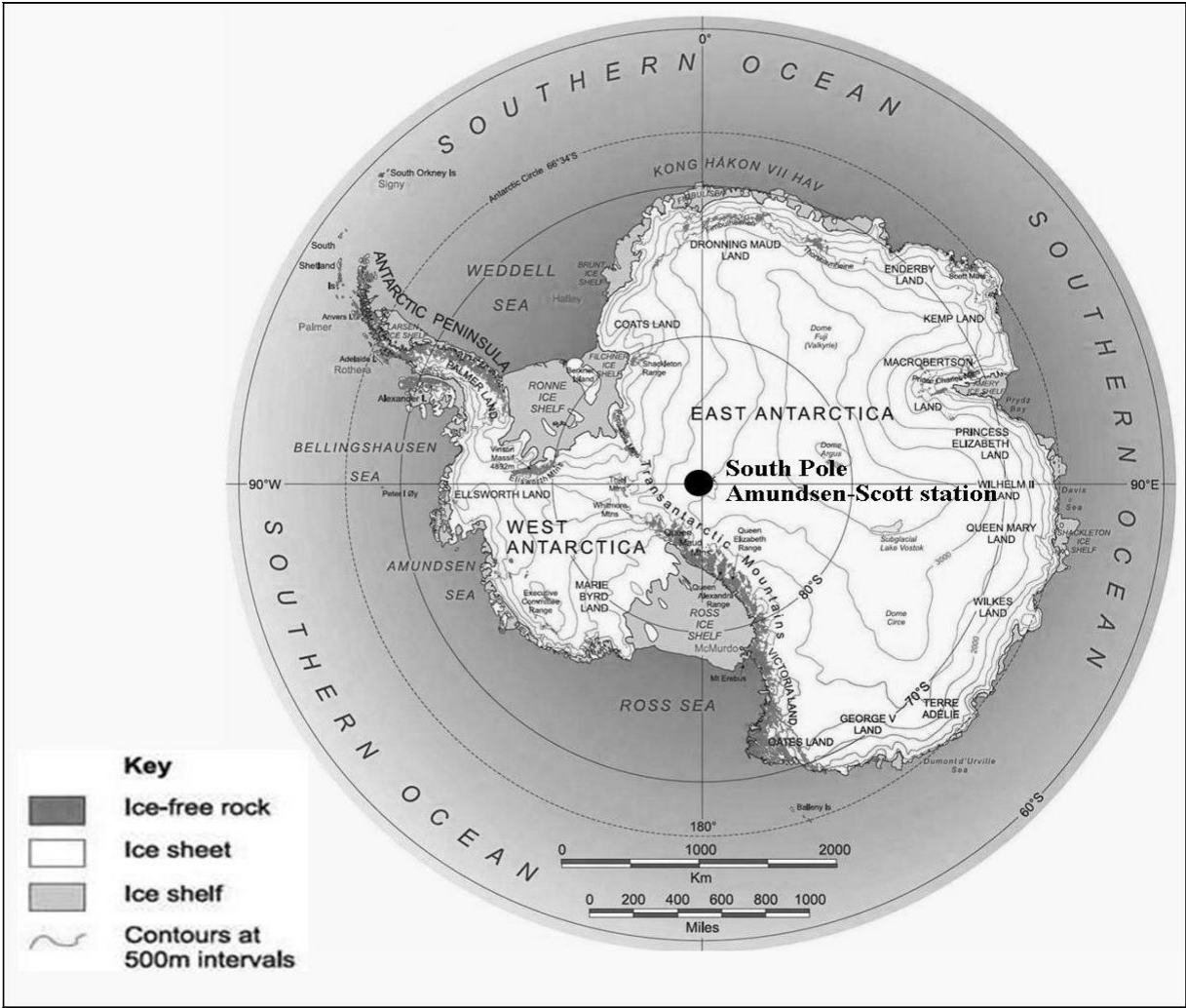


Fig. 1: location of the Amundsen-Scott station at the South Pole, in Antarctica, marked in black circle (Geology, 2010 with modification).

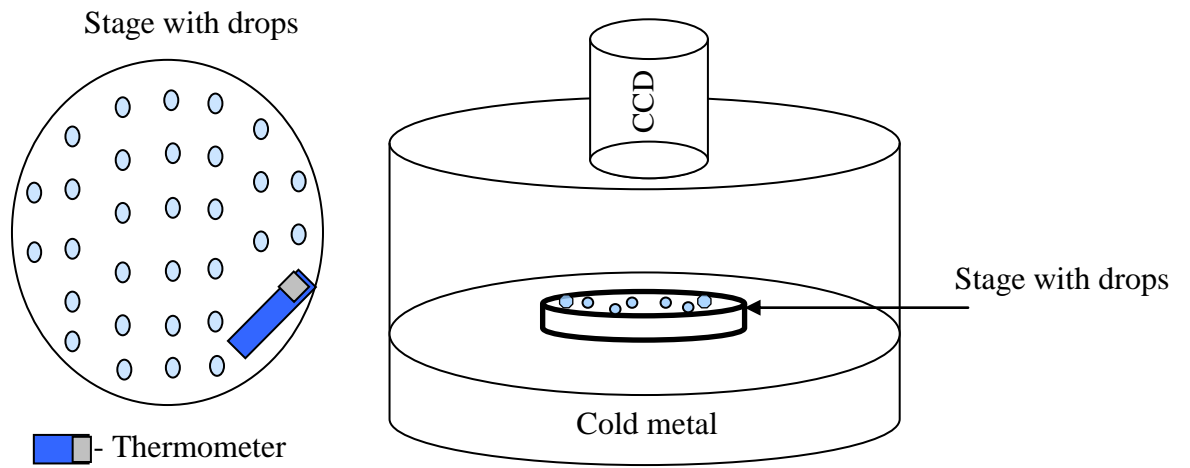


Fig.2: Schematic diagram of the immersion freezing method.

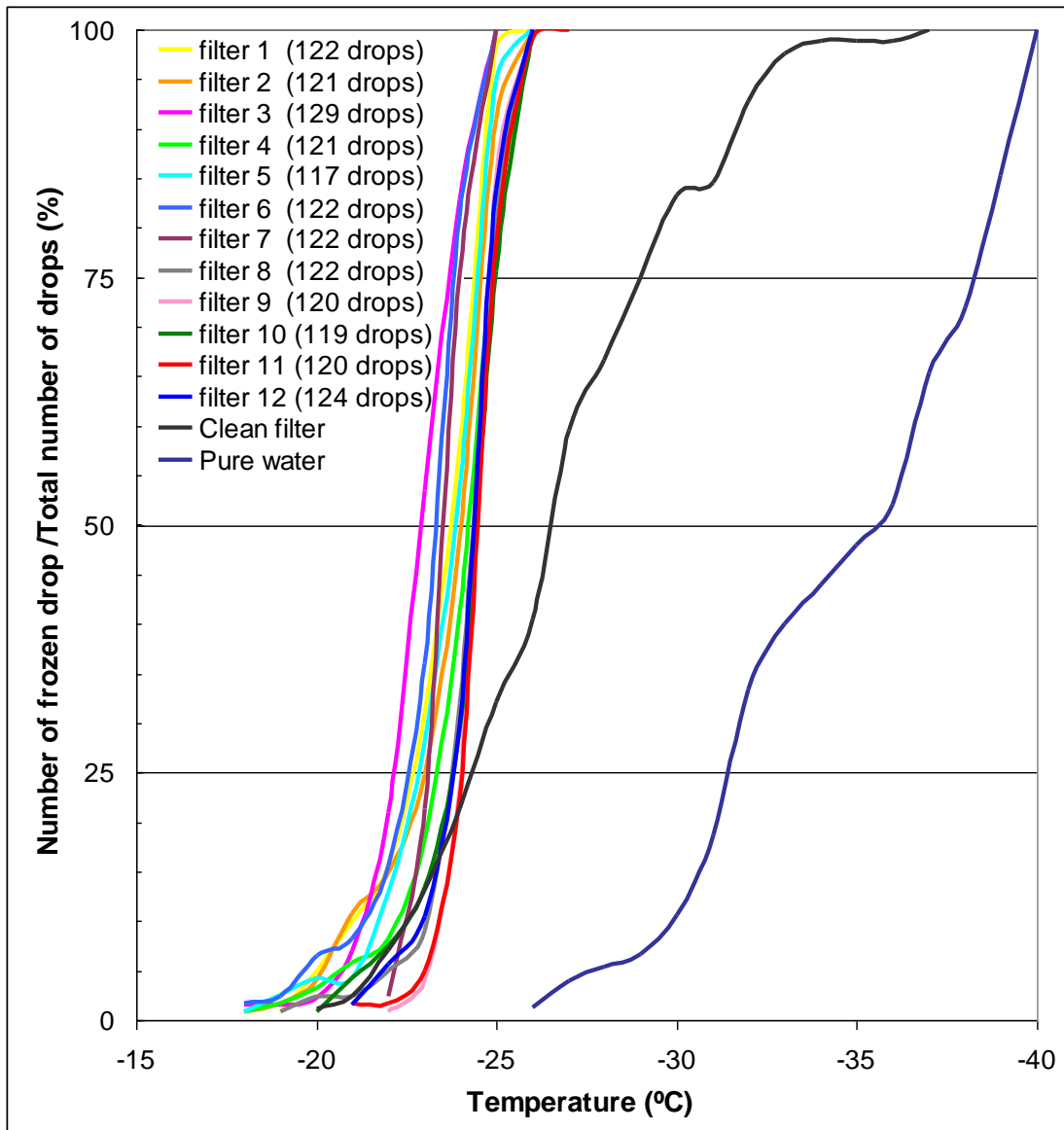


Fig. 3: The cumulative freezing spectrum of all the South Pole samples and the average spectrum of freezing temperature from clean filters and pure water.

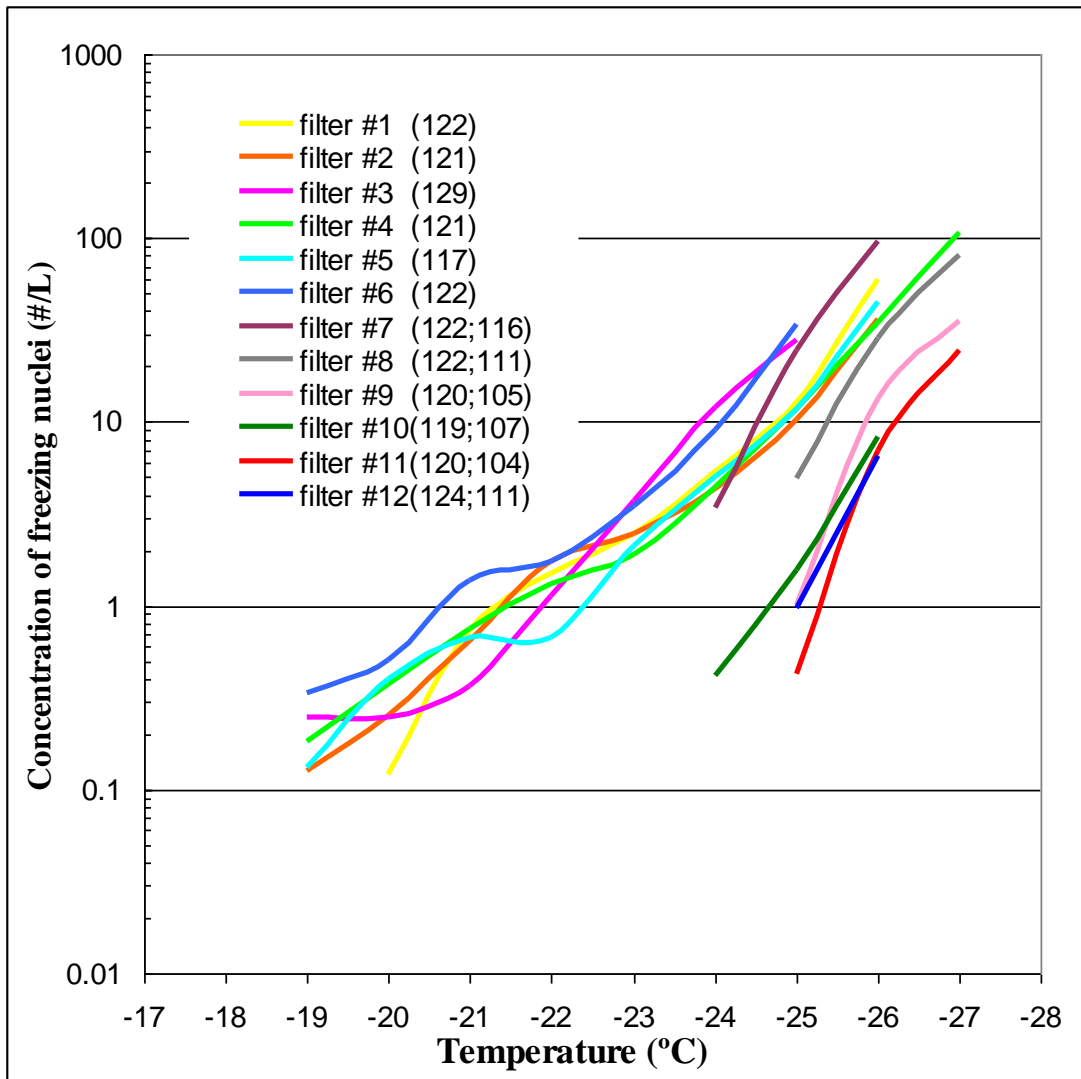


Fig. 4: Concentration of Freezing Nuclei calculated for the different samples. Note that for samples 7-12 all the drops that froze at similar temperatures to the blank filters were removed from the analysis. The first number in the parenthesis represents the number of drops used in the experiment and the second is the one used for the present calculations.

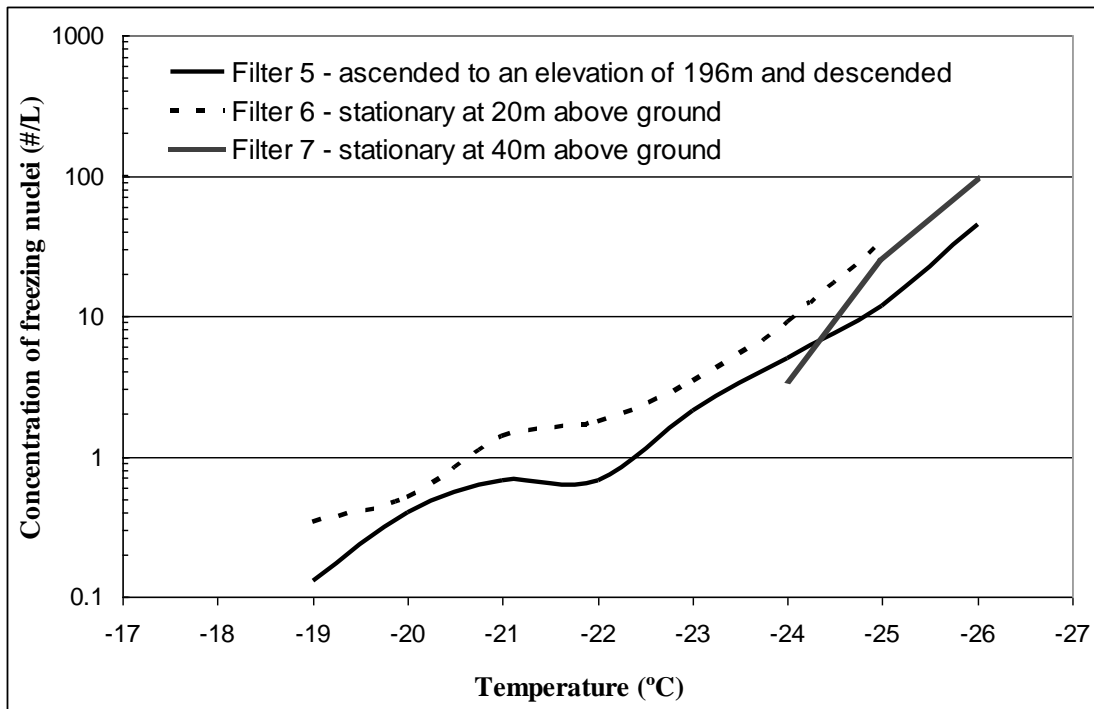


Fig. 5: Concentration of freezing nuclei from samples collected on the balloon.

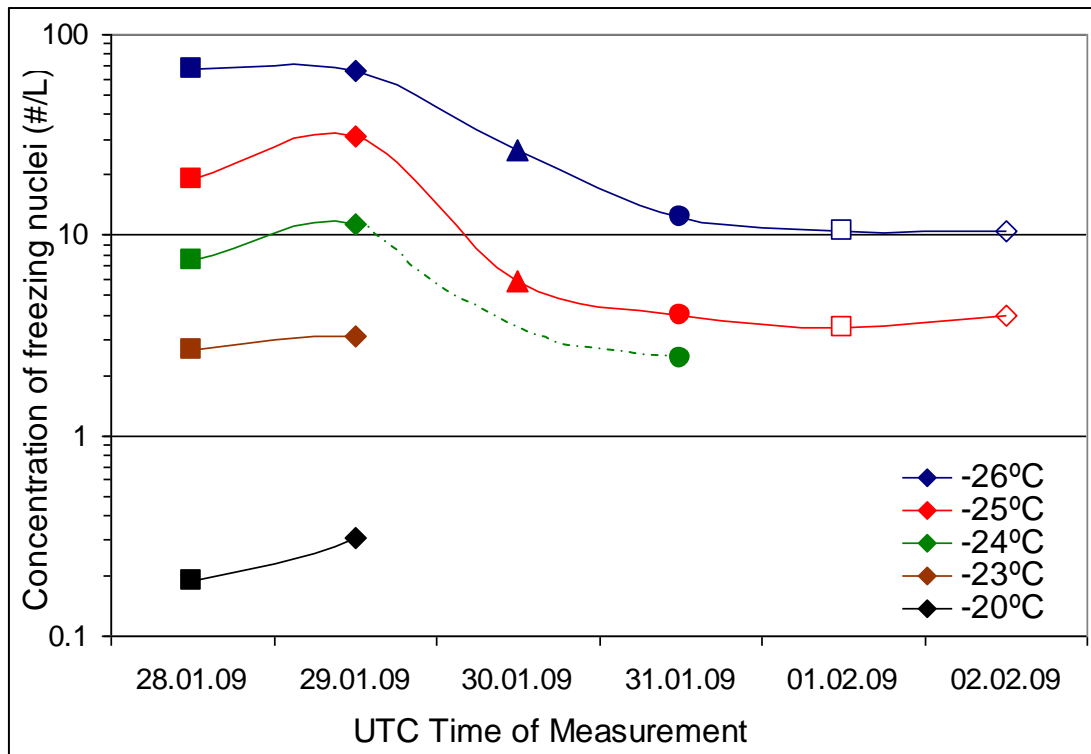


Fig. 6: Concentrations of Freezing Nuclei at different temperatures for the rooftop measurements on different days of the campaign. Each day is represented by a different symbol, and each temperature is represented by a different color.

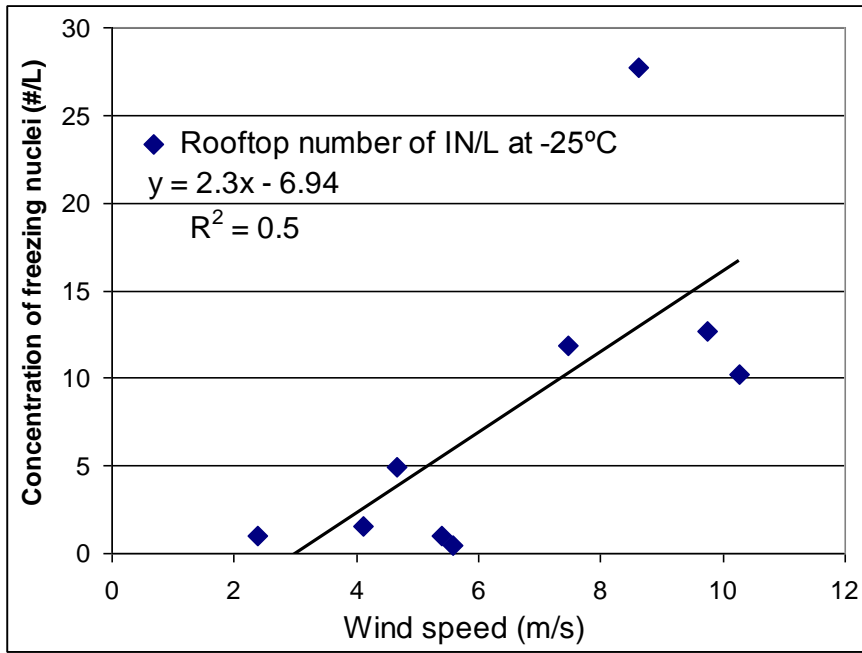


Fig. 7a: Concentration of freezing nuclei at -25°C as a function of wind speed.

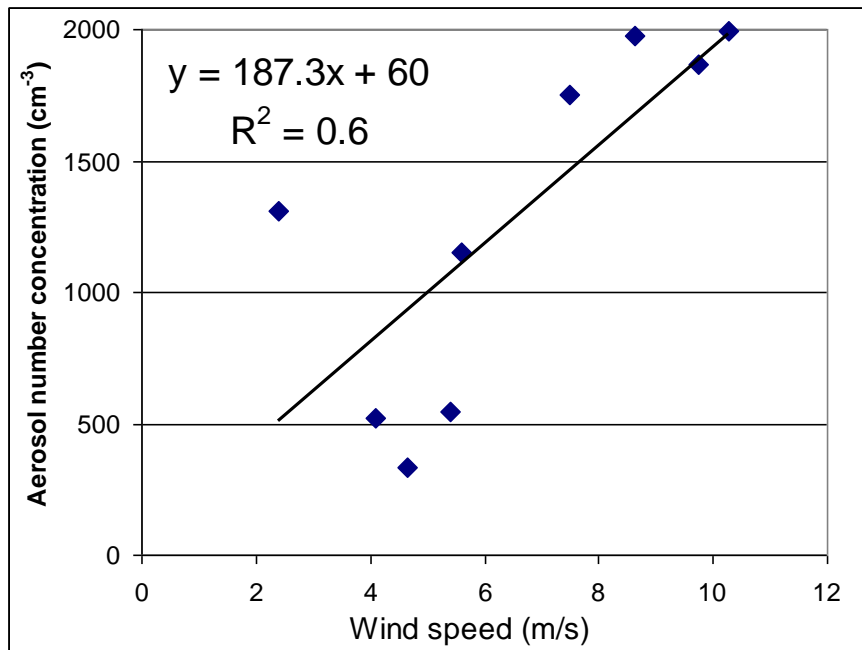


Fig. 7b: Concentration of aerosols > 0.01 μm as a function of wind speed.

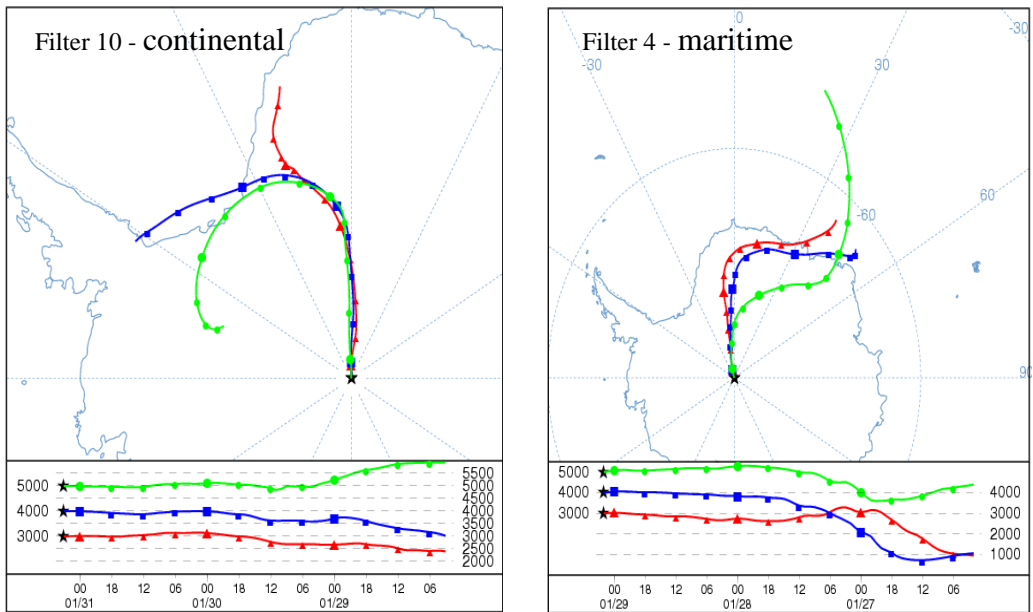


Fig. 8: Two examples of back trajectories of continental air collected on filter 10 and maritime air that was sampled on filter 4. The maps show the trajectory that the air parcels travel before reaching the station, located at an altitude of about 3000m ASL. The vertical projection for air arriving from 3000, 4000 and 5000 m is shown. Source: <http://www.arl.noaa.gov>.

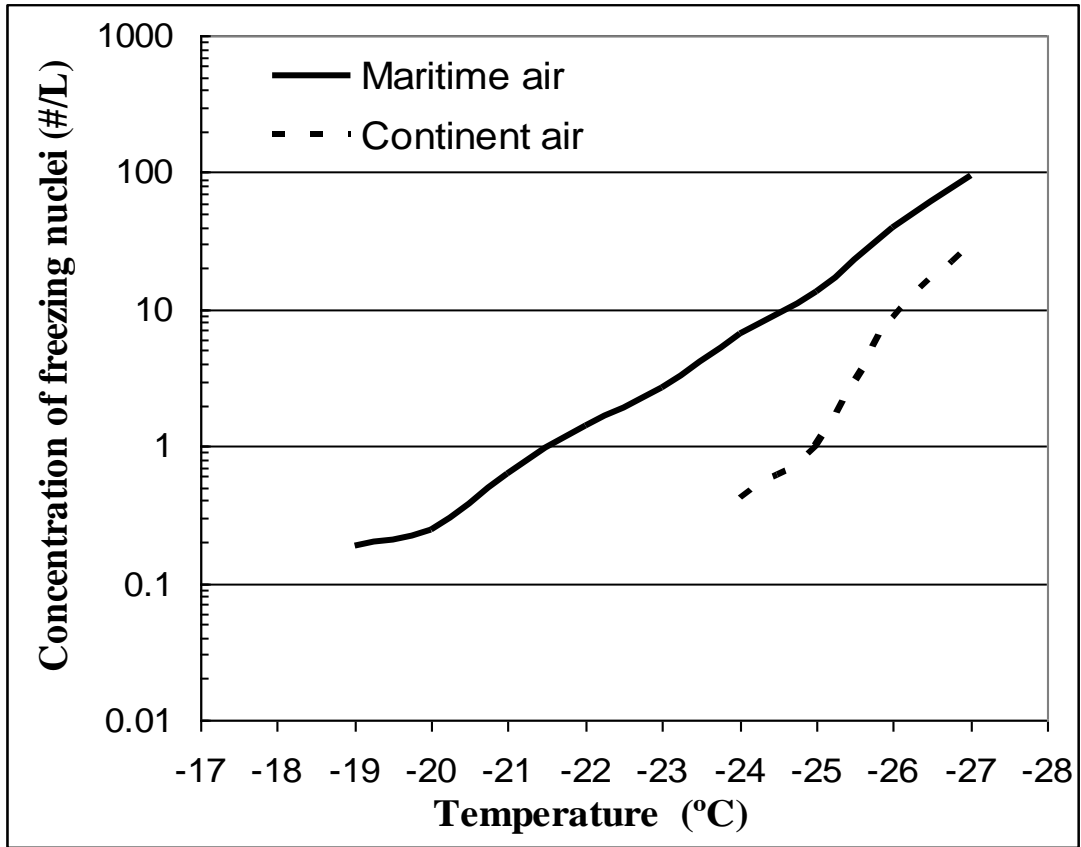


Fig. 9: Comparison between measurements taken on the rooftop based on air trajectory calculations. The solid curve represents ice nuclei concentrations in maritime air mass as measured from filters 1-4 and 8. The dashed curve is for ice nuclei concentrations in continental air mass as measured from filters 9-12. See text for definition of maritime and continental air.

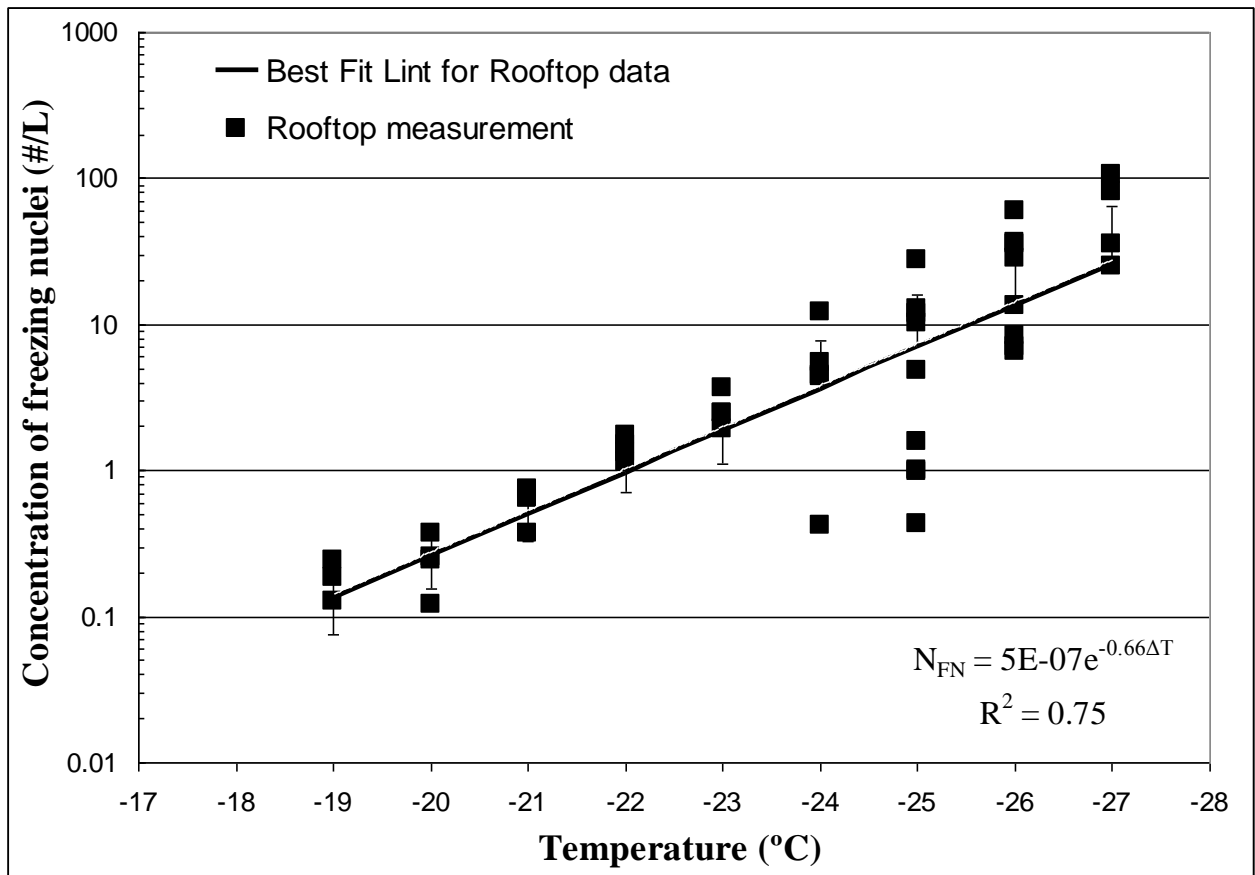


Fig. 10: The concentrations of FN in the air. The equation represents the best fit line to the rooftop data.

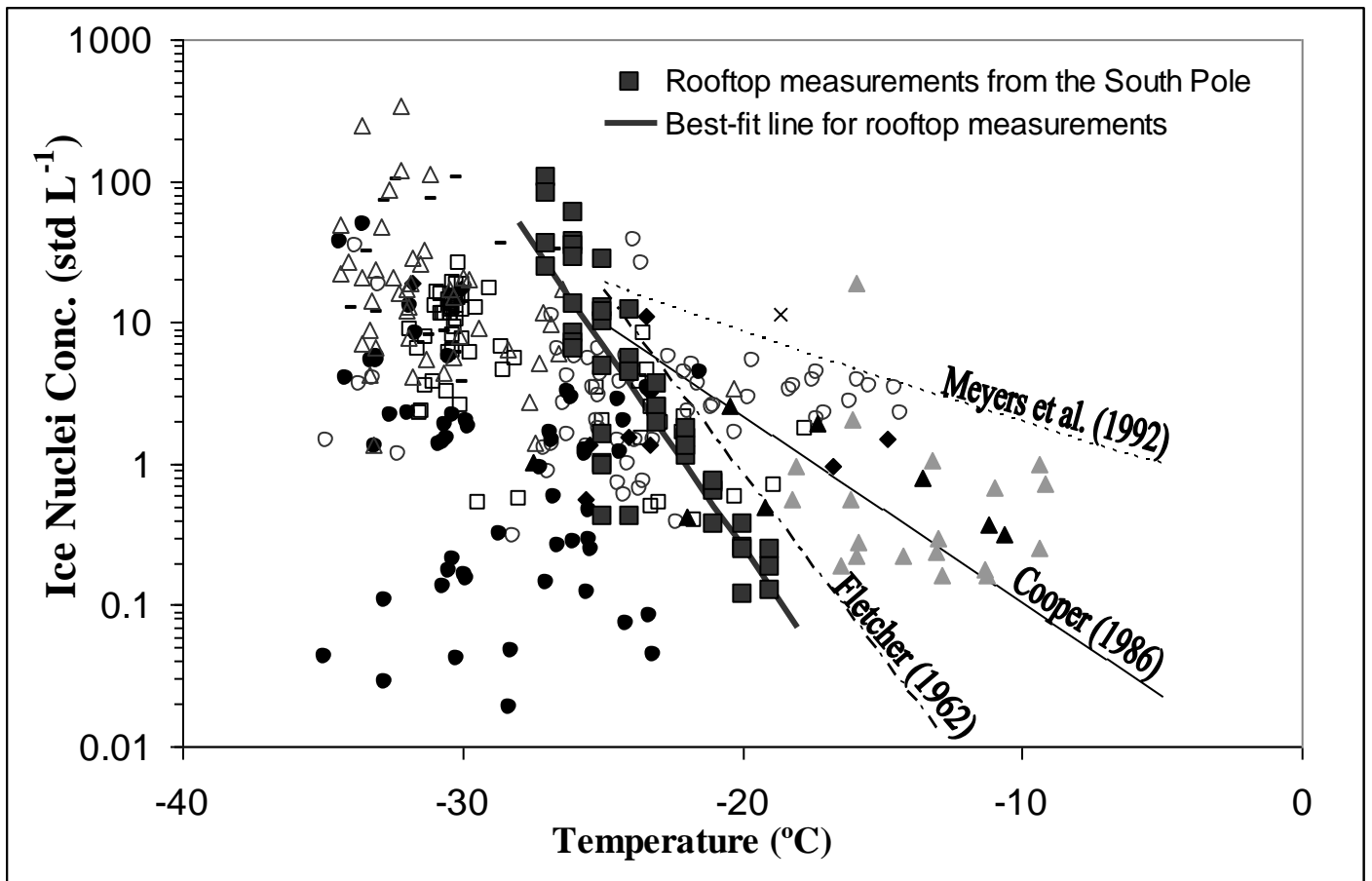


Fig. 11: Ice nuclei concentrations as a function of temperature from DeMott et al (2010) with the present measurements shown in black squares.