ATMOSPHERIC PARTICLES ACTING AS ICE FORMING NUCLEI IN DIFFERENT SIZE RANGES

G.Santachiara¹, L. Di Matteo¹, F.Prodi^{1,2} and F.Belosi¹

¹Institute ISAC-CNR, Clouds and Precipitation Group, Bologna, Italy ²Department of Physics, University of Ferrara, Dept. of Physics, Ferrara, Italy

1-INTRODUCTION

Aerosol particles that catalyze the formation of ice crystals in cloud are called lce Forming Nuclei (IFN or IN) and can form ice through different thermodynamic mechanisms or modes: deposition (or sorption), condensation-freezing, immersion and contact.

A wide variety of measurement techniques have been developed over the past 50 years for detecting IN and measuring their characteristics. These techniques include drop freezing, particle capture on supercooled droplets, particle sampling on filters followed by processing in static or dynamic chambers, continuous flow ice thermal diffusion chamber, and others.

It is generally agreed that ice will form on nuclei in response to different kinds of thermodynamic forcing, the primary variables being temperature and supersaturation with respect to ice and water (S_{ice} and S_{w} , respectively).

Even after 50 years of ice nuclei studies, a calibration ice nucleating material does not exist, and there is no general agreement on a standard technique for measuring IN. Ice nucleation measurement comparison workshops were held in the United States

in 1970 and 1975. Measurements from the various instruments did not produce tight quantitative agreement, but important factors for IN activation were identified, including sensitivity to vapour supersaturation, competition for vapour in filter processing, increasing activity with particle size.

Another important conclusion was the need to study with deeper insight the nucleation mechanisms.

The main goals of our experimental campaign can be summarized as follow:

a) Comparison of IN concentration in different size ranges, supersaturations and temperatures. b) Check a diurnal trend in the IN concentrations. c) Investigate the relationship between IN and condensation nuclei (CN).

2- EXPERIMENTAL

An experimental campaign was performed at a rural site (S.Pietro Capofiume, near Bologna) covering the period 09 -12 July 2007.

Simultaneous samplings of various aerosol fraction were taken on nitrocellulose membrane (Millipore HABG04700, nominal porosity 0.45 µm) of various aerosol fraction, i.e. PM1, PM2.5, PM10 and total suspended particles (PTS), four times a day (period 06 -22 h), at 3 m above ground level. The mean flow rate was 38.3 lpm with a sampling time of 10 min.

In addition the aerosol was sampled by means of an inertial spectrometer (INSPEC), which separates the particles on the basis of their aerodynamic size (Prodi et al., 1979). The flow rate was 20 I per hour and the sampling time was usually 12h, representing day-time and nigh-time samples.

Simultaneous measurements of particle number concentrations (CNC-TSI– Mod.3020, with 50% detection at ~10nm) and particle concentration in 15 different size classes starting from 0.3 μ m (Optical Spectrometer Grimm, Mod.1.108) were also performed.

Meteorological data (air temperature, wind speed, pressure) were recorded and the range of temperature, relative humidity (r.h.) and wind speed is reported in Table 1. The atmospheric pressure was about 1010 mbar.

Date of sampling	T, °C	r.h., %	v, m s⁻¹
09/07	25.4 -32	31-39	1.3-3.5
10/07	18.5 – 27.6	30-59	1.9-3.2
11/07	13.3-22.7	43-87	0.3-2.4
12/07	14.1-26.2	36-71	0.9-1.9

Table 1 – Details of the sampling campaign

Concentrations of IN were detected by the membrane filter technique, using a continuous diffusion chamber, which allows the control of the filter and of the air temperature, the latter saturated with respect to ice, thus obtaining different supersaturations. With this device the problem of competition among the nuclei on the filter for the vapour supply is minimized.

For each sampling, PM1, PM2.5, PM10 and PTS filters were cut into four pieces, and one piece for each fraction was inserted into the same metal plate in the diffusion camber and simultaneously developed.

The nucleation event was detected by counting on a monitor the number of ice crystal growing on the sampled filter, lighted with a grazing light.

Fig. 1 shows an example of ice crystals grown on different aerosol size range in the diffusion chamber.



Fig.1 – Picture of ice crystals grown on different aerosol size range

Measurements were made also at high supersaturation (Table 2), even through the common understanding is that the maximum supersaturation rarely exceeds 1% or 2% in natural clouds.

Table 2 – Operating conditions

T _{air} ,°C	T _{filter} , °C	S _{ice} , %	S _w , %
-15	-17	20	2
-15	-18	32	10
-17	-19	21	0

As a matter of fact the discrepancy between the concentration of ice nuclei and of ice crystals observed in clouds (Mossop et al., 1972) could be attributed to an occasional occurrence of very high supersaturations in clouds. During the entire freezing process the temperature of drop is 0°C, heat and water vapour are released into the surrounding air, whose temperature is lower than 0°C, and a region of supersaturated water vapour surrounds a freezing drop. Within this region of high supersaturation aerosol particles can act as CN or even IN (Rosinski, 1979).

3 - RESULTS AND DISCUSSION

Figs. 2, 3 and 4 show the time series of concentrations of aerosol particles active as IN in conditions reported in Table 2. Measurements below water saturation should allow to detecte deposition (sorption) nuclei, while those above water saturation allow the detection of deposition and condensation-freezing nuclei.



Fig.2 - Time series of concentrations of IFN (m^{-3}) active at T_{air} = -15°C, T_{filter} =-17°C



Fig.3 - Time series of concentrations of IN (m^{-3}) active at Tair = -15°C, Tfilter= -18°C



Fig.4 - Time series of concentrations of IN (m^{-3}) active at $T_{air} = -17^{\circ}C$, $T_{filter} = -19^{\circ}C$

From Table 3 it can be observed that PM1 fraction of the aerosol contributes to about 50% of the total measured IN for the atmospheric air sampled. Since the concentration of aerosol particles in the fine fraction is much higher than in the coarse one, it can be inferred that the nucleation efficiency, i.e. the fraction of natural aerosol particles nucleating ice at given temperature some and supersaturation. increases with increasing particle size. The increased ability of larger particles to act as freezing nuclei is counteracted in nature by the decrease in the concentration aerosol particles with increasing size.

This is in agreement also with the measurements performed with the INSPEC.

Table 3 –Percentage of IN obtained in PM1, PM2.5, PM10 fraction with respect to IN measured in the total suspended aerosol

	IN _{PM1} / IN _{PTS} , %	IN _{pm2.} / IN _{pts} , %	IN _{PM10} / IN _{PTS} , %
S _{ice} =20%; S _w =2%	61	82	70
S _{ice} =32%;S _w =10%	50	67	90
S _{ice} =21%;S _w =0%	49	62	83

The ratio between IN measured in the PM10 fraction and those in the PTS ranges from 70 to 90%, i.e. the dominant fraction of aerosol that can be activated concerns particles with aerodynamic diameter less than 10 μ m.

It is observed a positive correlation between higher S_w and S_{ice} values and IN concentration numbers. As a matter of fact the variation of filter temperature from -17°C to -18°C (T_{air} = -15°C) determines an increase of about three times (from 110 to 337 m^{-3}) in the IN average number concentration.

Experiments with $S_{ice} = 21\%$ and $S_w = 0\%$ (prevalently deposition nuclei) gives an IN concentration (about 200 m⁻³) between values obtained at $S_{ice}=20\%$; $S_w = 2\%$ and $S_{ice}=32\%$; $S_w = 10\%$ (110 and 337 m⁻³, respectively).

For comparison, Castro et al. (1998) in a rural area, far away from any potential source of pollution, measured at ground level a background concentration of IN active at -15°C, -19°C and -23°C at water saturation. of 7, 35 and 95 l⁻¹. There is not correlation between IN measured in the different size range and number particle concentration measured with the counter spectrometer (D>0.3 µm) and CNC, even if we can observe that the highest IN concentrations, measured when air masses came from west (10/07/2007), are coupled with high values of CN (Fig.5). Lower IN concentration values were measured when air masses came from N-NE.

The ratio of IN to CN concentration, depending on T, Sice and Sw , in our measurements ranges from about 1:108 to $1:10^7$. IN constitutes a very small fraction of the aerosol population.



Fig. 5 – Correlation between IN measured $(T_{air} = -15^{\circ}C ; T_{filter} = -17^{\circ}C)$ in total suspended aerosol (m⁻³) and CN concentration (cm⁻³)

There is no correlation between particle number measured with the optical counter and CN concentration (Fig.6). Therefore, it can be concluded that the variation in the particle concentration during the experimental campaign is mainly due to particles smaller than 0.3 μ m.

A few filters sampled with INSPEC during day (6 -18h) and night time (18-06 h) do

not allow to evidence a diurnal trend in the IN concentration.



Fig.6 - Time series of particle concentration measured with optical (I^{-1}) and CN counters (cm^{-3}) .

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