An Experimental Technique to Study the Effect of Size on Ice Nucleation

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12 August 1980

1. Introduction

The size of the particles acting as nuclei of the ice phase in the atmosphere is recognized as an important parameter in their mode of action. Nevertheless, information on their size distribution is scanty and sometimes conflicting.

A review on the status of ice nuclei spectrometry is presented by Roddy (1978). Georgii and Kleinjung (1967) have found ice nuclei better correlated with large particles, while Vali (1966) attributes a substantial fraction of freezing nuclei to small particles. The effect of particle size on the dynamics of artificial ice nuclei in real clouds has been specifically investigated by Langer *et al.* (1978).

Direct measurements of the size distribution of natural ice nuclei have been performed with a Goetz aerosol spectrometer. However, the instrument does not behave as a real spectrometer and is not suitable for field sampling. In addition, particles below a certain size are not deposited on the foil of the centrifuge; the size distribution of the particles below the critical size is not known and any information of the nucleating ability of this fraction is lost.

In this note a technique is presented which combines aerodynamic particle separation with deposition on a membrane filter. The device is portable and can be applied to field aerodynamic spectrometry of aerosol particles; in particular, the known membrane filter technique of ice crystal detection can be applied and the aerodynamic distribution of the ice nuclei can be measured.

2. Experimental technique

A novel type of size spectrometer has been recently developed that looks promising for aerodynamically separating aerosol particles [Inertial Spectrometer (see Prodi V. *et al.*, 1979)].

It is schematically composed of a 2 mm deep rectangular channel, with a 90° bend which is flushed with clear air (Fig. 1a). A thin aerosol sheet is injected upstream of the bend. While the streamlines follow the curvature, because of their inertia the particles tend to persist in the initial velocity at the bend. Therefore, the particles depart from the original streamline by a distance which is a function of the aerodynamic diameter. The external wall downstream of the bend is made of a membrane filter through which air is sucked. The particles are sepa-

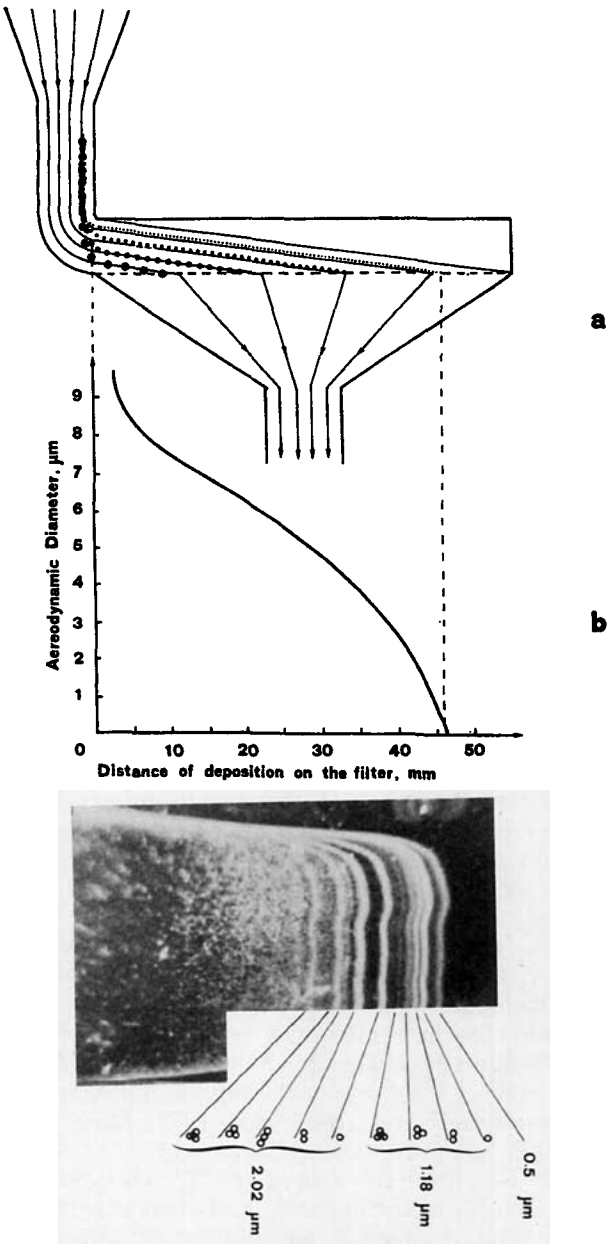

FIG. 1. Characteristics of the Inertial Spectrometer: (a) calibration curve of the instrument at 0.5 L min^{-1} sampled air and 5.5 L min^{-1} clean air flowrate; (b) portion of the surface of the membrane filter with a deposit of latex spheres of 0.5, 1.18 and 2.02 μm size; (c) the latex spheres are separated in singlets, doublets and higher multiplicities, which is an indication of the good resolution of the instrument.

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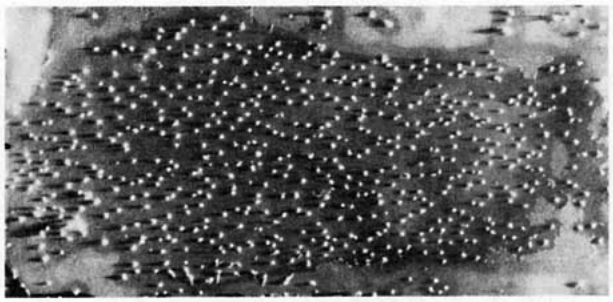

FIG. 2. Surface of a membrane filter after sampling urban air by the inertial spectrometer and after analysis for ice nuclei in a Stevenson Gagin diffusion chamber. Temperature of the filter, −19°C; temperature of the surrounding ice corona, −16°C.

rated according to their size while airborne; the small separation is strongly magnified by the aerodynamic projection to the filter surface.

The filter collects the aerosol with the characteristic efficiency of the membrane filters. Those particles too small to be deviated will deposit at the end of the filter; coarse particles are precipitated at the beginning and the intermediate sizes, between 0.5 and 11 μm, are continuously separated as is apparent from the calibration curve (Fig. 1b).

The resolution of the instrument is such that the latex sphere aerosol produced by atomizing monodisperse suspensions can be clearly separated in singlets, doublets, triplets and higher multiplicities (Fig. 1c). The figure shows also the effect of side walls—the useful part is that characterized by parallel deposition bands. The filter can be analyzed for the specific application required (microanalysis, scanning for radioactivity, etc.).

Ice nuclei measurement techniques using membrane filters are widely employed (Bigg *et al.*, 1963; Stevenson, 1968). It was therefore thought to combine the size separation and filter technique to produce an instrument which is able to determine the size effect on ice nucleation.

After exposing the filter to the aerosol stream in the inertial spectrometer the filter is impregnated from below in a gauged film of petroleum jelly on a metal plate, and developed in a diffusion chamber similar to that used by Prodi and Wirth (1972). The growth of the crystals is visually followed with the naked eye and with a low magnification microscope. Photographs of the filter are also taken at time intervals. As examples of the measurements performed by this technique we present the results from three tests.

In the first case urban air was sampled for 5 h at 0.5 L min^{-1}, with a clean air flowrate in the spectrometer of 5.5 L min^{-1}. The temperature during the development of the filter was −19°C and that of the heated ice corona −16°C. In this case the striking feature was a size dependent onset of crystal growth. After few minutes crystals appeared at the large

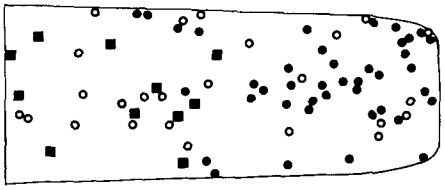

FIG. 3. Map of the filter surface drawn on the basis of serial photographs of the ice nuclei active after 2 min (black squares), 4 min (black circles) and 8 min (open circles). The filter sampled urban air.

particle side and the crystallization proceeded in fronts along equal size contours. The development was completed in 10 min with a formation of small and densely distributed crystals along the fine particle line. Fig. 2 shows the filter after development when lighted at grazing illumination; the picture is taken of the crystals grown to large sizes due to the poor contrast of the small crystals on the white background of the filters. The ice nuclei concentration in this case was of 2.3 L^{-1}.

The development of another filter which also sampled urban air in the same conditions of operation of the instrument was followed by serial photographs at 2, 4 and 8 min. The crystal onset after these time intervals was mapped from the photographs and is reported in Fig. 3. A dependence on size of the onset of crystal growth was observed also in this case though not so evident as in the previous test.

As an interesting application of the technique, the direct determination of the fraction of the aerosol particles which nucleates ice in each particle size interval can be performed. This is the parameter which has to be introduced into cloud models in order to appropriately reproduce the microphysics. This measurement has been performed by cutting in two, along its length, the filter after sampling. One half of it is inspected at the optical and electron microscope for counting the aerosol particles and the other is developed for ice nuclei detection. The results of this determination on a filter, which sampled

TABLE 1. Fraction of aerosol particles nucleating ice in the deposition mode at $T = -19$°C at the indicated size intervals [rural aerosol, Turbigo (Novara)].

Distance from the inlet slid (mm)	Particle aerodynamic diameter interval (μm)	Fraction of ice nuclei
0–5	>8.1	9.8×10^{-4}
5–10	8.1–7.5	4.8×10^{-4}
10–15	7.5–7.0	3.5×10^{-4}
15–20	7.0–6.3	3.3×10^{-4}
20–25	6.3–5.6	2.3×10^{-4}
25–30	5.6–4.7	1.5×10^{-4}
30–35	4.7–3.8	1.1×10^{-4}
35–40	3.8–2.5	9.0×10^{-5}
40–45	2.5–0.7	1.1×10^{-6}
45–50	<0.7	1.6×10^{-9}

rural aerosol at Turbigo (Novara) in the same operation conditons of the instrument and filter development, are reported in Table 1. The fraction of aerosol particles nucleating ice in this case ranged from 10^{-3} for particles of aerodynamic diameter $>8\ \mu$m to 10^{-6} for particles around 1 μm.

This has proven as a useful technique for a more detailed characterization of the ice nuclei population and more work on this is in progress.

Acknowledgments. The technical assistance of A. Malossini and M. Tercon, and discussions with Dr. J. Rosinski are gratefully acknowledged. The research is partly sponsored by the Piano Finalizzato "Promozione della qualità dell'Ambiente," CNR.

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