On the Use of Membrane Filters in Ice Nuclei Measurements

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1. Introduction

Since the first application of membrane filters to the detection of the ice nuclei content of atmospheric air (Bigg *et al.*, 1961, 1963), this method has probably become, with some important modifications by Stevenson (1968), Gagin and Arroyo (1969) and Gagin (1971), the most widely used. This is not surprising if one considers the many advantages of this method: chiefly an accurate control of both temperature and supersaturation, and the separation in space and time of the sampling from the "development"; both assure a relatively good reproducibility and the possibility of intercomparison of different measurements. In addition, the worldwide results obtained by membrane filters are well comparable with those obtained by other methods (e.g., different kinds of mixing and expansion chambers, acoustic counters, etc.) especially if one accepts that differences of, say, 50–200% have no practical consequences.

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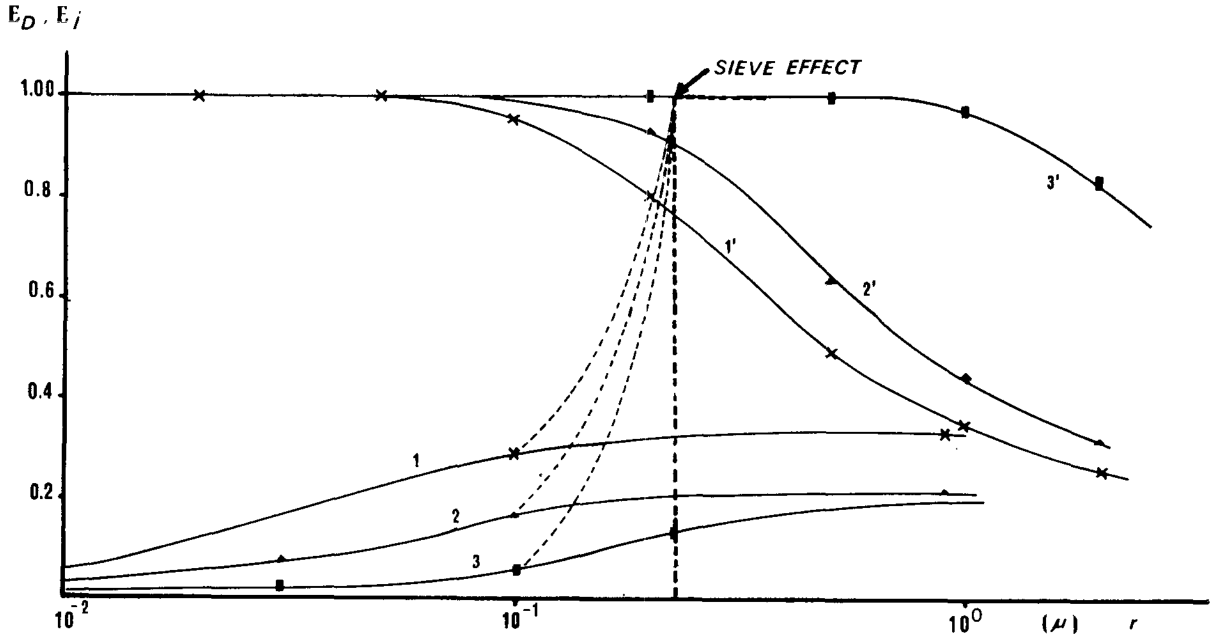

FIG. 1. Capture efficiency of membrane filters by diffusion inside filter pores (1′, 2′, 3′) and by impaction on the surface of filters (1, 2, 3) as a function of particles radius. Type 1: Synpore; 2: Millipore HA (I); 3: Millipore HA (II).

However, *in situ* measurements performed in and around the vicinity of clouds with not too low (but negative) summit temperatures (see, for example, Mossop and Ono, 1969; Mossop *et al.*, 1972) have demonstrated that a large discrepancy exists between the concentrations of ice nuclei and those of ice crystals. The difference in orders of magnitude can be as high as four, which arouses suspicion as to the reliability of ice nuclei measurements.

Secondary processes in the development (formation and propagation) of the ice phase in the cloud are hypothesized in order to account for the large discrepancies between ice nucleus concentrations and those of the ice particles. However, if we assume for a moment that no secondary process takes place, a direct consequence would be that our methods give only very rough estimates of the actual concentrations of ice nuclei. This would further imply that attempts to give definite answers to the questions concerning their origin, chemical composition, and size distribution should be considered as lacking an adequate physical and statistical basis and are thus all misleading. Therefore, the need is stressed for more critical investigations of the different methods of measuring ice nucleus concentrations.

TABLE 1. Filter, particle and airflow specifications.

	Synpore 6 (Czechoslovakian)	Millipore HA (I)	Millipore HA (II)
Velocity of airflow (cm sec^{-1})	63.6	36.6	4.2
Density of particle (gm cm^{-3})	1.5	1.5	1.5
Viscosity of air (poise)	1.8×10^{-4}	1.8×10^{-4}	1.8×10^{-4}
Porosity (ϵ)	0.67	0.79	0.79
Nominal pore radius (cm)	2×10^{-5}	2.25×10^{-5}	2.25×10^{-5}

2. The role of impaction and diffusion

We will consider now some physical processes involved in the membrane filter method. In using membrane filters in aerosol studies, in general, and in ice nuclei measurements, in particular, it is generally assumed that the particles, down to diameters of about two orders of magnitude smaller than the pore size, are deposited on the front surface (Mossop and Thorndike, 1966; Fuchs, 1964). This supposition, however, has to be verified by considering the processes involved in the filtration of particles by membrane filters. In fact, particles having sizes comparable with the diameters of the pores are captured mostly by two mechanisms: diffusion and impaction. The curves corresponding to the two processes have been computed (Fig. 1) for filters and air speeds specified in Table 1, and for particles of preselected density. The upper curves show the effectiveness of deposition by diffusion inside the filter pores calculated on the basis of Twomey's (1962) approach, while the lower curves show the effectiveness of deposition by impaction on the surface of the filters, on the basis of the Pich (1966) theory. The following equation of Pich has been used:

$$E_i = 2E^* - E^{*2}, \qquad (1)$$

where

$$\left.\begin{aligned} E^* &= \frac{1}{1+\alpha}\left[4K\sqrt{\bar{\alpha}}+8K^2\alpha\exp\left(-\frac{1}{2K\sqrt{\bar{\alpha}}}\right)-8K^2\alpha\right] \\ \alpha &= \frac{\sqrt{\epsilon}}{1-\sqrt{\epsilon}};\quad K=\frac{1}{9}\frac{\rho r^2 v_0}{\eta R}C;\quad C=1+\frac{6\times 10^{-6}}{r} \end{aligned}\right\}.$$

In this expression E_i is the impaction efficiency of the membrane filters, having uniform circular holes, ϵ the porosity of the filter, and K the Stokes' parameter in the expression for spherical particles of density ρ and radius r; v_0 the linear air velocity through the filter and η the dynamical viscosity of the air; R the radius of filter pores; and C the Cunningham correction factor (not applied by Pich).

In practice, when evaluating the relative effectiveness of these two mechanisms we are concerned only with the portions of the curves below the nominal pore sizes. So the curves of the effectiveness by impaction show a deep slope (dotted lines) due to the sieve effect. As a result, the collection efficiency of the filter surface by impaction is $\leqslant 30\%$ for particles $<10^{-5}$ cm, depending on the velocity of filtration. These particles, however, are retained by the filter inside the pores, with the effectiveness being very close to 100%. It is also observed that the efficiency of impaction increases with increasing Stokes numbers, while for non-inertial particles (Stokes numbers close to zero) this efficiency approaches zero.

The dependence of the efficiency by impaction on the velocity of flow is also observed in Fig. 2, which is a part of Fig. 4 from the work of Pich and Spurny (1965).

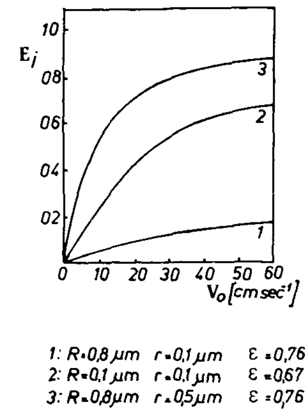

FIG. 2. Dependance of capture efficiency by impaction on the velocity of the air flow: R, pore radius; r, particle radius; ϵ, porosity; v_0, linear filtration velocity.

3. Conclusions

Accumulating evidence from experimental works (Vali, 1966, 1968) and findings of theoretical investigations (Bonis, 1972) as well, seem to support the hypothesis that the major source of the natural ice nuclei originates from the 0.01 μm (and smaller) size ranges. For particles of these sizes the impaction efficiency of filters—generally used with nominal pore sizes of 0.45 μm for ice nuclei detection and concentration measurements—varies between 1–10% depending on the linear filtration velocity.

If this is the case, all measurements of ice nuclei concentrations by membrane filters have been concerned solely with a very small fraction of these particles. In addition, the concentration of this fraction critically depends on the conditions of sampling and the developing procedures: that is, the complicated mechanism of the impaction around the pores of the filters is governed by a number of parameters appearing in (1). Therefore, it is not surprising that the measurable portion of ice nuclei could not unambiguously be related to other kinds of nuclei or to some simultaneously observed weather phenomenon. As for the agreement with the results of other techniques based mostly on mixing and expansion chambers, it might be that these, too, seriously underestimate the concentrations, possibly because of the short time of supersaturation maintained, inhomogeneities in supersaturation, and the effect of the walls in the mixing boxes generally used.

All this would suggest a need for the re-evaluation of the techniques, beginning from membrane filters. This might offer a key to a better understanding of several problems involving natural ice forming nuclei, of which the most important, from a theoretical point of view, is the relationship between ice nuclei and ice crystal concentrations in supercooled clouds.

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