Measurements of the Size of Natural Cloud Nuclei by Means of Nuclepore Filters

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ABSTRACT

The size of cloud nuclei acting at 0.75% supersaturation was estimated by using varying flow rates through Nuclepore filters to discriminate between different sizes. The nuclei, sampled in clean continental and maritime air at Robertson, N.S.W., were found to be small, not much greater than the theoretical minimum radius of $\sim 10^{-6}$ cm permitted by nucleation theory.

This result agrees quite well with previous estimates of cloud-nucleus size from experiments carried out at Chesapeake Bay, Md. It is concluded that the atmospheric residence time of cloud nuclei cannot be more than a few days and that they must be composed entirely or partly of a water-soluble material since an insoluble particle of such a small size could not nucleate condensation at supersaturations of the order of 1% or less.

1. Introduction

The size of cloud nuclei is relevant to the question of their composition and origin. It is also relevant to the possible absorption of visible light by clouds, which Danielson *et al.* (1969) suggested could be brought about by the presence of nuclei in cloud droplets. Previous measurements by the writer (Twomey, 1965) suggested an average particle radius of a few hundredths of a micron, which implied that the nuclei must be a soluble material (hence unable both because of their small size and their composition to give rise to appreciable shortwave absorption in clouds).

These earlier measurements (made at Chesapeake Bay, Md.) used a long length of stainless steel tubing as a size-dependent filter, the characteristics of which could be varied by changing the flow rate. With the advent of Nuclepore filters, a greater variety of filtration characteristics can be obtained in a much more convenient manner. The present paper will describe the results of a series of measurements made at Robertson, N. S. W., a site which experiences unpolluted maritime air, unpolluted continental air, and well-polluted urban-industrial air under different wind directions.

2. Experimental procedure

A Nuclepore[1] filter contains a large number of uniform pores of well-defined cylindrical geometry. Their filtration characteristics can thus be calculated theoretically with confidence from the physical equations for particle diffusion and inertial separation. Spurny *et al.* (1969) have compared experimental results with computed filtration efficiencies and concluded that theory predicted accurately the behavior of the filters in all respects except (not surprisingly) for the variation of filtration efficiency as the pores became partly clogged with collected material. (Even this variation was predicted reasonably well by theory.)

To measure the size of cloud nuclei, sample air from a reservoir was passed through filters and the number of nuclei remaining *after* filtration were measured for various flow rates.[2] Since there was no need to build up the material caught by the filter, the pads were changed frequently to avoid excessive accumulation and the resultant uncertainties caused by clogging of the pores.

3. Results

a. Average size of the nuclei

The first experiments utilized filter pads with 0.25, 1 and 4 μ pore radii. It was found that very few nuclei penetrated the 0.25 μ filter at the flow rates which could be obtained ($\leqslant 10$ cm^3 sec^{-1} with this pore size), so this filter was omitted in most later measurements. The removal of almost all the nuclei by this filter showed that there were very few nuclei in the radius range 0.1–0.2 μ for which up to 20% transmission is given by the 0.25 μ radius filter at flow rates of order 5 cm^3 sec^{-1} (see Fig. 1).

It was also found that the coarsest filter (4 μ radius) did not remove the nuclei very effectively and produced high flow rates which consumed the air samples quickly. That filter was therefore replaced by a 2.5 μ radius filter in later experiments.

The most useful information was given by the filter with a 1 μ pore radius. This filter could be made to pass most of the nuclei by using flow rates of ~ 100 cm^3 sec^{-1},

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[1] General Electric Company.

[2] Nucleus concentrations were measured by direct photography of the cloud formed in a thermal diffusion chamber.

but when the flow rate was reduced to 20 cm^3 sec^{-1} most of the nuclei were removed. The median transmission over a series of observations over the period July–October 1970 was 68% at 80 cm^3 sec^{-1} and 23% at 20 cm^3 sec^{-1}. The theoretical transmissions for various particle radii (assuming a particle density of 2.0 gm cm^{-3}) are as follows:

	Radius (μ)				
	0.01	0.02	0.05	0.1	0.2
Transmission at 80 cm^3 sec^{-1}	0.66	0.79	0.89	0.89	0.80
Transmission at 20 cm^3 sec^{-1}	0.17	0.54	0.81	0.87	0.86

Nucleation theory sets a minimum radius of approximately 0.01 μ for a soluble particle capable of nucleating condensation at 0.7% supersaturation. The experimental results accord best with an average size very close to this lower limit.

b. *Size distribution*

The measurement accuracy of cloud nucleus concentrations is not high enough to justify attempts to obtain any detailed information about the size distribution. Certain general aspects can, however, be discussed:

1) Since the number of particles emerging was found to increase with increasing flow velocity, the primary removal mechanism must be diffusion to the pore walls. Inertial removal contributes only a few percent (at most) to the filtration efficiency for particles $<10^{-5}$ cm radius at the flow rates used in these experiments. The fraction of particles surviving can therefore be obtained with little loss of accuracy by the diffusion loss equation (Twomey, 1963)

$$\frac{n}{n_0}=0.81904\exp(-3.6568\pi Lq^{-1}D)+0.09752\exp(-22.304\pi Lq^{-1}D)+\cdots,$$

where L is the length of a filter pore, q the volumetric flow rate through each pore, and D the particle diffusion coefficient; provided $n/n_0 \lesssim 0.7$ all but the first right-hand term can be discarded.

2) Füchs *et al.* (1962) have shown how a plot of the fraction of particles emerging from a rectangular channel (diffusion battery) against the logarithm of the diffusion parameter, $\xi=7.5407ba^{-1}Lq^{-1}$, can be used to obtain the logarithmic mean and standard deviation of a log-normal distribution. Their method can be applied almost unchanged when Nuclepore filters with cylindrical pores are used, provided the values of n/n_0 are sufficiently less than unity that only the first term is needed in each diffusion equation. The fraction emerging from a rectangular channel of depth a and width b ($b\gg a$) is given by

$$\frac{n}{n_0}=0.9104\exp(-7.5407ba^{-1}Lq^{-1}D)+0.05315\exp(-85.73ba^{-1}Lq^{-1}D)+\cdots,$$

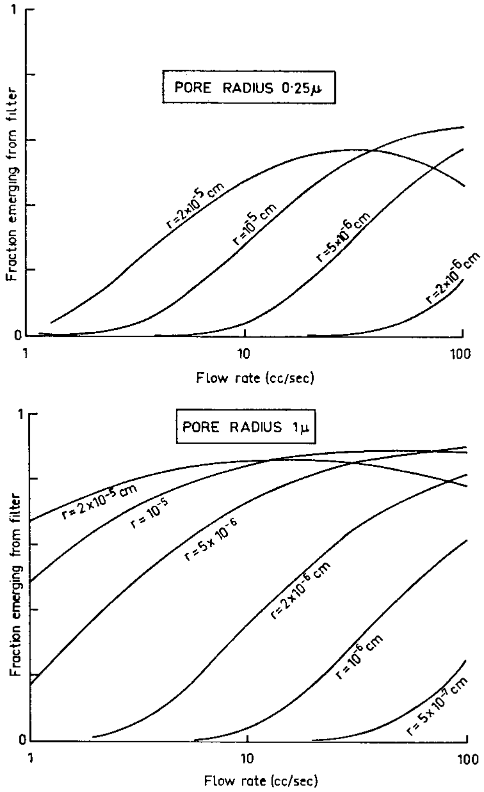

FIG. 1. Transmission through Nuclepore filters at varying flow rates for filters with pore radii of 0.25 and 1.0 μ.

which, for n/n_0 appreciably less than unity, simplifies to

$$\frac{n}{n_0}=0.9104\exp(-7.5407ba^{-1}Lq^{-1}D)=0.9104\exp(-\xi D).$$

For a cylindrical channel

$$\frac{n}{n_0}=0.81904\exp(-3.6568\pi Lq^{-1}D),$$

which can be rewritten as

$$\frac{0.9104}{0.81904}\frac{n}{n_0}=0.9104\exp(-3.6568\pi Lq^{-1}D).$$

Thus, if $3.6568\pi Lq^{-1}$ is used as the diffusion parameter and the fraction emerging is multiplied by 0.9104/0.81904, the method of Füchs *et al.* can be used equally well with cylindrical filter elements. The Füchs diffusion parameter $1.8852y$ is the same as ξ in the notation used above.

Fig. 2 shows median data for cloud nuclei sampled during May–November 1970 plotted on a reproduction of the penetration curves given by Füchs *et al.* The Füchs curves comprise 11 sets of five curves; for each set the logarithmic mean radius is the same, but the

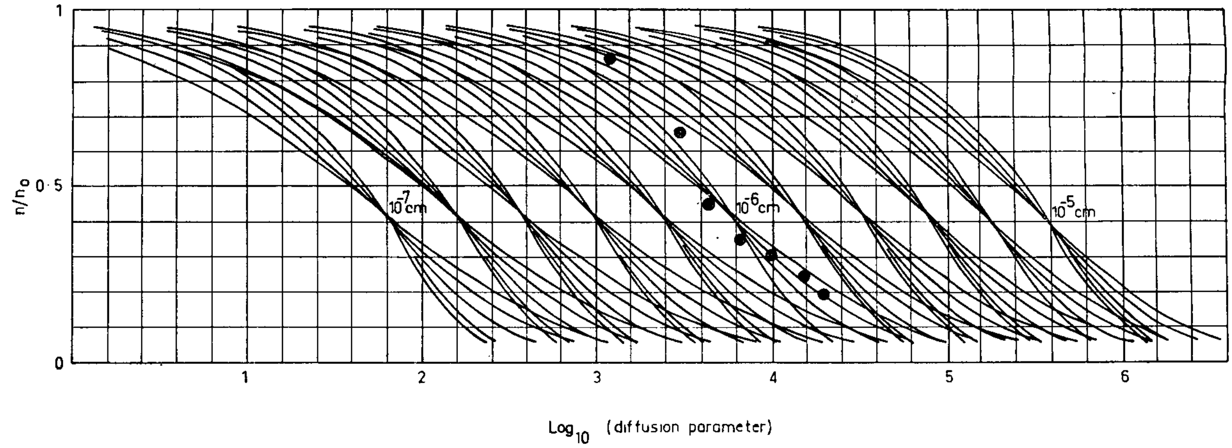

FIG. 2. Reproduction of curves from Füchs *et al.* (1962) for deriving logarithmic mean and standard deviation of log-normal distributions. Experimental points for cloud nuclei are superimposed, using the adjustments described in the text.

members of the set differ with respect to logarithmic standard deviation. The steepest curve of each set corresponds to a monodisperse aerosol (logarithmic standard deviation $\beta=0$), while the distributions become broader for the members of the set for which β takes successively the values of 0.1, 0.2, 0.3 and 0.4. The experimental curves clearly do not follow any of the Füchs curves closely, indicating that the distribution is not close to log-normal. However, they certainly suggest a logarithmic mean radius between 10^{-6} and 1.6×10^{-6} cm and a relatively broad distribution.

c. *Possible bimodality*

On many occasions it was found that when the flow rate was reduced until the number of nuclei emerging from the filter diminished to a low level, close to the minimum count which could be made reliably (~10 cm^{-3}), further quite large reductions in flow rate failed to eliminate the remaining few nuclei. This suggested that these remaining nuclei were distinctly larger than the rest, with a correspondingly smaller diffusion coefficient. It was also found that the number of these few larger particles was comparable to the number of nuclei remaining when the aerosol was passed through heated tubes known to be hot enough to evaporate ammonium salts but not sodium salts (Twomey, 1971). There is, of course, no assurance that the same nuclei survived in the two situations but the suggestion is strong that there is a break or at least a deep minimum in the size distribution of natural cloud nuclei separating the smaller, volatile (probably ammonium sulfate) particles from the larger, fewer and much less volatile (probable sodium chloride) fraction. The measurements suggest that radii of the latter are greater than at least 0.05 μ. Cascade operation of the boiler apparatus and the filters would help to elucidate this point.

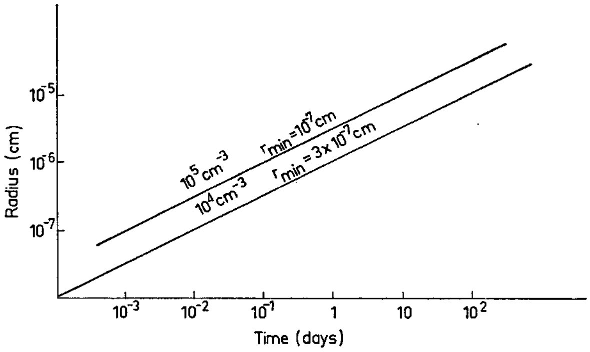

FIG. 3. The relationship between radius and coagulation "time constant" for particles coagulating with an aerosol following a Junge distribution down to a minimum radius $r_{\min}$.

4. Discussion

The size suggested by these experiments for nuclei acting at a supersaturation up to 0.75% is 10^{-6} to 2.5×10^{-6} cm radius. The minimum size permitted by nucleation theory is a little below 10^{-6} cm. If one makes a few reasonable simplifying assumptions (i.e., that the supersaturation spectrum of the nuclei is approximately $N\propto S^k$, that the critical supersaturation of a soluble particle of radius r is proportional to $r^{-\frac{3}{2}}$, and that its diffusion coefficient $D(r)$ is proportional to r^{-2}), then the mean diffusion coefficient $\bar{D}$ of the population of nuclei is readily calculated from

$$\bar{D}=\frac{\int_{r_{\min}}^{\infty}D(r)\frac{dN}{dr}dr}{\int_{r_{\min}}^{\infty}\frac{dN}{dr}dr}=r_{\min}{}^2D(r_{\min})\frac{\int_{r_{\min}}^{\infty}r^{-\frac{3}{2}k-3}dr}{\int_{r_{\min}}^{\infty}r^{-\frac{3}{2}k-1}dr}$$

$$=\frac{3k}{3k+4}D(r_{\min}),$$

which is the diffusion coefficient of a particle of radius $[(3k+4)/3k]^{\frac{1}{2}}r_{\min}$. For $k\gtrsim0.4$ this would be no greater than $2r_{\min}$, so that an inferred radius down to twice the minimum possible radius is reasonable (for observations show that k is typically of the order of $\frac{1}{2}$). Since the sizes inferred are close to the minimum possible for soluble particles, very little increase of particle size by coagulation with the inactive aerosol particles can have occurred. The rate of growth by coagulation can readily be estimated using the Smoluchowski equation. If an r^{-2} dependence is again assumed for D, and if the total aerosol follows the Junge size distribution (constant mass per log-radius interval) down to a radius $r_{\min}$, then the rate of growth of a particle of radius R is easily calculated. Results for two reasonably representative aerosols are shown in Fig. 3. It is seen, with a moderate concentration of fairly large particles, that $n \sim 10^4$ cm^{-3}, $r \sim 3\times10^{-7}$ cm; these values are typical of what one might expect in unpolluted continental air, where growth from, say, $r=10^{-6}$ to $r=2\times10^{-6}$ cm takes several days. Provided the residence time of the particles is no greater than a few days, the experimental finding of cloud nucleus sizes close to the minimum is reasonable. Measurements of the large-scale distribution of cloud nuclei (Twomey and Wojciechowski, 1969) also suggested a residence time of a few days for cloud nuclei.

5. Conclusions

Filter measurements of the size of natural cloud nuclei yield values close to the theoretical minimum particle size of about 10^{-6} cm permitted by nucleation theory. This leads to the conclusion that these nuclei must be composed of a water-soluble material and that their atmospheric residence time is no more than a few days.

The experiments give some indications that the size distribution of atmospheric cloud nuclei may be bimodal, with the few sodium chloride or sea-salt particles in the larger (of order 10^{-5} cm) size group.

REFERENCES

Danielson, R. E., D. R. Moore and H. C. Van de Hulst, 1969: The transfer of visible radiation through clouds. *J. Atmos. Sci.*, **26**, 1078–1087.

Füchs, N. A., I. B. Stechkina and V. I. Starosselskii, 1962: Determination of particle size distribution in polydisperse aerosols by the diffusion method. *Brit. J. Appl. Phys.*, **13**, 280–281.

Spurny, K. R., J. P. Lodge, E. R. Frank and D. C. Cheesley, 1969: Aerosol filtration by means of Nuclepore filters. Structural and filtration properties. *Environ. Sci. Technol.*, **3**, 452–468.

Twomey, S., 1963: Equations for the decay by diffusion of particles in an aerosol flowing through circular and rectangular channels. *Bull. Obs. Puy de Dôme*, 173–180.

——, 1965: Size measurements of natural cloud nuclei. *J. Rech. Atmos.*, **2**, 113–119.

——, 1971: The composition of cloud nuclei. *J. Atmos. Sci.* **28**, 377–381.

——, and T. A. Wojciechowski, 1969: Observations of the geographic variation of cloud nuclei. *J. Atmos. Sci.*, **26**, 684–688.