Observations of the Geographical Variation of Cloud Nuclei

S. TWOMEY[1] AND T. A. WOJCIECHOWSKI

Naval Research Laboratory, Washington, D. C.

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ABSTRACT

Airborne measurements at flight level of cloud nuclei have been carried out during approximately 100,000 mi of flight over wide areas of the world. A thermal diffusion chamber was used to obtain cloud-nuclei spectra which present the concentration of active nuclei as a function of supersaturation over the range of supersaturation of interest in cloud formation. The results confirm previous conclusions that continental air masses are systematically richer in cloud nuclei and indicate that the median of cloud nuclei over the open ocean is reasonably predictable and varied little from region to region. Over continents the median spectrum does not seem to vary greatly even when North America was compared to Africa and Australia; however, the variability about the median is much greater over land when compared to ocean areas. The results indicate that the lifetime of cloud nuclei, at least over the oceans, is about 3 days. The results require a widespread and relatively uniform source of cloud nuclei both over the oceans and over the land, and it is indicated that the sources of nuclei are not largely dependent on industrial or other man-made pollution.

1. Introduction

During the period 1965–67 a number of long-distance flights were made to sample cloud-nuclei distributions in different parts of the world, with the aim of assembling the rudiments of a climatology of cloud nuclei —which have been found to control to a major extent the number concentration of cloud droplets (Twomey and Squires, 1959; Twomey and Warner, 1967). Indirectly, therefore, these nuclei influence the average size of the cloud drops, the colloidal stability of the clouds, and the optical density of the cloud. The solar radiation reflected by a cloud, for instance, depends not just on cloud thickness and liquid water content, but also on the droplet size and concentration; thus, it also depends on the cloud nuclei content of the air.

Because of their influence in these respects, it is desirable to know what sort of cloud nuclei numbers may be typical of various ocean and land regions. The flight results to be described here represent only a very sparse sampling, but they do provide a picture of how the general levels vary from place to place.

2. Method

The apparatus consisted of a thermal diffusion chamber approximately 7.5 cm diameter and 1 cm high, with appropriate sampling tubes and valves, temperature sensors, etc. It has been described in more detail elsewhere (Squires and Twomey, 1966).

The measurements were made aboard a Super Constellation (EC-121) aircraft operated by the Naval Research Laboratory. In most instances the aircraft flew straight long paths between stops. Since the flight level was dictated to a large extent by operational factors, the sampling achieved was therefore far from random; sampling remote from land tended to be at higher levels, samples over land were at levels dictated by air traffic control, and so on. Samples at or near cloud base, while clearly useful for cloud physics, were a distinct minority even though sought whenever possible. A total of approximately 100,000 mi was flown in obtaining the data discussed below.

[1] Present affiliation: Radiophysics Laboratory, CSIRO, Epping, Australia.

3. Results

The result of an observation normally consisted of a nucleus spectrum, in which the number of activated nuclei was plotted against the supersaturation in the approximate range 0.2–2%. It is obviously out of the question to present the spectra individually; mostly the presentation of results will be confined to median spectra for various regions or situations. When a single number had to be used, the number of nuclei at 0.75% supersaturation was chosen, on the grounds that this is more or less the average upper limit of natural cloud supersaturation, and also a region in which the observational data are believed to be most accurate.

a. Maritime-continental contrast

It is now generally accepted that continental air masses are systematically richer in cloud nuclei (Twomey, 1959a, 1963; Squires and Twomey, 1966; Jiusto, 1966). The present observations amply confirm that conclusion, the medians of *all* continental observa-

tions and of *all* maritime observations being shown in Fig. 1. If one restricted the data to subcloud air samples and to some minimum overland or over-ocean trajectory, the contrast would be greater; this will be seen from some figures in the following sections.

b. Slope of the spectra

The slope of the spectra tended to be higher than earlier observations had suggested. The slope is itself of some importance, since it determines how droplet number and maximum supersaturation vary with updraft speed (Twomey, 1959b). Fig. 2 shows the distribution of the parameter k for all subcloud spectra which could reasonably be described by the expression $N=cS^k$ (N being the total number activated at supersaturation S). The marine data used in producing the figure was restricted to situations where onshore airstreams prevailed, subcloud measurements in mid-ocean being too few to give an adequate sampling.

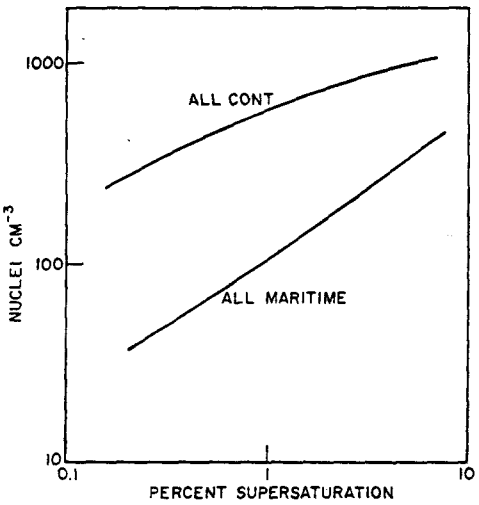

FIG. 1. Median of all observations overland and over the ocean.

To a good approximation the maximum supersaturation during cloud condensation is proportional to the $3/(2k+4)$ power of the updraft speed and the droplet number concentration to the $3k/(2k+4)$ power. From Fig. 2 it follows that the values of these exponents is typically about 0.6 and 0.3 in maritime air (k typically $\sim$0.5) and about 0.5 and 0.4 overland (k of the order of 0.7).

c. Marine atmospheres

Median spectra for several maritime regions for which sufficient data were gathered are shown in Fig. 3. The most notable feature of this figure is the uniformity of the median nucleus spectrum over the various oceans. It suggests an aerosol produced by a widespread and relatively uniform source, rather than a continental aerosol depleted of nuclei during its sojourn over the sea.

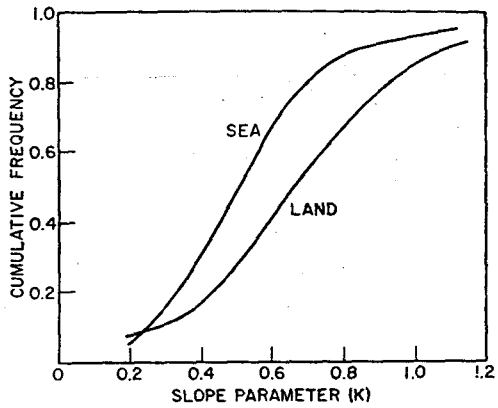

FIG. 2. Distribution of the parameter k (slope of spectrum on a log-log plot).

It is of obvious interest to ascertain how a continental aerosol is changed to a maritime one over the ocean. Unfortunately, reliable trajectory information was not available for the samples collected; some information may be obtained, however, by utilizing the wind at the sampling point, climatological wind data, and synoptic information. This has been done to produce Figs. 4a and 4b which relate to mid-latitude observations and tropical or subtropical observations, respectively (obviously more confidence can be attached to the extratropical data). Both sets of data suggest that the characteristic time for the delay from continental to maritime levels of concentration may be about 3 days.

This estimate of lifetime presumably applies equally to nuclei being produced in the oceanic atmosphere and, in conjunction with the observed median concentrations and height distributions, can be used to estimate the rate of production of maritime cloud nuclei. Height distributions observed over the ocean (see Squires and Twomey, 1966) represent a total nuclei concentration of

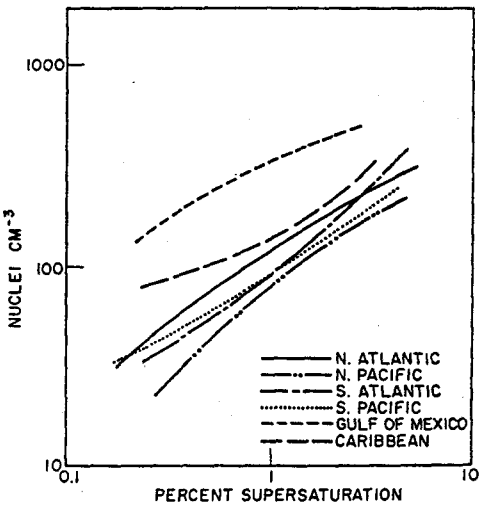

FIG. 3. Medians of maritime subcloud samples for various regions.

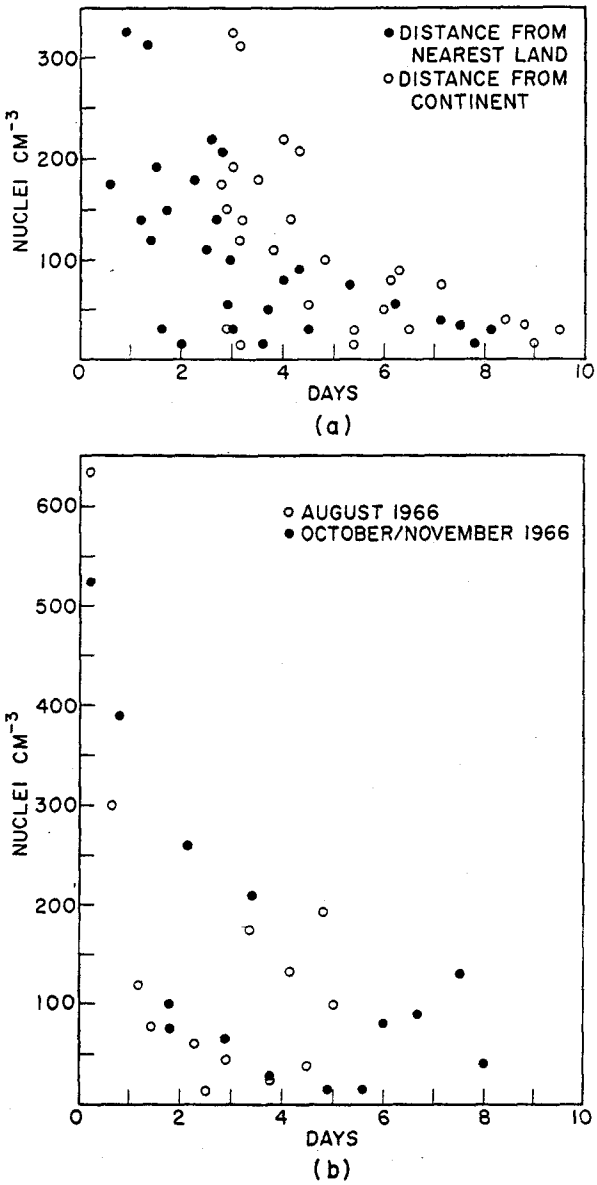

FIG. 4. The decay of nucleus number (for 0.75% supersaturation) in continental air moving out over the ocean: mid-latitude observations (a); tropical or subtropical observations (b).

about 5×10^7 nuclei in an atmospheric column 1 cm^2 in cross section. With a residence time of 2.5×10^5 sec, the nuclei production rate required to maintain that concentration is some 200 cm^{-2} sec^{-1} if they originate at the surface, or 3×10^{-4} cm^{-3} (STP) sec^{-1} if they originate within the atmosphere. Since laboratory experiments show that a few hundred cloud nuclei can be produced by a bursting bubble at the surface of sea water and other aqueous solutions, an oceanic bubbling rate of the order of 1 bubble cm^{-2} sec^{-1} would account for the observed quantities. There is, however, nothing in the observations which demands that the marine nuclei originate at the sea surface rather than within the atmosphere; this contrasts with the situation overland, where the marked decline in concentration with height is strongly suggestive of a surface origin.

d. Continental atmospheres

Because of the greater number of nuclei found overland, and the steeper lapse of cloud nuclei number with height overland, a widespread surface source of nuclei seems to be demanded by the observations. Overland fluctuations around the median were obviously greater and more frequent than was the case in maritime air. To that extent it might be inferred that the main source of nuclei overland was more variable in space or time, or in both. The most obvious possible source is, of course, industrial and other man-made fires, smokes and effluents. However, the observational data does not support the contention that this is the major source, since median nuclei levels were higher for flights across Australia and Africa than for flights across North America (Fig. 5). Furthermore, a division of the North American data into groups, contrasting the western half with the eastern half or the western, central and eastern thirds, showed no significant differences between the groups. Higher continental concentrations tended to occur over arid and semi-arid regions; over eastern North America, modified Gulf air was systematically richer in nuclei content than the cooler drier Canadian air.

The conversion of an air mass from maritime to continental, although of obvious interest, could not be studied in any detail through these observations since the crossing of a coastline by an extensive onshore stream, without the complications of rain, large urban areas and wind shifts, rarely occurred. The best that could be gleaned from the observations was that the addition of nuclei overland was a slow process; 5–6 hr overland is not sufficient to raise the numbers by a factor of 2 (much less than this could not be distinguished from

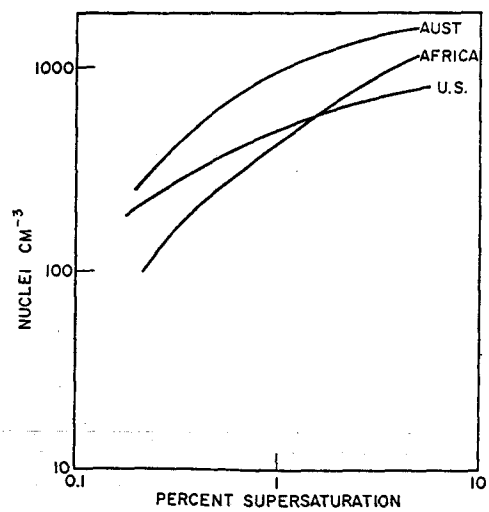

FIG. 5. Medians of continental spectra.

normal fluctuations), and the results generally suggest that several days are required to convert maritime nuclei levels to continental. Much better trajectory information would be needed before anything more specific could be inferred about the process or processes which produce nuclei overland. It may also be noted that the mere crossing of a coastline does not seem to have an appreciable influence on an airmass as a whole, although the surface count may be greatly elevated in areas immediately downwind of the shoreline.

e. Vertical distribution

The flights were not suitable for the best determination of median profiles over land or oceans. The profiles given in Squires and Twomey (1966) represented better samplings than could be obtained during long, essentially point-to-point, flights such as are under discussion here.

It was, however, noticeable well out over the ocean that higher nuclei counts tended to be found above than below clouds. In Fig. 6 median and extreme spectra are shown for flights across the North, Equatorial, and South Atlantic regions in February 1967. The figure utilized only those observations which were made below or above cloud, cloudless or between-cloud situations being eliminated. It shows about twice as many nuclei above cloud as compared with subcloud air. The difference most probably was caused by nothing more than washout of nuclei in the subcloud layer; however, production of nuclei at or above the cloud top level cannot be ruled out entirely since on several occasions extremely high nuclei concentrations were found immediately above cloud tops, the largest being 4300 cm^{-3} at 15,000 ft west of the Marshall Islands in November 1966. Such a count is quite unusual even in continental air, so even undiluted continental air would not be expected to produce so high a count.

f. Removal of nuclei from the atmosphere

In the dry state, cloud nuclei have been found to be a few hundredths of a micron in radius, corresponding to a diffusion coefficient of the order of 10^{-5} cm^2 sec^{-1} (Twomey, 1965). At concentrations of hundreds cm^{-3} the fractional coagulation rate, as given by the Smoluchowski equation, is only of the order of 10^{-7} sec^{-1}, and would have a negligible effect in a day or several days. Coagulation with the more numerous and more mobile Aitken nuclei will be considerably faster, but such encounters will not affect a cloud nucleus unless it can be poisoned by the material of the Aitken nucleus. Apart from that possibility, the lifetime of a few days suggested by the present observations can only be explained by cloud and rain processes or by the actual destruction of the nuclei (by eventual evaporation of the solid nucleus).

Within a cloud, the activated nuclei grow into cloud droplets several microns in radius while the inactivated

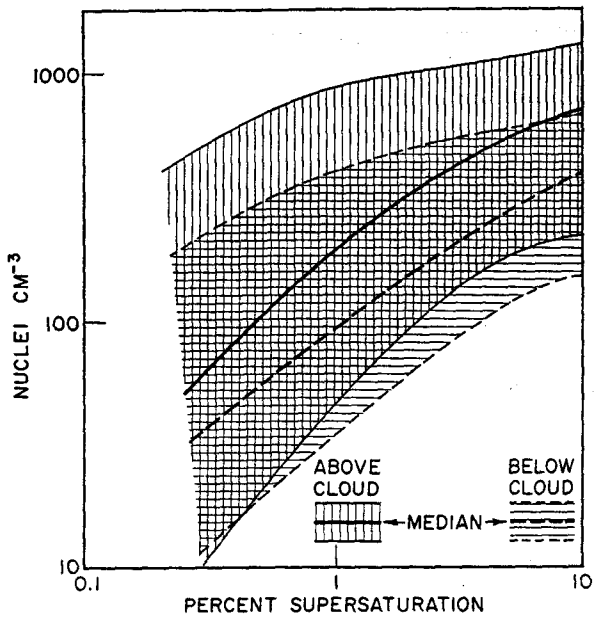

FIG. 6. Contrast of maritime samples below and above cloud.

nuclei remain below the critical radius (approximately 0.1 μ for a 0.75% supersaturation, for instance). The latter have an almost negligible diffusion coefficient and are too small to be collected efficiently by larger droplets falling through them, and will therefore tend to persist while in cloud (this may explain the "elbow" or even S-shaped bend sometimes found in cloud nuclei spectra around 1% supersaturation). Washout by rain will clearly diminish the number of activated nuclei, since each raindrop contains a large number of cloud droplets. If, for example, the average annual rainfall is 100 cm, the cloud liquid water 0.3 gm m^{-3}, and there are N cloud droplets cm^{-3}, then it is readily seen that about 3×10^8N cloud droplets cm^{-2} come to the surface of the earth per year, an average of about $10N$ cm^{-2} sec^{-1}. With a typical marine vertical profile there will be some $5\times10^5\times N$ nuclei in the entire atmospheric column, so that the overall fractional removal rate becomes about 2×10^{-5} sec^{-1}, corresponding to a residence time of the order of a day.

These semi-quantitative arguments suggest that washout is sufficient to account for the observed depletion rate in the case of the nuclei which grow into cloud droplets and become involved in coalescence and rain formation. The removal of the inactivated nuclei, which necessarily remain below, but close to, their critical radius while the air is saturated, is not so readily explained.

g. Variation with latitude and seasons

Insofar as the data could discriminate, no marked seasonal or latitude variations could be detected in the median spectra. It was, however, noticeable that when

unusually low counts (of the order of 10 cm^{-3} or less) did occur, they were found at higher latitudes or in air recently arrived from higher latitudes (i.e., polar maritime or modified polar maritime air).

h. The supersaturation in cloud condensation

Once the nuclei spectrum is known, one can readily derive [precisely by numerical integration or approximately by methods given in Twomey (1959b)] the maximum supersaturation during cloud condensation at prescribed temperatures and updraft speeds. The following data was obtained in this way, using the subcloud spectra of Figs. 3 and 5. The supersaturations in the table

Updraft ($m\ sec^{-1}$)	Typical maximum supersaturations (%)	
	Maritime	Continental
0.3	0.4	0.3
1	0.75	0.4
3	1.25	0.75
10	2.5	1.5

do not differ very much from those derived in the earlier paper cited above. It should be emphasized that these values are for the maximum supersaturation, which is attained during the earliest stage of condensation; thereafter in a steady updraft the supersaturation falls off, so that the average supersaturation will be quite different. The quasi-steady state supersaturation, which is attained when condensation just accommodates the excess vapor supply from the cooling of the air, can be derived from the updraft velocity and the cloud droplet-size distribution, but is typically found to be of the order of hundredths of one per cent or less; the generally prevailing supersaturation, away from cloud base and regions of sizeable acceleration, will tend to be of this order of magnitude.

4. Conclusions

The measurements suggest that the median of cloud nuclei spectra over the open ocean seems to be reasonably predictable and to vary little from region to region. Over continents the median spectrum does not seem to vary greatly even when North America was compared to Africa and Australia; however, the overland variability about the median is much greater.

The results suggest that the lifetime of cloud nuclei, at least over the oceans, is of the order of 3 days. This is somewhat shorter than is believed typical of aerosols generally, the shorter time possibly resulting from the role of these nuclei in the formation of cloud droplets and, eventually, precipitation.

The results demand a widespread and relatively uniform source of cloud nuclei both over the oceans and over the land. The comparison of populous and sparsely populated regions and of the Northern and Southern Hemispheres suggests that the sources of nuclei are not largely dependent on industrial or other man-made pollution.

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