A Comparison of Cloud Nucleus Measurements over Central North America and the Caribbean Sea

P. SQUIRES

National Center for Atmospheric Research, Boulder, Colo.

AND S. TWOMEY

U. S. Naval Research Laboratory, Washington, D. C.

(Manuscript received 18 February 1966)

ABSTRACT

The mean concentration of cloud nuclei over Colorado is found to be about three times that over the Caribbean up to 5000 feet above the surface. The contrast decreases upwards and at 17,000 feet there is no appreciable difference.

1. Introduction

In certain regions, observations of the concentration of droplets in convective clouds have given systematically larger values over continents than over oceans. Squires (1956, 1958) reported median values for the Australian region of 228 and 45 cm^{-3}, respectively. Battan and Reitan (1957) found average values of 200 cm^{-3} over the central United States and 55 cm^{-3} over the ocean near Puerto Rico. Eaton (1963) has reported values of 600 to 1000 cm^{-3} in Colorado and MacCready and Takeuchi (1965) even larger values in Arizona, both semi-arid regions. In the Australian observations, the continental clouds were much more variable than the maritime clouds, and there was a suggestion that the highest concentrations occurred in dry weather, the absolute maximum of 2800 cm^{-3}, for example, occurring under drought conditions, while the lowest continental values were found during a period of wet weather several hundred miles inland. These latter clouds scarcely differed from typical maritime clouds.

It was considered by Squires (1958) that the differences observed by him could be explained only by postulating that continental surfaces generate cloud nuclei, that is, condensation nuclei with critical supersaturations of the order of 1 per cent. Twomey (1959) confirmed this speculation by direct measurements of the spectrum of cloud nuclei, finding, at the surface, but upwind of sources of local contamination, about ten times more nuclei activated at a supersaturation of 1 per cent in continental air than in fresh maritime streams, and even more under drought conditions. He was able to show further that with plausible updraft speeds at cloud base his measured spectra of nuclei would lead to droplet concentrations of the observed magnitude. Working at an inland location, Twomey and Squires (1959) made simultaneous observations of nucleus spectra and droplet concentrations in cumuli. The latter ranged from about 100 to 400 cm^{-3}, and were in reasonable agreement with the values which would be predicted from the nucleus spectra.

Twomey's conclusions regarding the spectra of condensation nuclei were in general agreement with those of Junge (1952) concerning the spectrum of particle sizes of a typical aerosol in oceanic air and over the continent of Europe. However, there is probably no one-to-one correspondence between the critical supersaturation of nuclei, which is the important parameter in cloud physics, and the size of these particles which often consist only partly of soluble material (Junge, 1952). Moreover, Europe is a densely inhabited, highly industrialized and humid area whereas the source region of the Australian continental air masses is very sparsely inhabited with little or no industrial activity and of a semi-arid nature. The general concordance of the measurements in these two areas cannot, therefore, be taken to imply that the same phenomena are involved. Indeed, observations of cloud droplet spectra reported from Europe such as those of Diem (1962, 1963) give little indication of the existence of a systematic difference of the kind discussed here.

It is, therefore, open to question, to what extent the contrast found in the Southern Hemisphere can be identified in the Northern, where the ratio of land to water surface is much larger, and where man's contamination of the air reaches much greater proportions. The purpose of this paper is to compare observations of cloud nucleus spectra over the eastern Caribbean Sea and over the plains of Colorado.

2. Measurements

A thermal diffusion chamber operating as described by Wieland (1956) was used in each series of measurements. The instruments, which differed in many details, are briefly described below.

a. Caribbean measurements. Airborne equipment was employed in which air samples were brought into temperature and pressure equilibrium with the aircraft cabin and then passed into a thermal diffusion chamber where droplets grew on the activated nuclei and were photographed by a motion picture camera. The device was practically equivalent to ground based devices which have been used for a number of years (Twomey, 1963), differing mainly in the much more rigid mounting of the components needed in the aircraft installation.

The cloud chamber was a shallow glass-walled cylinder, three inches in diameter and one-half inch deep, with heavy metal top and base, the latter being cooled thermoelectrically to produce the required supersaturation, and both top and bottom lined with wet filter paper. Droplet concentration was determined by photographing at 90° the droplets contained in a thin, vertical, intensely illuminated region of known volume in the middle of the cloud chamber.

Sampling of the outside air was done by flushing air at a high flow rate through the connecting (stainless steel) tubing and through the conditioning chamber, a shallow one-half liter cylinder with a thin metalized Mylar diaphragm. When an observation was to be taken, the conditioning chamber was closed off from the lower outside pressure and the sample air within the chamber was brought to cabin pressure by bringing the other side of the diaphragm to cabin pressure. After a waiting period of one-half minute or so for temperature equilibrium, sample air was passed into the cloud chamber and the resulting cloud formation photographed. The air flows used were much larger than needed to assure negligible loss of nuclei by diffusion.

Observations were taken at heights from 500 to 17,000 ft over the eastern Caribbean in an area south to southeast of Puerto Rico, during the first two weeks of August 1965. The air was generally maritime in character but, in view of the land-locked nature of the Caribbean, was undoubtedly subject to some recent land influence. Every observation, however, was included in the results. More than sixty spectra were compiled in all, each spectrum containing droplet concentrations at several supersaturations between 0.2 and 2 per cent.

b. Colorado measurements. The majority of the measurements were made on samples obtained in the vicinity of Denver, Colo., but during the summer of 1965, some were taken near Fort Collins, 70 miles to the north. Both places are in the immediate lee of the Rocky Mountains, at similar elevations (about 5000 feet MSL) in gently rolling, semi-arid terrain. A few samples were obtained at various levels in the first 5000 ft above the surface during the latter part of 1964, but the main series of measurements began in March 1965 and continued on the average about once a week until November 1965. A total of some 150 free air spectra were measured. With few exceptions, the samples were taken at 8, 12, 18 and 25 thousand feet MSL.

The sample air flowed at a rate of 5-10 liters sec^{-1} through a tube of total length of about 1.5 m which protruded about 35 cm from the top of the aircraft. The air was stored for later measurement on the ground in a 300-liter Mylar bag, metalized on the inside to form a Faraday cage. The rate of decay of the concentration of nuclei in the air sample in the bag was estimated from experiments in which successive measurements were made on the air contained in a bag. The rate of decay of nucleus concentrations amounted to about 10 per cent per hour for a full bag, and 20 per cent per hour for one which was half-empty. Since after the capture of the sample, the delay to the measurement was rarely as much as an hour and never less than half an hour, the measurements as made were increased by 5 per cent to allow for storage losses. A further correction is described below.

The diffusion chamber was made in two parts. The lower cooler surface was a layer of water about 3 mm deep in a flat bottomed dish of optical grade Plexiglas. A heavy brass dish of similar dimensions, previously warmed by about 5C relative to the water layer, was inverted inside the glass dish, so that the water formed a seal. The inside surface of the brass was covered with black cotton cloth which was kept wet. The chamber so formed ($21\times23\times2.2$ cm) was flushed by a flow of about ten chamber volumes of sample air at a rate of 200 cm^3 sec^{-1} through two sloping inlets in the upper plate, setting up a vigorous horizontal circulation. A horizontal exit tube, 11 cm long, prevented turbulent entrainment of room air into the chamber during the period between stopping the inflow and the decay of the circulation. A pencil of light from a mercury arc source entered through the exit tube and formed an image of an aperture in the center of the chamber. Using light scattered forward and downward at 45° from the horizontal (i.e., through the water layer and Plexiglas bottom), photographs were taken of the cloud so illuminated. In order to increase the volume in sharp focus, (typically about 0.1 cm^3), the film plane was approximately vertical. Some five photographs were taken at intervals of about two seconds, showing the end of the period of droplet growth and the beginning of the decay of the cloud by sedimentation. No correction was applied for the effects of the side walls since they were remote from the region photographed. As the brass top of the chamber cooled, successive samples of air were introduced on the order of one per minute and the resulting clouds photographed. The temperature difference between the brass top and Plexiglas bottom, measured by thermocouples, decayed with a time constant of about 2 min.

c. Comparison of the instruments. A diffusion chamber of the same design as that used in the Caribbean measurements was compared with the one used in Colorado. On nine days samples of surface air were

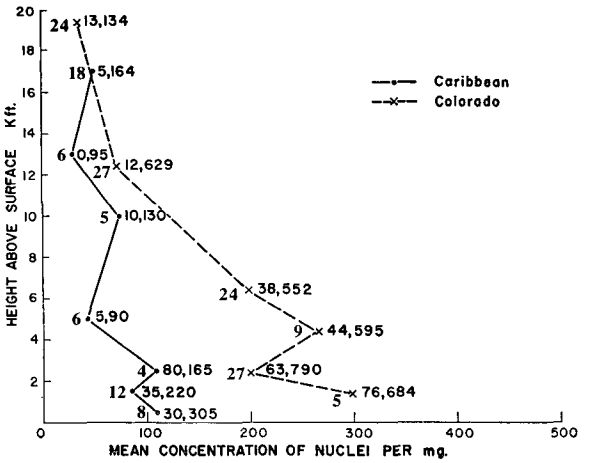

FIG. 1. Mean concentrations, per mg of air, of cloud nuclei activated at a supersaturation of 0.35 per cent over the Caribbean Sea (full lines and dots) and over the plains of Colorado (broken lines and crosses), against height above the surface. The numbers to the left of the plotted points give the number of samples at each level while the minimum and maximum concentrations observed are given to the right of each point.

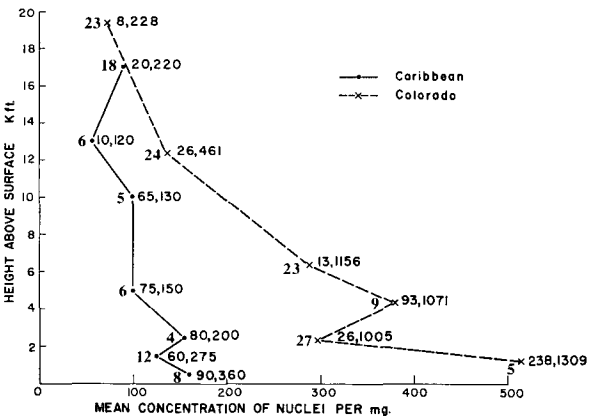

Fig. 2. Same as Fig. 1 except for a 0.75 per cent supersaturation.

stored in the 300-liter Mylar bags, and measured in the two counters. The Colorado instrument gave results which in the mean were 20 per cent higher at supersaturations up to 0.75 per cent. The reason for this discrepancy was not discovered. In order to make the two series of measurements comparable, taking account of the discrepancy found during the comparison of the instruments and of the storage losses which occur in the Colorado measurements (the 5 per cent previously mentioned), the values obtained from the photographs of the clouds in the diffusion chamber in Colorado were reduced by 15 per cent.

3. Results

The mean concentrations of cloud nuclei activated at a supersaturation of 0.35 per cent at various heights in the two areas are compared in Fig. 1. The number of samples and the minimum and maximum concentrations observed are given for each level. A similar comparison is shown in Fig. 2 at a supersaturation of 0.75 per cent. Concentrations have been expressed per milligram of air (1 cm^3 at 20C and 840 mb). Some details of the treatment of the data are given below.

a. Colorado. At times, surface measurements at the airport showed quite high concentrations which appeared to be due to local artificial sources, such as the city of Denver (population 520,000). The free air samples were taken upwind of Denver, but the wind flow in the lower levels in the lee of the mountains is often complex. In order to obtain results which would be representative of the area as a whole, only those values are used in this study which were obtained from samples taken at levels where the possibility of contamination was excluded, either by the structure of the wind with height (for example, winds from north or west at all levels), or by a radiation inversion as shown from the temperature sounding taken from the aircraft. On some occasions when no inversion was present, the height to which surface mixing extended was indicated by cumulus formation, and in such cases samples from higher levels only were included. Altogether, about 25 per cent of the Colorado samples were rejected.

In this way any influence from immediate local sources has been excluded from the Colorado data. Although the sampled air may include contributions from remote centers of population or occasionally additions originating the previous day from local sources, it can be assumed that similar concentrations of anthropogenic nuclei would occur anywhere in the area. The question of the relative importance of natural and cultural sources, although of great interest, is irrelevant to the present comparison.

At all levels, the mean concentrations in the rejected data were higher than those in the data which was retained, the ratio being about 2:1 for the lower layers and decreasing upwards. Surface samples, which often had Aitken counts exceeding 10,000 mg^{-1}, and were certainly at times contaminated, gave average concentrations of cloud nuclei comparable with the maximum values found in the free air samples which were accepted as being free from local effects.

On some days with westerly winds where rather low concentrations were observed in the lower levels in Colorado, it seemed clear that nuclei had been removed from the air as it passed through an extensive (but typically not very deep) sheet of orographic cloud on the western slope of the Rocky Mountains, the existence of which could be inferred from the presence of a föhn wall cloud at the continental divide, visible from the plains. This may be the reason for the anomolous variation of the mean concentrations with height in the first few thousand feet above the surface in Colorado which will be noted in Figs. 1 and 2. A few samples which were taken in stratiform cloud over the

plains in fact gave very low counts; these were excluded as unrepresentative.

The fact that the observations were not simultaneous in the two areas gives rise to some difficulty in interpreting the comparisons of Figs. 1 and 2, as the majority of the Colorado observations were in the period March to November 1965, and those in the Caribbean, in August 1965. However, the indications are that simultaneous observations in sufficient numbers would have given a very similar result since the mean concentration found in all observations at 2400 and 6400 feet above the surface on nine days during the months of July, August and September in Colorado was within 3 per cent of that found in the whole series.

b. Caribbean. The Caribbean data comprised over sixty spectra which were observed at a variety of altitudes. To produce the figures, the numbers were grouped in the following altitude intervals: 500±500 ft; 1500±500 ft; 2500±500 ft; 5000±1000 ft; 10,000 ±2000 ft; 13,000±2000 ft; 17,000±2000 ft. The heights on the figures are the midpoints of the intervals.

4. Conclusion

The data plotted in Figs. 1 and 2 show a clear difference in the cloud nucleus concentrations over the Caribbean Sea and over the plains of Colorado at the same height above the surface. The mean values in Colorado are about three times those over the Caribbean in the lower levels, but the difference decreases with increasing height, as might be expected. At the highest levels observed there is no appreciable difference.

The contrast reported here may perhaps be to some extent an underestimate, because of the cleansing influence of orographic clouds on some of the low-level continental samples, an effect which is no doubt peculiar to the region of the central North American plains which lies close to the Rocky Mountains. As the data stand, however, the contrast between maritime and continental air masses in the North American region does not appear to be as clear-cut as that found in Australia by Twomey (1959). Table 1 compares the mean concentrations found here up to 5000 ft above the surface with those reported by Twomey. The Australian data refer to surface samples, but since maritime air was sampled at the coast and continental air upwind of appreciable sources of local contamination, they are probably comparable with the free air data from North America. The Australian continental samples show higher concentrations than the American, and the maritime samples, lower. These comparisons are based on a quite limited sample, and may well be modified by more extended measurements. However, the difference found in each case is in the sense which might perhaps have been expected. On the one hand, the arid and semi-arid region of North America is not so extensive as that in Australia. On the other hand, it seems possible that the maritime air masses over the Caribbean are subject to a residual influence from land exceeding that in the air masses which were sampled in the Southern Hemisphere. Further measurements, expecially over the open ocean, are already under way to examine this question further.

TABLE 1. Mean concentrations of cloud nuclei in the lower levels of maritime and continental air masses in the North American and Australian regions.

	Concentrations of nuclei per mg activated at supersaturations of:			
	0.35 per cent		0.75 per cent	
	Maritime	Continental	Maritime	Continental
North America (0–5000 ft) present data	87	266	133	401
Australia (surface) after Twomey (1959)	65	415	80	625

Acknowledgments. In the Caribbean, Mr. Theodore A. Wojciechowski made the aircraft observations. In Colorado, Mr. Gary B. Corless acted as flight observer and the measurements in the diffusion chamber were carried out by Mr. William D. Booton. The aircraft were operated by the Naval Research Laboratory and by the Facilities Division of the National Center for Atmospheric Research. The cooperation and assistance of the manager of the Jefferson County Airport, Colorado, is gratefully acknowledged.

REFERENCES

Battan, L. J., and C. H. Reitan, 1957: Droplet size measurements in convective clouds. *Artificial Stimulation of Rain*, New York, Pergamon Press, 184–191.

Diem, M., 1962: Zur Struktur der Wolken. *Arch. Meteor., Geophys., Bioklimat.*, **A13**, 34–56.

——, 1963: Zur Struktur der Wolken, II. *Arch. Meteor., Geophys., Bioklimat.*, **A13**, 461–480.

Eaton, L. R., 1963: Great Plains cumulus cloud droplets. Thesis, Colorado State University.

Junge, C. E., 1952: Die Konstitution des atmosphärischen Aerosols. *Ann. Meteor., Beiheft*, 1–55.

MacCready, P. B., and D. M. Takeuchi, 1965: Precipitation mechanisms and droplet spectra of some convective clouds. Army Report (MR 165 R-243), Analysis of Flagstaff Data, Report No. 2 USAERDL Contract DA 28-043-AMC-00406 (E).

Squires, P., 1956: The micro-structure of cumuli in maritime and continental air. *Tellus*, **8**, 443–444.

——, 1958: The microstructure and colloidal stability of warm clouds. *Tellus*, **10**, 256–271.

Twomey, S., 1959: The nuclei of natural cloud formation. *Geofis. Pura Appl.*, **43**, 227–249.

——, 1963: Measurements of natural cloud nuclei. *J. Rech. Atmospheriques*, **1**, 101–105.

——, and P. Squires, 1959: The influence of cloud nucleus population on the microstructure and stability of convective clouds. *Tellus*, **2**, 408–411.

Wieland, W., 1956: Die Wasserdampfkondensation an natürlichem Aerosol bei geringen Übersättigungen. *Z. Angew. Math. Phys.*, **7**, 428–460.