A STUDY OF SEA-SALT PARTICLES OVER PUERTO RICO

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ABSTRACT

A total of eleven flights was made between 12 December 1953 and 16 April 1954 by Air Force B-17's for the purpose of measuring chloride particle concentrations at various altitudes near Puerto Rico. Further, a number of ground samples was taken. The results of these observations are presented, together with tentative explanations of the variations exhibited by the data.

1. Introduction

The question of the role of sea salt in meteorological phenomena is an old one, and is by no means answered at this time. Kohler (1925; 1936), Cunningham (1941) and others found salt a common contaminant in cloud and fog waters, and numerous investigators identified chlorides in precipitation. It was first suggested that sea salt was the chief contributor to the atmosphere's budget of condensation nyclei. Simpson (1941) pointed out that this called for an improbably large rate of production of salt particles, the more so since he felt that only spray whipped up by a storm, or surf along a coastline, could introduce such particles into the atmosphere. Wright (1940), however, invoked the hypothesis ot Melander (1897) which appeared to show that the simple evaporation of salt solutions introduced salt particles into the air. In this view, the entire sea would be a constant source of such nuclei. Recent work by Lodge *et al* (1954) seems, however, to make the Melander hypothesis quite untenable. Further, the investigations of Kuroiwa (1953) indicate that relatively few cloud particles form on any sort of hygroscopic nuclei. The indications at this time are that nucleation is nonspecific as to the chemical composition of the nuclei.

There remains the theory that large salt nuclei may become the large cloud droplets required for the production of rain from warm clouds. Woodcock (1949; 1950a; 1950b; 1952; 1953) has investigated numerous aspects of this problem and has shown that hygroscopic particles of sufficient size and in adequate numbers to satisfy the requirements of this mechanism are present at cloud levels over the sea.

No exhaustive studies have been made of variations of particle concentration with altitude, meteorological environment, or overland trajectory. To help this situation, the Cloud Physics Project undertook a program of measurements of salt-particle concentration both on the ground and in the air in connection with its flight program in the vicinity of Puerto Rico.

2. Procedures

Two different collection and identification techniques were used. The majority of the samples were collected and analyzed by the impaction method of Seely (1952). Five flights, however, used the Millipore technique of Lodge (1954). All ground collections used the former method.

In both procedures, the final identification hinges upon the formation of a precipitate with mercurous fluosilicate. Hence, for purposes of this article, "sea salt" is defined as any substance which gives a typical halide reaction with this reagent. However, in view of the geographical location of Puerto Rico, it seems unlikely that there can be any significant source of such particles other than the sea. No known man-made sources of halide were near the areas where samples were taken.

The ground-level collections were all made with use

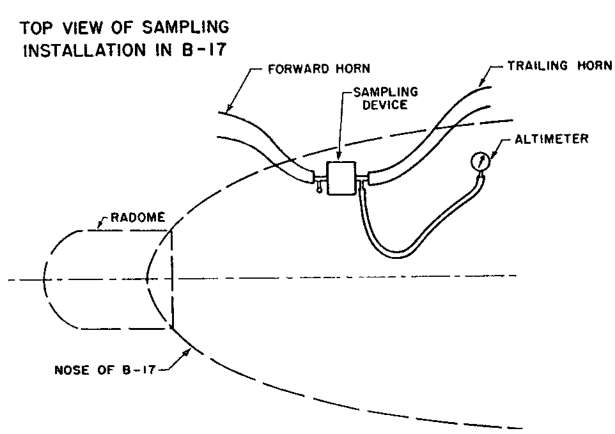

Fig. 1. Arrangement used for sampling from aircraft.

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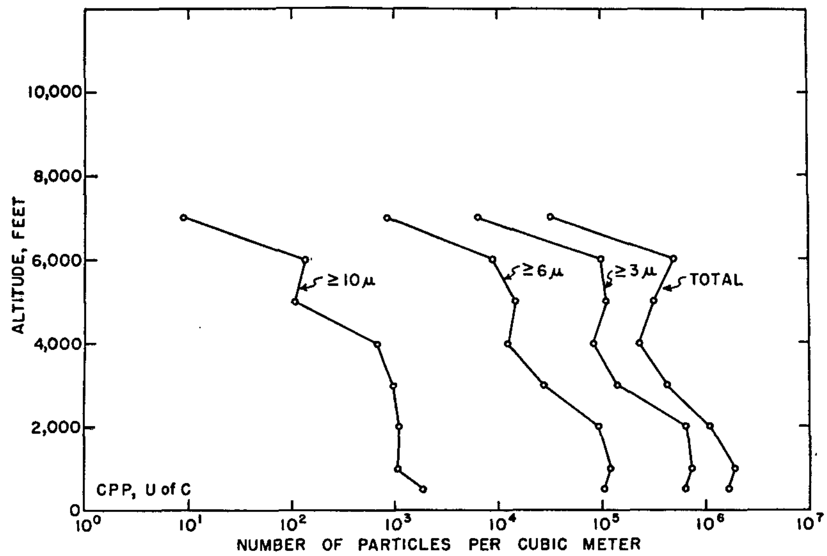

FIG. 2. Flight 10C. Test of oversampling. Data taken in morning, 15 April 1954, on windward side of Puerto Rico, using ¼-in intake. Millipore sample.

of a commercial tank-type vacuum cleaner as the sampling pump. The samples taken aloft involved the arrangement shown in fig. 1, ram pressure and the suction of the trailing tube being used to force air through the collectors — the impactor or the Millipore filter holder. The air-flow rates for the aircraft samples were determined by measuring with an altimeter the pressures ahead of, and behind, the collector. The altimeter readings were converted for this purpose from feet of pressure-altitude to millimeters of mercury by means of the U. S. National Advisory Committee for Aeronautics standard atmosphere, and thence to flow rates according to a previous laboratory calibration. The sampling rate of the impactor–vacuum-cleaner combination was known and assumed constant.

In the impactor samples, no evidence was found of particles smaller than 3 μ equivalent dry diameter. Apparently these either were not collected or faded before they were counted (Pidgeon, 1954). Even some particles larger than 3 μ apparently were not registered, judging by the relative number of particles in the 3- to 6-μ range in the impactor and Millipore collections. For this reason, it is necessary to employ a practical definition of the term "total concentration" as the number of particles *found* per unit volume by the particular analytical method, and to place a similar limit on the meaning of the concentration of particles in any given size interval. On the other hand, it is felt that the values found with the Millipore technique, which is subject neither to fading nor to considerations of collection efficiency, are very near the true values.

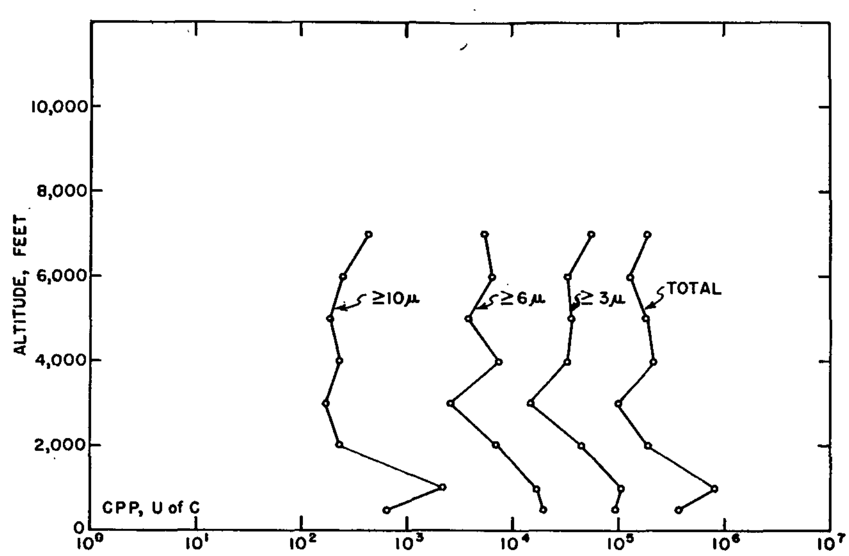

FIG. 3. Flight 11C. Data taken in afternoon, 15 April 1954, on windward side of Puerto Rico, using 4-in intake. Millipore sample.

TABLE 1. Test of oversampling. Ratios of number of particles >10 μ to number of particles <0.7 μ.

	Altitude (ft)							
Flight	500	1000	2000	3000	4000	5000	6000	7000
10C:	0.01026	0.00511	0.00513	0.05783	0.02824	0.00526	0.00270	0.00071
11C:	0.01353	0.01670	0.00605	0.01000	0.00783	0.00667	0.01143	0.01133

The samples collected by the Seely method were all counted as soon as possible after collection — in all cases within 4 hr. The Millipore samples, being far less sensitive to prolonged storage, were returned to the laboratory for processing. In either case, the raw data consisted of the sampling rates, determined as described earlier, the locations and/or altitudes at which the samples were taken, and the counts according to size of the reaction spots. Work in the laboratory, described in a manuscript soon to be published, provided a means of calculating the equivalent dry diameter of the original salt particle from the measured reaction spot-size in both techniques.

In the collections taken aloft, by either method, the intake velocity of the sampler was very much less than the airspeed of the ship. There was, therefore, some concern that oversampling of the larger particles had occurred. In fact, the theory of Langmuir and Blodgett (1946) indicated that 10-μ particles should be oversampled with respect to 3-μ particles by some three orders of magnitude. This possibility was rejected on four counts: first, the uncorrected data accord generally with those of Woodcock in which this airspeed problem did not enter; second, the percentages of 10-μ particles in the total impactor collections on the ground and in the air were the same within statistical error; third, the uncorrected data from the Millipore method showed the expected logarithmico-normal distribution, whereas the data corrected according to the Langmuir theory did not; and fourth, the uncorrected data fit Junge's (1953) distribution law,

$$r^3\, dN/d(\ln r) = k,$$

where N is cumulative total number, r particle radius, and k is a constant.

An independent check of the degree of oversampling may be made by varying the size of the sampling inlet. The flights designated 10C and 11C (figs. 2 and 3) constitute such a test. Both were made the same day, with use of the Millipore method. Flight 11C used the 4-in intake used on all the other flights; on 10C this was replaced by a length of $\frac{1}{4}$-in ID copper tubing.

Using reasonable values for the variables in Langmuir's equations, we find that the oversampling correction for the larger intake for 10-μ particles should be 3×10^{-4}, and for the smaller intake 4×10^{-2}. The theory indicates no oversampling of particles less than 0.8 μ. Hence, the ratio of the number of 10-μ particles to the number of 0.8-μ particles with the large horn should be approximately 100 times that with the small tube. The measure actually employed was the ratio of the number of particles larger than 10 μ to the number smaller than 0.7 μ, since these numbers arose from data already available and should, if anything,

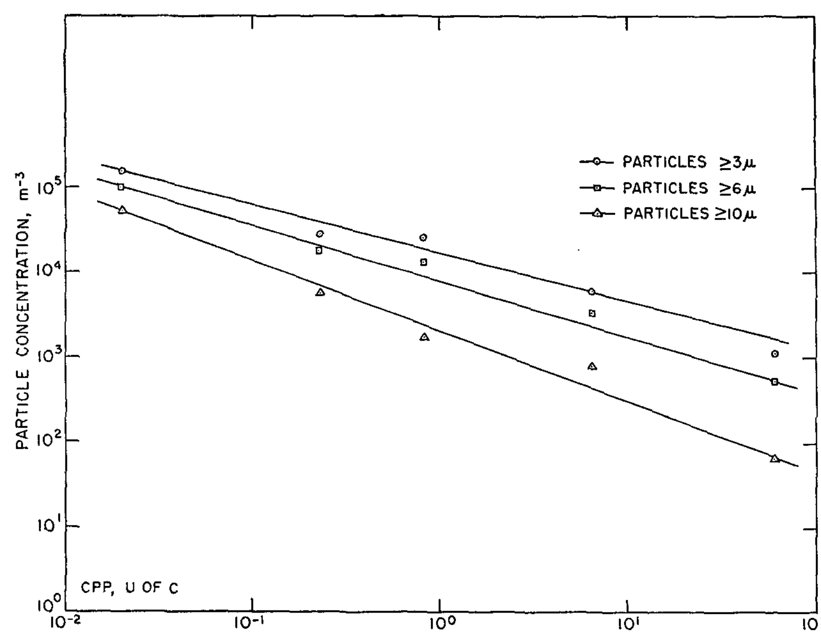

FIG. 4. Variation of chloride particle concentration at ground level with distance inland, 12 to 15 December 1953. Impactor sample.

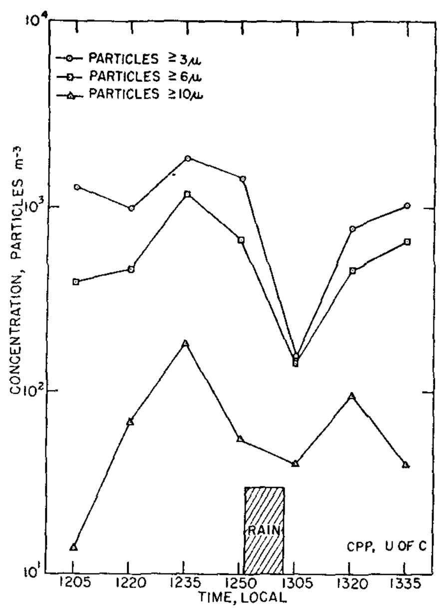

FIG. 5. Time variation of chloride particle concentration at Ponce, 13 December 1953, showing effect of brief rain shower. Impactor sample.

show an even larger effect. The values of these ratios are shown in table 1. The respective mean values are: for flight 10C, 0.0144 ± 0.0195; and for flight 11C, 0.0104 ± 0.0036. Application of the t test to the means gave $t = 0.564$ for 14 degrees of freedom, which shows the difference between the means to be insignificant even at the 50 per-cent level.

Apparently the approximations necessary for the development of Langmuir's formulae lead to erroneous answers when applied to this problem. There may be some slight oversampling, but it is certainly not of the magnitude indicated by Langmuir's analysis. The data are therefore presented here without correction.

3. Ground collections

The data from the ground samplers are summarized in figs. 4 and 5. As shown in fig. 4, there appears to be an orderly decrease in sea-salt concentration with distance inland. The term "distance inland" here means the straight-line distance upwind to the coast. It is realized that this definition is somewhat loose, but it is obviously the only one that can have any numerical meaning with the sparse wind data available. The samples were taken at various sites in and around Ramey Air Force Base, near the northwest tip of Puerto Rico, and at Ponce on the south coast. They were taken at various times during a period of four days of fairly constant meteorological conditions, with the winds rather uniformly between 7 and 10 kn. The number of samples is too small to give certainty to the linear log-log relationship indicated in fig. 4; hence, little quantitative significance can be attached to it. However, it is certain that the qualitative picture given here is essentially correct — a general downward trend, with the largest particles decreasing most rapidly in concentration with increasing distance downwind from the surf.

Fig. 5 shows the variation of the individual samples taken at Ponce and represented in the mean by the points at the far right of fig. 4. The interesting feature is the great decrease in particle concentration immediately following the brief rain shower which occurred between the fourth and fifth samples. This decrease might be ascribed either to washout or to the lower concentration of salt in the air of the downdraft associated with the rain, or to both.

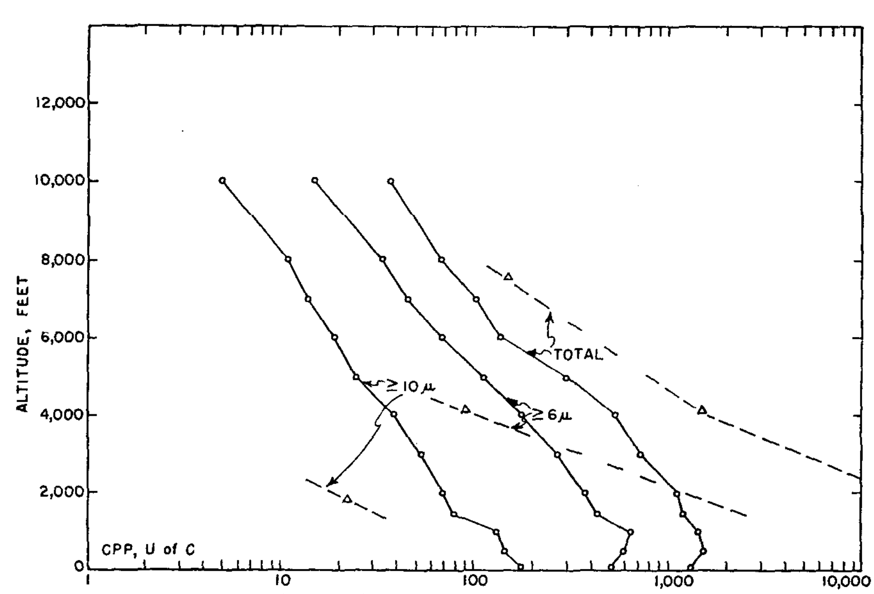

FIG. 6. Mean distribution of chloride particles with altitude on windward side of Puerto Rico, 11 to 15 December 1953. Average of five flights. Dashed line shows data of Woodcock (1953) for Hawaii, wind force 3. Impactor sample.

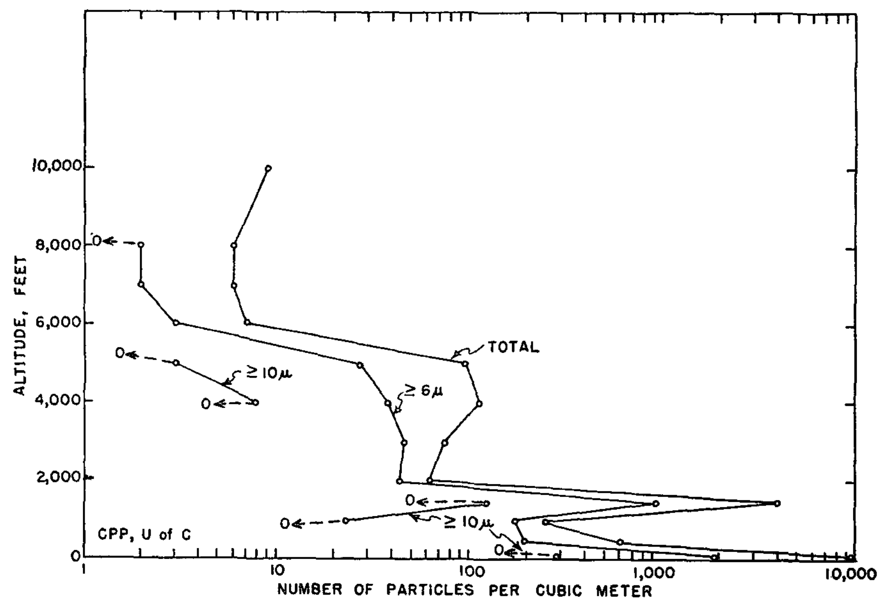

Fig. 7. Distribution of chloride particles with altitude on leeward side of Puerto Rico, 15 December 1953. Impactor sample.

4. Flight collections

The data taken aloft are shown in figs. 2, 3, 6, 7, 8, 9 and 10. Fig. 6, representing the average of five flights over the sea on the windward side of Puerto Rico, exhibits two noteworthy features. The first is the close approach to a Boltzmann distribution of the particles with altitude. The second is the decrease in concentration from 500 to 100 ft, which was found in four of the five flights. This indicates, of course, that the sea is here acting as a net sink; but this is merely a restatement of the problem, not an explanation. It is possible that, with no whitecaps, a turbulent layer some few hundred feet thick may exist over the ocean surface. With no new salt arising locally from the sea, all chlorides measured at the point in question would have to arise from areas of disturbance at a distance. Thus, the local, low-altitude turbulence would result in a net transport of salt downward in this lower layer only.

An alternative explanation has been proposed. It is suggested that the land breeze at night carries out to sea a lower layer depleted in salt by the action of the land, which retains its identity during the greater part of the day, moving back toward the land. A recent flight, not yet completely analyzed, shows this same inversion more than 50 mi from the nearest land, which seems to disprove this idea.

For comparison, the data of Woodcock (1953) for comparable particle sizes over Hawaii at Beaufort wind force 3 are plotted on the same graph. The qualitative agreement appears to be acceptable.

Fig. 7 is the record of a single sampling mission on

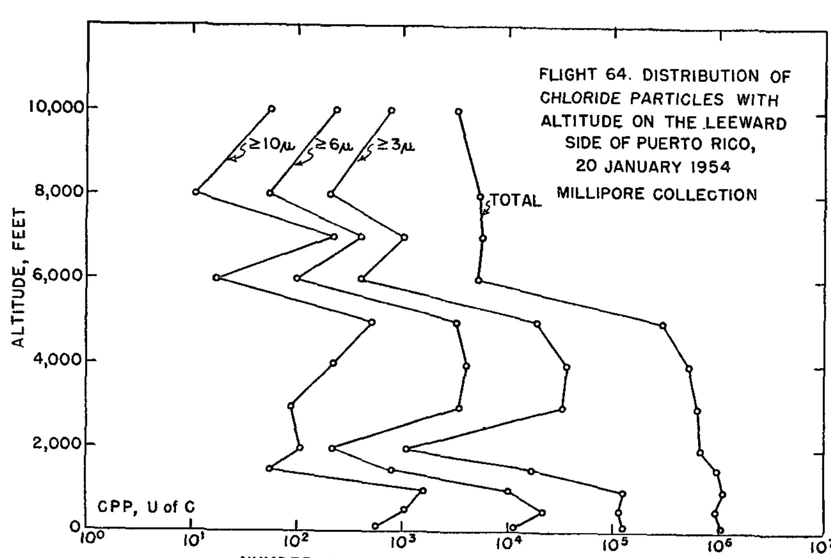

Fig. 8. Distribution of chloride particles with altitude on leeward side of Puerto Rico, 20 January 1954. Millipore sample.

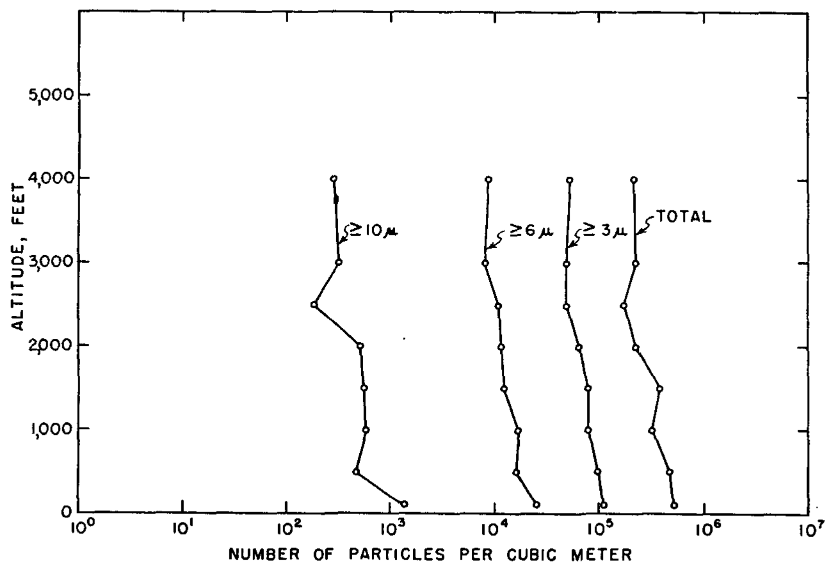

FIG. 9. Distribution of chloride particles with altitude on leeward side of Puerto Rico, 16 April 1954. Millipore sample.

the lee side of the island. The upper portions of the curves are rather typical of any of the individual flights. (The separate data contributing to the averages shown in fig. 6 exhibited similar irregularities, which tend to vanish in the mean, due to daily fluctuations.) In this particular case, the concentration of salt is low, and substantially constant, above cloud top level, which is about 6000 ft.[3] Below this, there is rapid increase. The distinctive features of this collection, as contrasted with samples on the windward side, occur at the lowest levels. These are the two large peak values, one at 1500 ft and the other at 100 ft. Since these samples were taken over the ocean very close to the beach, it seems logical to associate the upper maximum with salt from the surf on the windward side of the island, and the lower with that from the nearby leeward shore. An alternative explanation, that this is a sea-breeze circulation phenomenon, might explain the data in fig. 8, but could hardly account for the lower-level maximum here. The available pibal data for the two days involved do not exclude a lower-level component from the sea, but fail to show the upper return flow at the altitudes required.

Fig. 8 presents data from a similar flight in which the Millipore sampling technique was used. The strong double maximum in the larger particles gives credence to the idea that this is indeed a lee-side flight, although the available wind data do not make it certain that this is the case. At this time the winds were somewhat stronger, about Beaufort force 4 to 5. In this case, the probing aircraft flew somewhat farther out from the shore — some 2 to 4 mi. Hence, the two

[3] The San Juan sounding for 1500 GCT, approximately the time of this flight, showed the trade-wind inversion at 5000 ft.

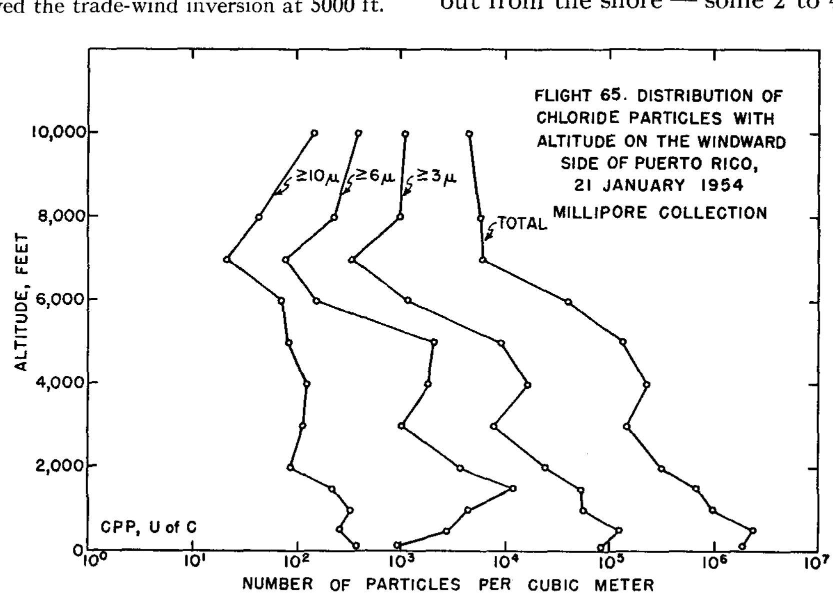

FIG. 10. Distribution of chloride particles with altitude on windward side of Puerto Rico, 21 January 1954. Millipore sample.

maxima are broadened and shifted toward higher altitudes. Cloud tops, as one might expect from the graph, were at 5500 to 6000 ft.[4] The much more efficient sampling of the Millipore method may be readily seen.

A later attempt to study the structure of these maxima (fig. 9) failed to show any maxima at all. Several explanations may be advanced for this. The effect noted in the earlier flights may be purely adventitious. As noted earlier, wind data are sparse here, and the sampling may not have been in the lee of the island at all. Alternatively, the major cause of the phenomenon may be washout of salt in the zone between the two maxima, and there may have been insufficient precipitation on 16 April 1954 to accomplish this.

The results of a Millipore sampling mission on the windward side are shown in fig. 10, which may be compared with fig. 6 to show the relationship between the two sampling methods. The wind force was again 4 to 5. Once more the Boltzmann-type distribution may be noted, as well as the decrease in concentration near the surface. Cloud tops were near 6000 ft,[4] and the slight decrease at 3000 ft may be associated with cloud base.

Figs. 2 and 3, showing flights made primarily for the purpose of checking oversampling, reveal an interesting movement of salt to higher altitudes in the course of a single day, apparently due to development of convection from mid-morning to mid-afternoon.

One interesting feature of some of the Millipore collections is that they show a higher percentage of large particles at high altitudes than at low. No explanation of this phenomenon can be offered, but it appears to be real.

Further experiments are planned in all phases of this work.

5. Summary

Measurements in the vicinity of Puerto Rico have substantiated Woodcock's findings that large sea-salt nuclei occur in sufficient concentrations at cloud level over the ocean to account for the initiation of precipitation. In addition, further information on the vertical distribution of salt particles has been obtained. However, if the concentration of such particles in the free air drops off with distance away from the sea at the rate shown for ground samples, there is grave doubt that such a mechanism can be effective very far inland.

[4] The trade-wind inversion was shown about this level on the San Juan sounding for 0300 GCT, but was not distinguishable at 1500 GCT, which is roughly the time of this flight.

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