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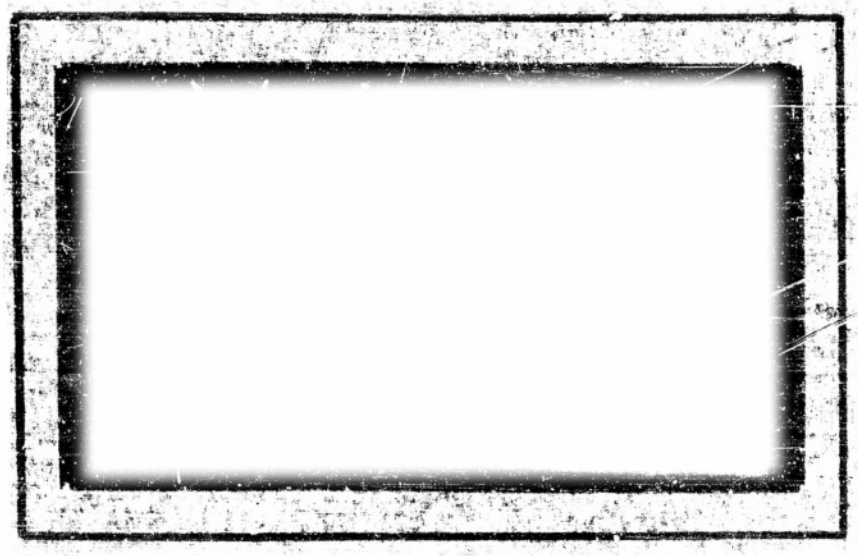

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Reference No. 53-97

SEA SALT NUCLEI STUDIES

"Salt Nuclei in Marine Air as a Function of Altitude and Wind Force"

and

"Remarks on 'Atmospheric Salt Particles and Raindrops'"

by

A. H. Woodcock and James E. MacDonald

Technical Report No. 7
Submitted to Geophysics Branch, Office of Naval Research
Under Contract Nonr-798(00)(NR-085-001)

December 1953

APPROVED FOR DISTRIBUTION _____
Director

Reprinted from JOURNAL OF METEOROLOGY, Vol. 10, No. 5, October, 1953, pp. 362–371
Printed in U. S. A.

SALT NUCLEI IN MARINE AIR AS A FUNCTION OF ALTITUDE AND WIND FORCE

By A. H. Woodcock

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By A. H. Woodcock

Woods Hole Oceanographic Institution [1]

(Manuscript received 24 February 1953)

ABSTRACT

Large differences are shown to occur in the numbers and sizes of sea-salt particles in marine air over the sea as the altitude, position, and the time of sampling are varied. Increases in the amount of air-borne salt near cloud base are related to increases in wind force at the sea surface. The greatest proportionate increase in particle number occurs at the large end of the weight range. Most of the samples reported here were taken near the Hawaiian Islands. The differences in nuclei number and size with increasing altitude in the lower atmosphere are similar in pattern in Hawaii, Florida and South Australia.

It is suggested that bursting air bubbles in "white caps" on the open sea are a major source of the salt nuclei, and that a greater portion of the sea surface may act as a source of these particles during average winds than might be judged from the relatively small area usually covered by white caps.

1. Introduction

Recent developments in the study of rain-forming mechanisms have indicated the importance of very large condensation nuclei in the formation of rain by accretion [2;10]. Large sea-salt nuclei have been found at cloud levels in marine air over land, and their number and weight have been shown to agree in part with the assumption that each salt particle becomes a raindrop [19]. The purpose of the present paper is to indicate some of the great variations which occur in the number and size of large air-borne salt particles at cloud levels in marine air, as a contribution to the detailed study of the role of sea-salt in atmospheric processes. For example, knowledge of the details of these variations is essential to study the relationship of the salt particles in the clear air in the environment of clouds to the precipitation elements within these clouds [14;19].

2. Methods

The salt particles impinge upon and adhere to small glass slides which are exposed from aircraft. The slides are then taken to the laboratory where measurements and counts are made with a microscope of the sizes of individual particles at a relative humidity of 90 per cent, the weight being computed from these size measurements using an isopiestic method [6]. The values for salt-nuclei weight derived in this manner have been tested by direct titration of chlorides. Details concerning sampling methods have been published by Woodcock and Gifford [17] and Woodcock [19], and no further discussion of them will be given here.

3. Increasing wind force and increasing salt content of air

Moore [12] and Fournier [5] have measured an increase in the number and size of large salt particles in the air near the sea surface as the wind speed increases. For cloud physics studies, however, it seems necessary to know the distribution of these particles at cloud heights.

Over the sea in Hawaii, salt-particle samples were taken at cloud levels on thirty-seven days. On twenty-four of these days, most of the range of particle size present on the sampling slides was counted and measured. The average size distributions of the particles in the air on these twenty-four days are given on fig. 1. (On the remaining days only the large nuclei were measured.)

The distribution curves on fig. 1 show that greater wind force in the Hawaiian area is associated with a rather consistent pattern of increase in numbers and sizes of particles near cloud base. It is expected that a comparable pattern of increase will probably be found in most marine atmospheres having a similar fetch, and which also have a similar water-vapor, temperature and velocity distribution throughout the previous several days of movement of the air.

The differences between the values obtained at a given wind force in Hawaii, Australia and Florida, are attributed primarily to differences in the history of the air preceding its arrival at the time and place of sampling, and to errors inherent in estimations of wind force. Note on fig. 6 that the observed force

[1] Contribution No. 643 of the Woods Hole Oceanographic Institution, Woods Hole, Mass. This study was supported by the Office of Naval Research, under Contract Nonr-798(00) (NR-085-001).

3–4 winds are associated with a salt distribution near cloud base (790 m) equal to the average found during a force 5 wind in Hawaii, as shown on fig. 1. Conversely, on fig. 7 note that the observed force 6 wind is associated with a salt distribution curve near cloud base (640 m) which falls between the force 3 and 5 curves on fig. 1. A further discussion of sources of error in relating wind force to salt particle distribution

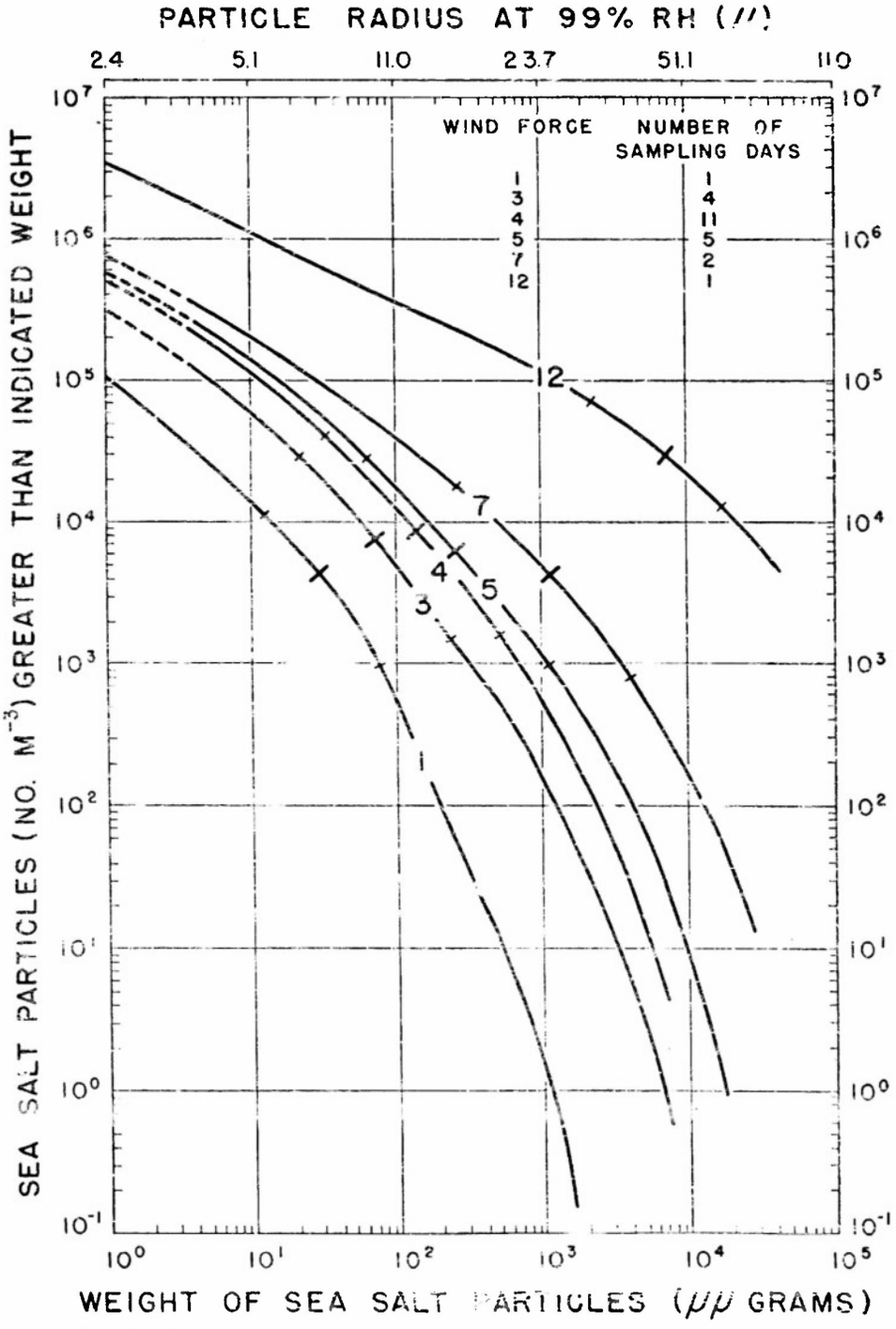

FIG. 1. Effects of varying wind force upon number and weight of large sea-salt particles near cloud base over sea in Hawaii area. Smoothed distribution curves for forces 3, 4, 5 and 7 based upon averaging of results obtained on days when these forces were observed (see legend). Curves for forces 1 and 12 based upon observations for one day only. Three short transverse lines on each curve mark first quartile, median and third quartile weight-distribution points, reading left to right. Distribution curves are read as follows: in force 5 wind, there are about 10,000 particles m^{-3} larger than 170 $\mu\mu$g. Force 12 curve based upon measurements made in Florida within a tropical storm, and included here as indication of probable maximum amount of airborne salt.

seems unnecessary at the present stage of the study.

As indirectly indicated in fig. 1, a great increase is observed in the total amount of sea salt in the air as wind force increases. This is shown by fig. 2, which gives values for all of the thirty-seven sampling days,[2] and also a few measurements obtained in hurricane force winds. Measurements made by Jacobs [7] at La Jolla, California, are also shown on fig. 2. The marked difference in the amounts of salt given by Jacobs and the author is probably due to the fact that Jacobs' samples were taken on the shore at La Jolla behind the surf line, whereas most of the samples reported here were taken near cloud base.

Emphasis should be given to the fact that the individual values for total sea salt on fig. 2 represent, in each case, averages obtained along flight paths thousands of meters in length, and that they cannot be regarded as representing the salt present in any small portion of the paths traversed. As an indication of the variability along flight paths of similar length, seven salt samples were taken on a 65-km horizontal run with constant course in the cloud layer, at an altitude of 1220 m. Each sample represented an averaging in 2700 m of air, with a 7200-m interval between them. Surface winds were force 3. These seven samples showed a variation in total weight of sea salt of from 2×10^{-6} to 4×10^{-6} g m^{-3}. If the sampling had been limited to shorter air paths, a greater variability would probably have been found.

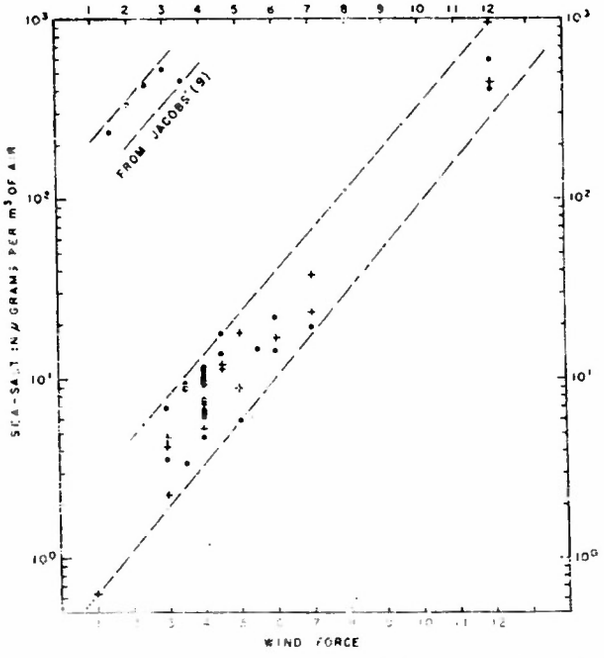

FIG. 2. Variation in total amount of air-borne sea salt as wind force varies. Most of samples taken at cloud levels in Hawaii, June 1951 to July 1952. Upper symbols represent data taken from Jacobs [7]. His measurements were obtained near seashore at Scripps Institute, La Jolla, Calif. Dashed lines are added as aid to note broad trend of observed values.

[2] As previously noted, only the larger nuclei (> 10 to 100 $\mu\mu$g) were measured on some of the sampling days. To save time and effort, these data were not included in deriving the average curves on fig. 1. There is no indication that their inclusion would significantly modify these curves. The addition of the total salt values from these data to fig. 2 is useful, however, and is justified by the fact that the larger nuclei which were measured contain most of the salt (see mass distribution points on fig. 1).

It is expected that further measurements, made during winds in excess of force 7, will reveal the presence at cloud levels of particles as large or larger than 10^5 $\mu\mu$g. At the small end of the weight spectra the curves shown on fig. 1 are ended at about 4 $\mu\mu$g because, on many of the sampling days, the smaller particles on the slides were not counted.

The great increase in the number and size of sea-salt particles at cloud level, which is associated with higher winds, is thought to be due primarily to an increase in the number of bubbles produced in "white caps" at the sea surface. In the laboratory, the bursting of air bubbles (estimated diameters 50 to 500μ) at the surface of sea water is found to project small droplets of various sizes into the air. These droplets contain sea salt which has a weight range comparable to that observed among particles found in the air over the sea. Other laboratory observations concerning this bubble-droplet mechanism have been made in distilled water by Stuhlman [16], and in sea water by Aliverti [1] and Facy [4]. As suggested by the above laboratory tests, the bubble patches which result from breaking waves at the sea surface are found by the author to be sources of miniature clouds of droplets which are rapidly dispersed by the wind. Similar observations of bubbles as a source of nuclei have been made by Boyce [3] in the surf on a Carolina beach. As the foam patches on the open sea become more numerous and greater in extent with higher winds, a larger proportion of the sea surface becomes a source area for particles. Individual foam patches, during a force 5 wind, remain clearly visible from aircraft as turbid or cloudy areas for more than two minutes, indicating the presence of bubbles slowly rising to the surface long after the occurrence of the obvious white cap. Thus a larger proportion of the sea surface is probably a source of particles arising from bursting bubbles than might be judged from the number of readily visible white caps.

As a result of the above preliminary observations, a further study is now being made of the role of bursting bubbles in the formation of salt nuclei in the air overlying the sea.

4. Increasing altitude and decreasing salt

Some indication of the variability of salt content with altitude and with wind force in Hawaii is shown on figs. 3, 4 and 5. In both the subcloud and cloud layers of air there is a general increase in the amount

of salt as the wind increases. At the highest sampling levels, in cloud-free dry air, the quantity of salt is very low and the variations in the amount present seem to be unrelated to surface wind.

The most obvious feature of the distribution curves on figs. 3, 4 and 5 is the rapid decrease of salt with height. On the basis of these and other samples [19], the lower atmosphere in the trades can be divided roughly into three layers: the layer extending from about 50 m above the sea to cloud base, the layer from cloud base up to the trade-wind inversion where the clouds are usually stopped [13], and the air above the inversion. Ascent from one of these layers to the next always reveals a marked decrease in salt. This decrease also appears in samples taken in marine air in Florida and Australia (see figs. 6 and 7). The altitudes of the clouds and the temperature distributions are shown by the insert graphs on each figure.

The environment of cumulus clouds (which develop in the air represented by figs. 3 through 8) is, there-

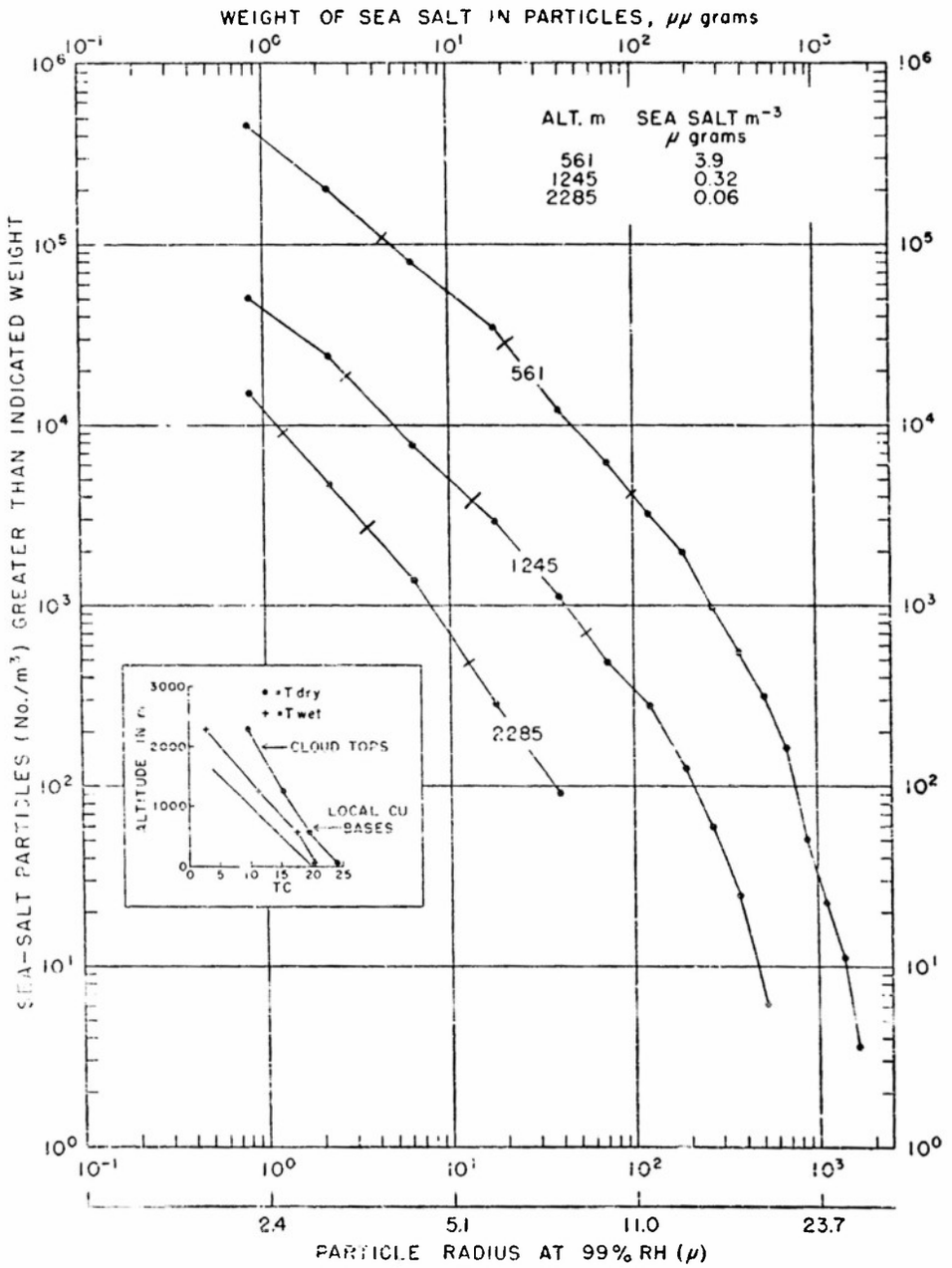

FIG. 3. Vertical distribution of salt particles over sea in Hawaii (21°30'N, 157°40'W), 4 June 1951, 1230 local time. Surface wind 60 deg, force 3. Widely scattered cumulus clouds. See legend to fig. 1.

fore, one of rapid change in the numbers and sizes of salt particles. The clouds may draw much of their air from the subcloud layer, while their tops commonly extend up into relative salt-free air. The number of large salt particles found within these clouds at any given height will depend, in part, upon the relative

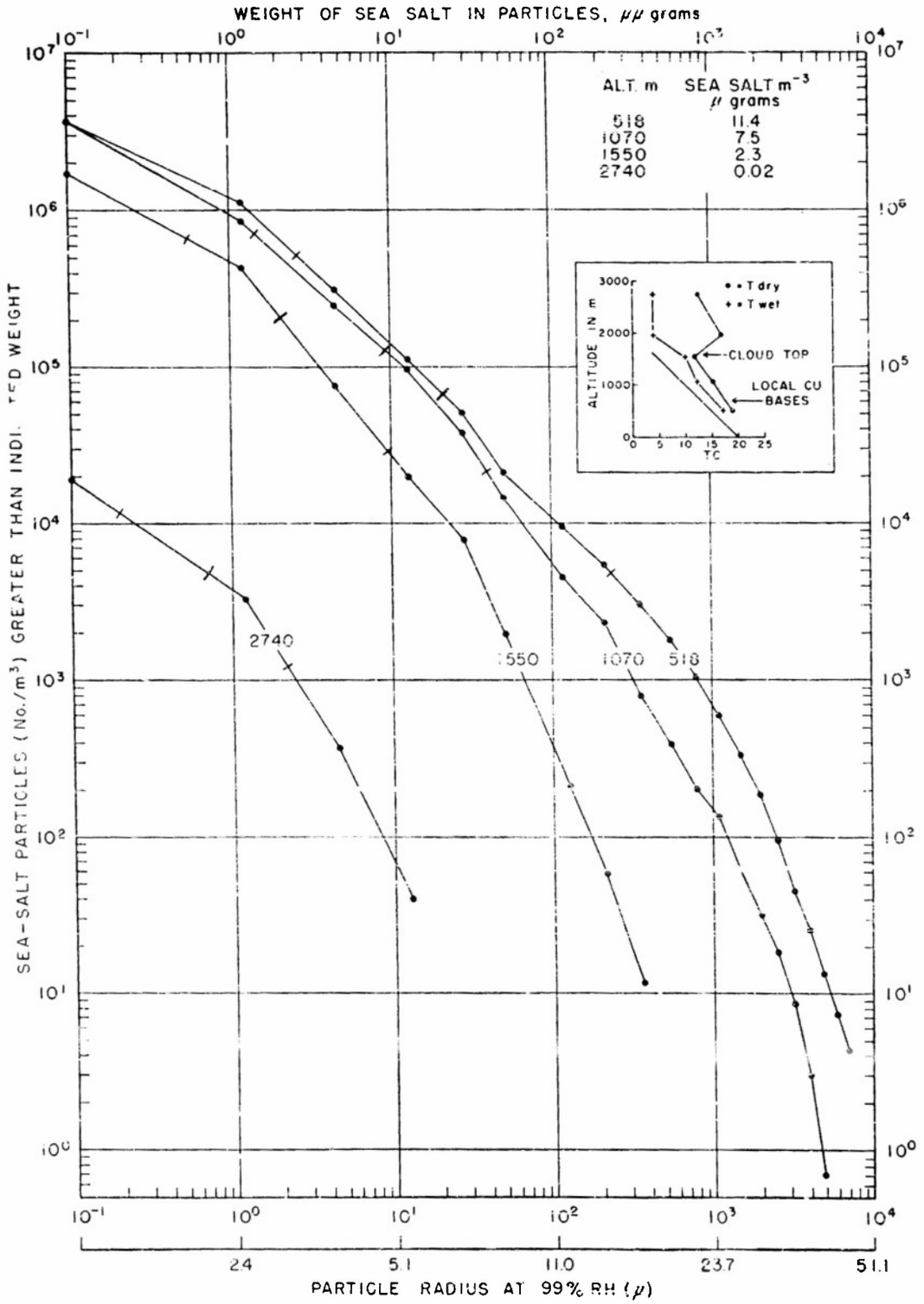

FIG. 4. Vertical distribution of sea-salt particles over sea in Hawaii (21°30'N, 157°40'W), 30 May 1952, 1300 local time. Surface wind 80 deg, force 4 to 5. Height of scattered cumulus cloud bases varied from 640 to 760 m. Temperature at 2740 m, 12.8C. See legend to fig. 1.

proportions of cloud air which are "entrained" as the cloud ascends [11;15]. Thus it may prove to be difficult to assign very definite numbers to the nuclei which should be found within a cumulus cloud at various levels, on the basis of measurements of the nuclei sizes in the clear air near the cloud base.

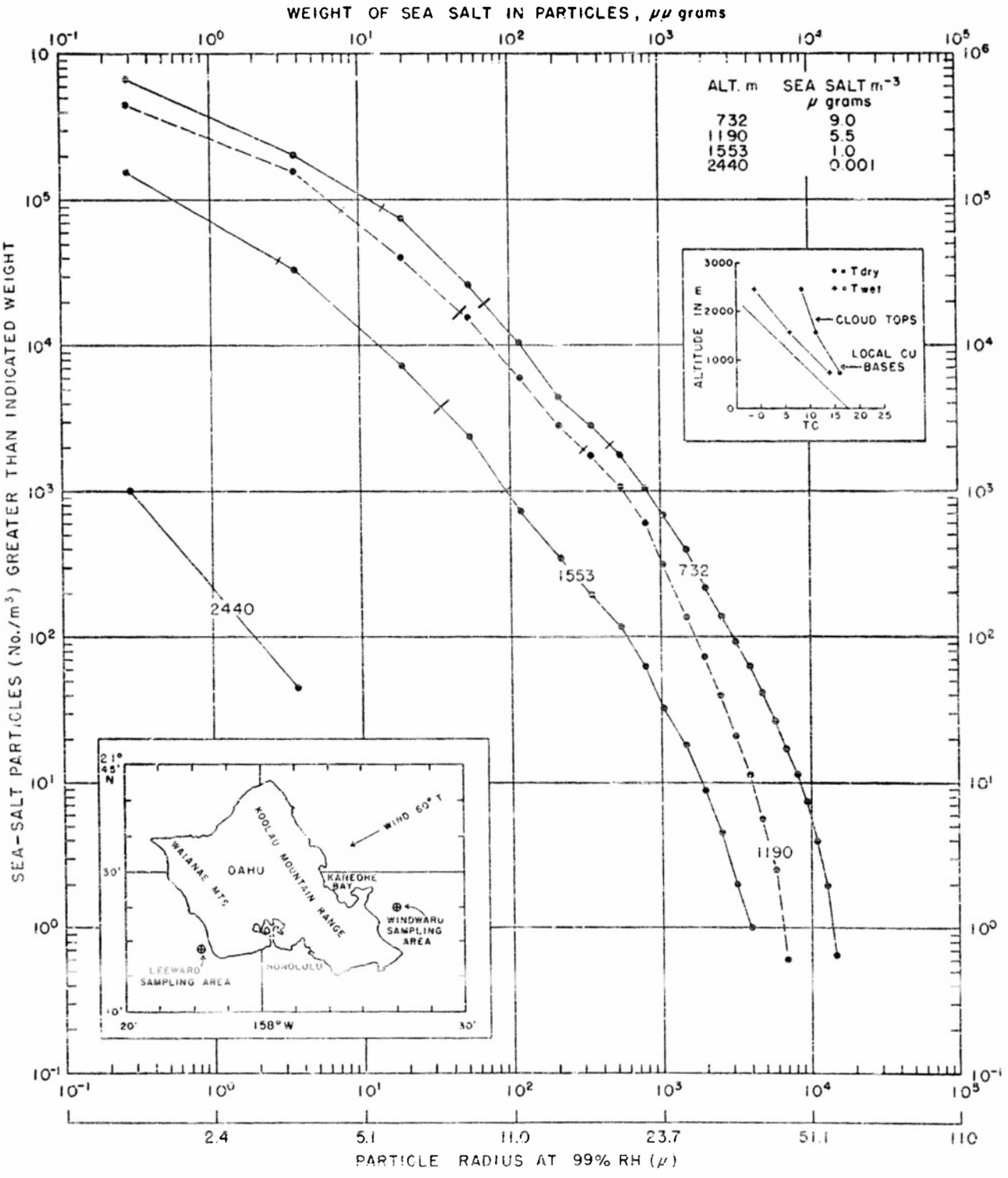

FIG. 5. Vertical distribution of salt particles over sea (21°25'N, 157°40'W) on windward side of Oahu on 22 April 1952, 1140 local time. Surface wind 70 deg, force 5. See legend to fig. 1. Observed points connected by dashed line (1190 m) represent particle distribution on lee side of Oahu (see map), 1500 local time, 22 April 1952.

At present, little is known about the relationship between the numbers of large salt particles in the clear air and the numbers of the sparsely distributed large droplets within the clouds, though a beginning has been made in a recent study by Squires and Woodcock of cumulus and strato-cumulus clouds in maritime air in southern Australia [14]. Many similar observations are needed to enable us to relate the size distribution of the largest cloud droplets in the numerous cloud types to the size distribution of the salt nuclei in the clear air surrounding the cloud.

Most of the studies of salt distribution (figs. 3–8) have been made in regions of large scale divergence and sinking within the atmosphere. Samples taken in the same areas during periods of large scale convergence and ascent should reveal a very different graphical picture of the distribution, the salt aerosols in this case being carried to much higher levels.

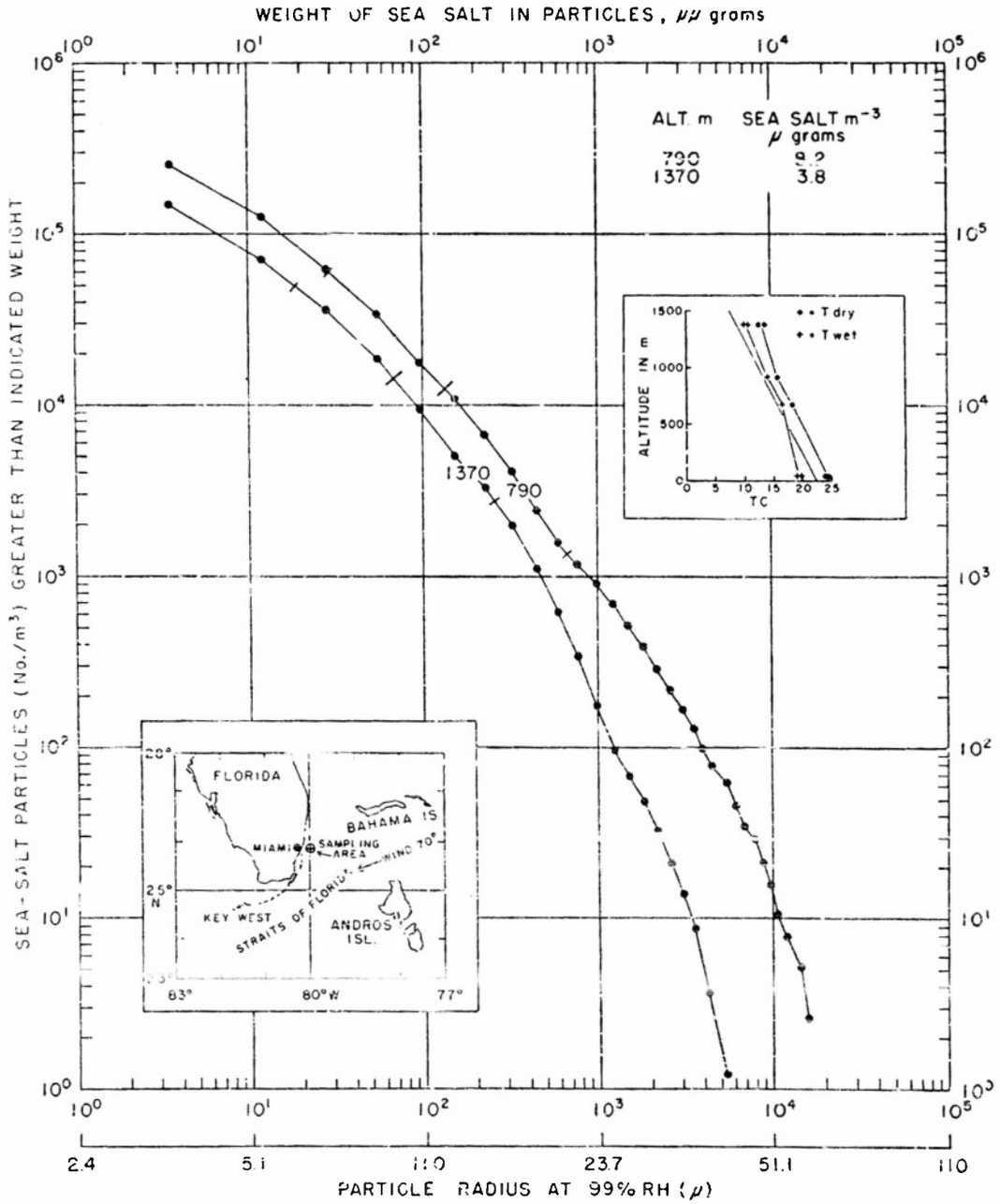

FIG. 6. Average distribution of sea-salt particles in oceanic air over sea east of Florida (see inset map), 8 and 14 November 1951. Surface winds force 3 to 4, 65 deg. Data from 790 m represent samples taken about 100 m below local cumulus clouds, and data from 1370 m represent samples taken in clear air among clouds. See legend to fig. 1.

5. Modification of salt-particle distribution by passage over islands

On 22 April, 1952, samples were taken on the windward and leeward sides of the island of Oahu (see positions on insert map, fig. 5). The continuous lines on fig. 5 show the distribution of particles on the windward side before the air arrived over the island, and the broken line shows the distribution just below cloud base after the air had passed over the island. No profound change has occurred in the numbers of particles as a result of passage over about 32 km of land and over a mountain range having an average altitude of about 600 m. It was observed that no rain was falling in the mountain areas at the time of the lee-side sampling.

On the large island of Hawaii however, a very great decrease was found in the amount of salt near the bases of large cumulus clouds over land, when a com-

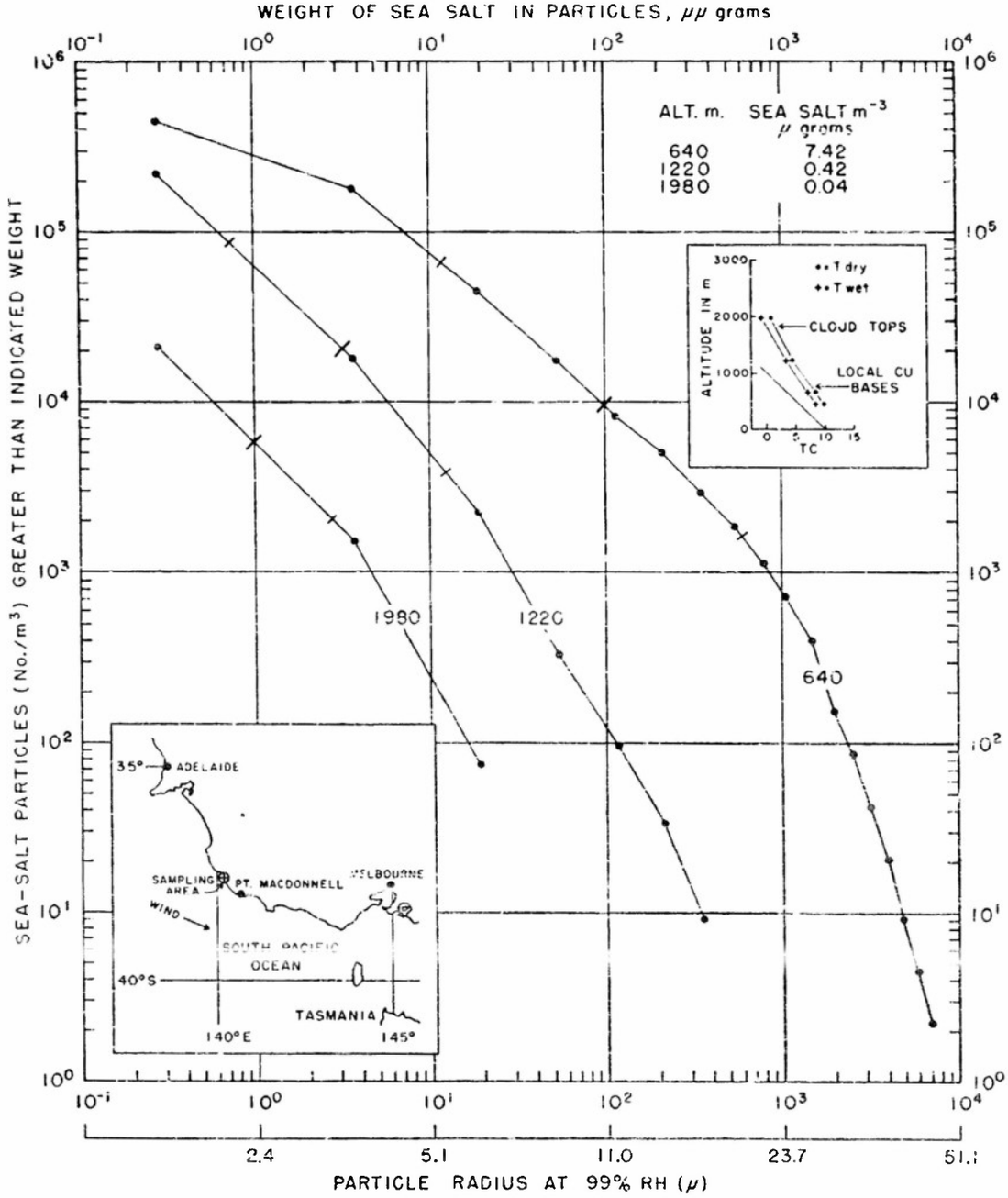

FIG. 7. Vertical distribution of sea-salt particles over Southern Ocean, off coast of S. Australia (37°40'S, 140°12'E), 9 August 1952, 1630 local time. Surface wind 300 deg, force 6. Height of scattered cumulus cloud bases varied from 305 m to 790 m during observation period; tops from 1830 to 1980 m. Rain showers visible during flight. Temperature at 1980 m, 0.5 to 1.4C. See legend to fig. 1.

parison was made with the amount present near cloud base over the windward sea (see comparative distribution curves and sampling positions on fig. 8). The curve representing salt particles over land is, in this case, similar to those derived from samples taken at the highest levels over the sea (see figs. 3

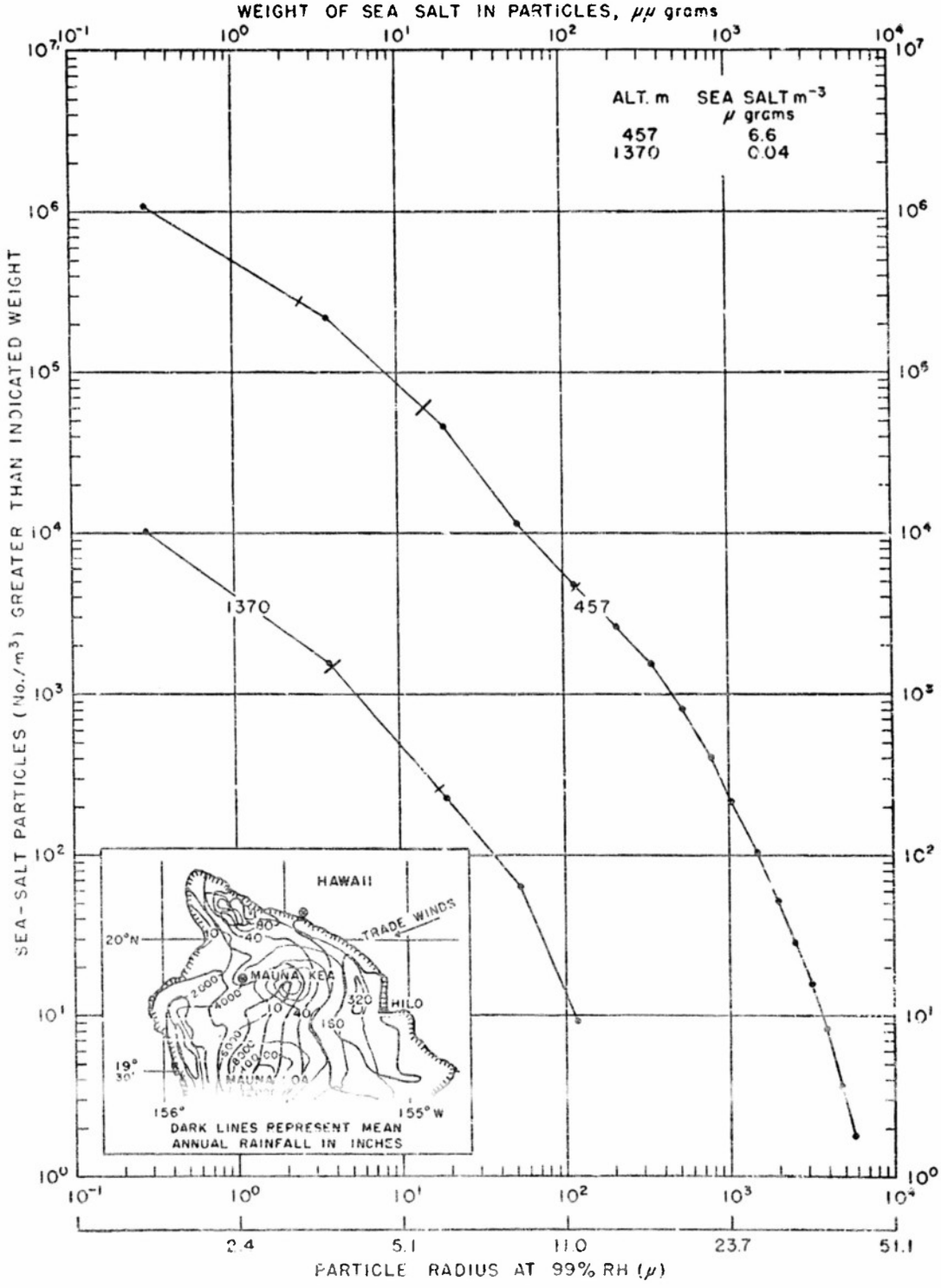

FIG. 8. Comparison of amounts of salt at height of 457 m over sea on windward side of Hawaii, and that over land on lee side of great mountain Mauna Kea at altitude of 1370 m. Sampling positions shown by circled X marks on map. Samples taken on 20 and 21 March 1952, about 100 m below bases of local cumulus clouds. Isohyets on map taken from Leopold [10].

and 4). The great decrease observed over the land area is tentatively attributed to rain-out of the large salt nuclei in the windward high-rainfall areas (see isohyetal lines on map), and perhaps in smaller part to descent of salt-free high level air in a "land-breeze" which, according to Leopold [9], occurs at night on the windward face of the great mountain Mauna Kea.

The presence of well developed cumulus clouds over land in air relatively free from large salt particles is of interest to those concerned with inducing rain by accretion processes. Initially, these clouds will most probably contain very few large droplets, as compared to clouds developed in the air flowing in from the sea on the windward shore (see relative particle sizes on scale at base of diagram, fig. 8). If it is assumed that the larger particles grow to raindrop size more rapidly [8], it is reasonable to suppose that clouds containing smaller salt particles will require more time to produce rain naturally [10]. Exploration of the frequency of occurrence of relatively salt-free and moist air in Hawaii and other regions may prove useful, for the seeding of clouds formed in this air with larger water or salt particles, is more likely to reduce markedly the time required for them to produce rain.

Acknowledgments.—The writer is indebted to Mr. W. A. Mordy of the Pineapple Research Institute, Experiment Station of the Hawaiian Sugar Planters' Association and the Territorial Cattleman's Council, for the opportunity to work in Hawaii, and to Dr. E. G. Bowen of the Commonwealth Scientific and Industrial Research Organization, Sydney, for the opportunity to work in South Australia. The writer is also grateful to Dr. G. Walton Smith for the use of the facilities of the Marine Laboratory, University of Miami, Coral Gables, Florida.

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CORRESPONDENCE

Remarks on "Atmospheric salt particles and raindrops"

BY JAMES E. MCDONALD

Dept. of Physics, Iowa State College, Ames

1 August and 10 October 1952

In a recent paper in the JOURNAL, Woodcock[1] reported the results of a series of measurements of salt-particle sizes at various altitudes above and distances from the sea. The sampling techniques employed were successful in extending the range of collection upward to quite large particle sizes and in establishing that these large particles are carried up to cloud-base levels over and near the sea. In the same paper, Woodcock presents arguments supporting his view that the larger salt particles grow to raindrop size by condensation plus coalescence with smaller drops. I feel that there are two features of the argument which must be questioned.

The first and principal question concerns Woodcock's figs. 6 and 7. It will be seen that fig. 6 constitutes the one check between the salt-particle hypothesis and independent raindrop data, and that this figure depends in turn upon the content of fig. 7, which relates rainfall intensities to rain chlorinities. It seems to be implied that the hypothetical (solid) curves of fig. 6 were computed with use of chlorinities given by curve 1 of fig. 7, but the latter curve is by no means a close representation of the observational points plotted in fig. 7. Inasmuch as it seems improbable that curve 1 could have been obtained by any curve-fitting technique applied to the observed chlorinities of fig. 7, and is presumably not based upon any independent physical hypothesis (since none is mentioned), I am led to ask whether curve 1 was *computed* by starting with some particular points of the solid curves of fig. 6 (such as those at the ordinate of 10^2 drops m^{-3} mm^{-1}, where the solid and dashed curves cross)? If so, the pertinent test of Woodcock's hypothesis would be not the implied one of the agreement between solid and dashed curves of fig. 6, but rather the degree of fit between curve 1 of fig. 7 and the therein plotted points. The strong discrepancy between curve 1 and the datum points weighs heavily against accepting the hypothesis as presented.

A second feature of the argument which I find disturbing is the requirement that the accretional growth of the large salt particles involves coalescence with droplets of virtually zero salinity. Though there is, to be sure, present doubt as to whether sea-salt particles do comprise the primary source of condensation nuclei, it seems inconsistent to assume that the very rare, large salt particles grow readily while the far more numerous small salt particles do not, which is apparently implied by Woodcock. And if the intended implication is rather that the small particles, whose large numbers are so clearly shown by Woodcock's extensive measurements, do grow to cloud-drop size but are still outnumbered by other non-saline drops, it is quite indispensable to an appraisal of the hypothesis to have some estimate of the total weight of salt which might be added by coalescence with smaller saline drops, in order to be sure that the latter do not raise the chlorinity to values inconsistent with the hypothesis. The reason given for not making such an estimate is that one cannot be sure of the weights of salt particles comprising the nuclei in cloud drops, a view which seems incompatible with Mr. Woodcock's willingness to build a hypothesis upon just the upper end of the range of his own numerous salt-particle size measurements.

[1] A. H. Woodcock, "Atmospheric salt particles and raindrops," *J. Meteor.*, 9, 200–212, 1952.

Reply

By A. H. WOODCOCK

Woods Hole Oceanographic Institution, Woods Hole, Mass.

22 September and 31 October 1952

Prof. McDonald is quite right in his emphasis upon the critical importance of the salt-concentration values used (fig. 7) when computing the raindrop size distributions of fig. 6 from the salt-particle distribution (curve 1) of fig. 5. However, in the second paragraph of p. 205, it has been clearly stated that the curves on fig. 7 "represent assumed distributions of chlorides in rains of various intensities." In other words, values of concentration were assumed which, when applied to the salt-particle distributions shown in fig. 5, gave the computed drop distributions shown in fig. 6. The justification for this assumption is that one is thereby able to construct a physically reasonable hypothetical relationship between the observed salt-particle weight distributions at cloud level and raindrop size distributions. On the basis of the results arising from this assumption, "it is suggested that a family of curves similar to those in fig. 7 will eventually be found, each representing rain from an atmosphere containing a characteristic distribution of sea-salt particles" (see p. 205).

As noted on p. 206, "the scatter of the observed points on fig. 7 is attributed to time variations in the salt content of the air and to the collection of other salt particles by accretion as the raindrops fall from cloud to earth." To test the hypothesis that each

large salt particle becomes a raindrop, it has been necessary in subsequent observations to develop new rain-sampling techniques, and to sample exclusively within orographic clouds. This has now been done, and the first tests of the hypothesis are being analyzed.

Prof. McDonald's second point concerns the more numerous small sea-salt particles (weight < about 100 $\mu\mu$g) which are sampled, and their possible effect upon the salt particle–raindrop hypothesis. The number and total mass of these salt particles are not great enough to affect seriously the hypothesis, even if one assumes that all of them coalesce with the larger particles used in deriving the computed drop sizes shown in fig. 6. As shown in table 6, the number of droplets in trade-wind clouds is about 100 times greater than the number of salt particles sampled. These numerous cloud droplets are presumed to be the major source of relatively non-saline water for the accretional growth of the droplets formed on the giant salt nuclei. The weights and the nature of the condensation nuclei on which these cloud droplets form are unknown to me. However, as stated on p. 207, ". . . the quantity of salt present in the larger particles is sufficient within the range of the data presented here to account for all of the salt found in rain and cloud waters." The implication of this fact is that the numerous cloud droplets contain relatively very little sea salt.