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WOODS HOLE OCEANOGRAPHIC INSTITUTION

Woods Hole, Massachusetts

Reference No. 52-75

SEA-SALT NUCLEI STUDIES

Atmospheric Salt Particles and Raindrops

and

Remarks on "Sea Salt in a Tropical Storm"

by

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Director

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ATMOSPHERIC SALT PARTICLES AND RAINDROPS

By Alfred H. Woodcock

ATMOSPHERIC SALT PARTICLES AND RAINDROPS

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Woods Hole Oceanographic Institution[1]

(Original manuscript received 18 May 1951; revised manuscript received 27 December 1951)

ABSTRACT

Marshall and Palmer (1948) have shown that, for rains of a given intensity, there is a definite distribution curve of number of raindrops of a particular range of diameter. The writer has indicated here that the chlorinity of rains also varies with rain intensity. Recently obtained data, concerning atmospheric sea salt, are presented in the form of distribution curves. These curves show the number of sea-salt particles sampled at different altitudes, of a given weight range, plotted against the weight. A computation is made, using a salt-particle distribution curve obtained at cloud levels, in which water is added to each particle until it reaches an assumed chlorinity for a given rain intensity. Each particle is thereby increased in size and becomes a drop of a new weight. The distribution curves of these computed drops are compared to the observed distribution curves of Marshall and Palmer, for various rain intensities, and are found to be remarkably similar. This result implies that, in the process of growth, the droplets containing each salt particle grow to raindrop size through coalescence with much more numerous and relatively non-saline cloud droplets.

The numbers of droplets in cumulus clouds over the sea are compared to the numbers of condensation nuclei in the sub-cloud layer and to the number of larger sea-salt particles.

A method of sampling the large sparsely-distributed salt particles in the atmosphere is described briefly.

1. Introduction

The presence of variable quantities of sea salts in cloud, fog and rain waters is well known (Köhler, 1936; Houghton and Radford, 1938; Takeuchi, 1949; *etc.*) and has led to much discussion of the role of sea-salt particles in rain formation (Simpson, 1941a; Wright, 1940; Köhler, 1941; Findeisen, 1937). Crystalline particles have been sampled in relatively clear marine air at low altitudes by Owens (1926). In clear marine air at cloud levels, Woodcock and Gifford (1949) have sampled salt particles which have a wide range of weights. Thus, salts are found in precipitation elements and also in the relatively clear air in which these elements develop.

Little discussion has been found in the meteorological literature about possible relationships between the number and weight of salt particles in the clear air and the salt in cloud and raindrops. The present paper indicates that a relationship exists between the distribution of weight of salt particles in clear air and the distribution of size of raindrops of varying salt content.

The primary purpose of this paper is to show that large particles of sea salt are present at cloud levels in marine air, and to give evidence that these particles take part in the formation of rain. In this study, three kinds of observational data are used:

1. The weights and numbers of the larger sea-salt particles sampled in the lower atmosphere over the sea and over the land.
2. The chloride content of waters from rains of varying intensity.
3. The size and number of drops in rains of varying intensity.

[1] Contribution No. 563 of the Woods Hole Oceanographic Institution. This study was supported by the Office of Naval Research, under contract number N6onr-277, T.O. II.

TABLE 1. Multi-slide data from air-salt samples taken on a pier at Woods Hole, Mass., 1140 to 1711 EST 15 September 1950. Wind: SW. Visibility: 3 mi. Clouds: cu, 5/10. T: 18.7 to 19.0C; T_w: 18.0 to 18.1C. Sampling height: 5 m above sea surface.

	Sampling strip widths (mm) 1	3	10	30	100	1 (Ag rods)	3
1. Sampling method used	impact	impact	impact	impact	impact	impact	precipitation
2. Slide-exposure duration (sec)	52 (av.)	513	3,750	11,700	18,850	18,850	15,200
3. Average wind speed (m/sec)	9.6	9.1	9.1	8.8	8.9	9.1	9.8
4. Slide-exposure length (m)	500	5000	34,100	103,000	133,400	171,500	—
5. Sea salt cm^{-3} of air ($\mu\mu g$)			8.87			9.21	8.64
6. Number of salt particles cm^{-3} of air			0.35			—	0.66
7. Covering fraction*	.062	.035	.069	.078	.020	—	.0003
8. Number of particles measured			2002			—	1240
9. Method used to measure salt wt.			isopiestic			titration	isopiestic

* See footnote, table 2.

For present purposes, data from category 3 have been taken from the literature. Concerning classes 1 and 2, the observational data available in the literature are inadequate. Hence, new observations of the larger salt particles in the air and of the chloride in rain waters were made and are presented below. The observations of the larger salt particles were made by use of a simple modification of sampling methods already described by Woodcock and Gifford (1949). This modified technique is described at the end of this paper. A standard method was used to determine the chlorides in rain waters. The silver nitrate required to precipitate the chlorides was measured by micro-burette.

2. Vertical distribution of salt particles over the sea

Fig. 1 shows the distribution of sea-salt particles sampled at the end of a dock in Woods Hole, Mass.

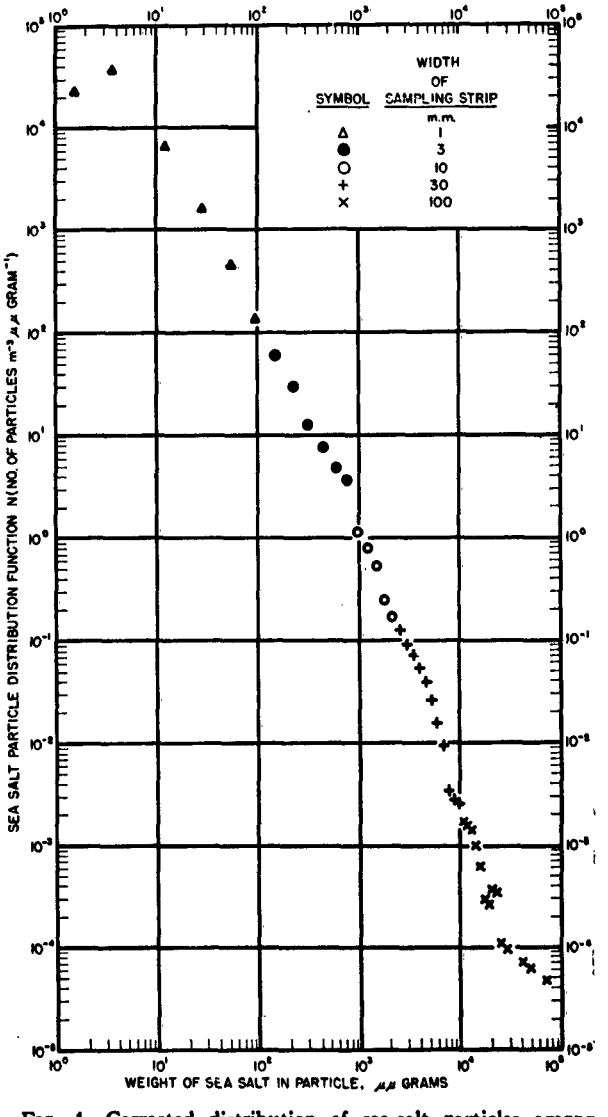

FIG. 1. Corrected distribution of sea-salt particles among samples taken 15 September 1950, 5 m above sea surface. Symbols indicate widths of strips used to sample various particle sizes. See table 1 and text for further discussion.

Table 1 gives other sampling conditions. Previously the largest and most sparsely distributed particles commonly sampled in marine air weighed about 10^{-9} g ($= 10^{3}\mu\mu g$) and were present in numbers of about four m^{-3} of air $\mu\mu g^{-1}$ range in weight. In fig. 1 it can be seen, however, that the new sampling technique extends this limit to about 10^{-4} particles $m^{-3}\mu\mu g^{-1}$ range, at a weight of about $3 \times 10^{4}\mu\mu g$. Questions arose concerning the distribution of these larger nuclei in marine atmospheres at cloud levels.

The distribution of the weight and number of salt nuclei at various altitudes over the sea east of Miami, Fla., is shown on figs. 2 and 3, and tables 2 and 3 show other relevant data. Winds were easterly at all sampling levels. These data show again the very similar distribution of nuclei weight and number at different heights within the sub-cloud layer (see also Woodcock and Gifford, 1949, fig. 15). In addition, the data show that the larger nuclei present near the sea surface are also carried up to cloud levels.

In considering the significance of these large salt

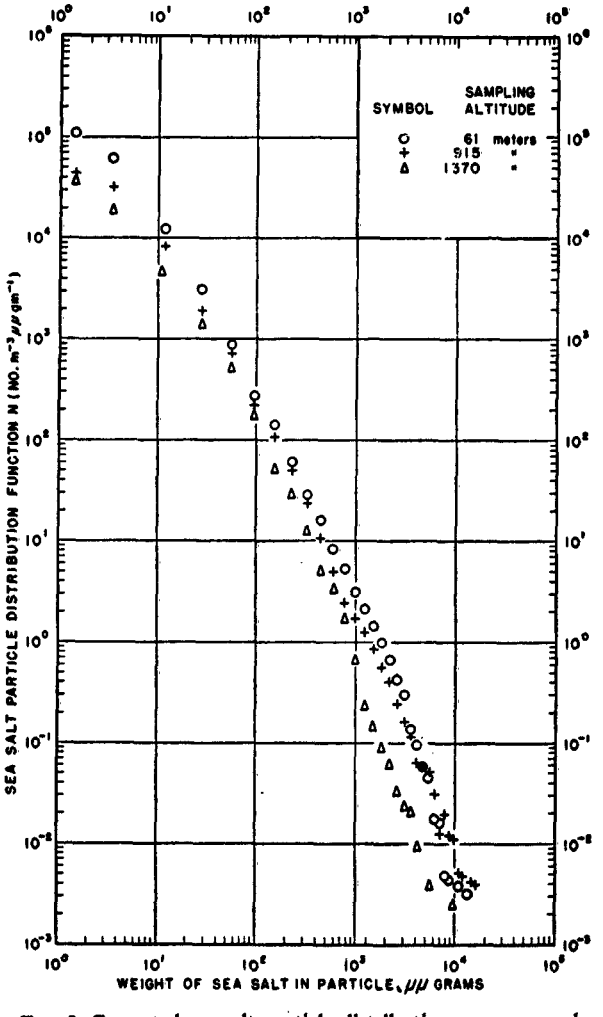

FIG. 2. Corrected sea-salt particle distribution among samples taken in lower atmosphere over sea on 8 November 1950. See table 2 and text for further information.

particles in the formation of rain over land, it is necessary to show that they are present at cloud levels at considerable distances from the sea.

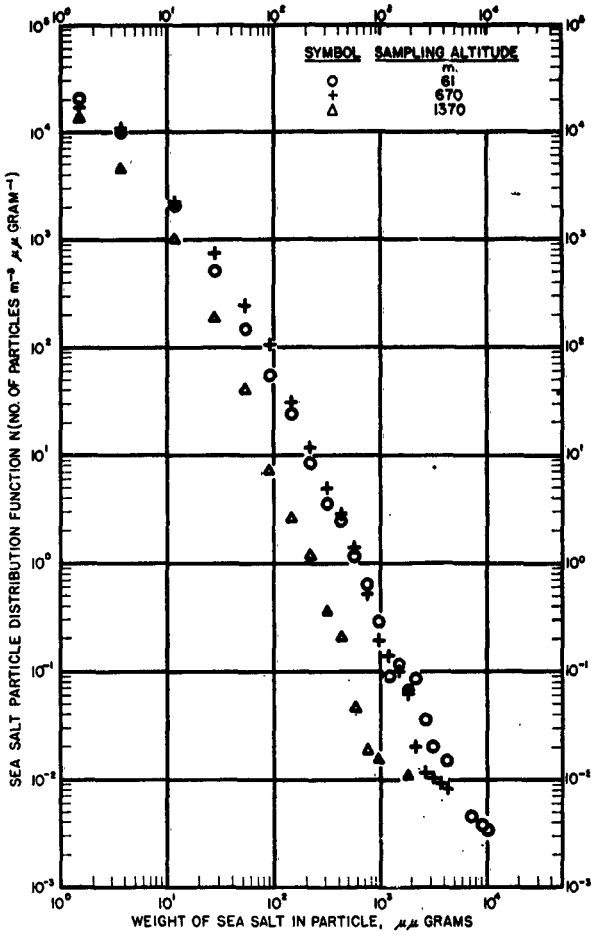

FIG. 3. Corrected distribution of sea-salt particles among samples taken in lower atmosphere over sea on 14 November 1950. For further information, see table 3 and text.

TABLE 2. Data concerning air-salt samples taken over the sea from an airplane near Miami, Fla., 8 November 1950 (lat. 25°51′N, long. 80°00′W). Surface wind: ENE, force 3. Sampling speed: 26.8 m/sec. Scattered cu clouds, bases at 1000 m. Rain showers visible over sea.

	Height (m)		
	61	915	1370
1. Time (EST)	1135	1310	1425
2. Pressure (mb)	1010.5	913	864
3. T (deg C)	24.2	15.9	12.4
4. T_w (deg C)	18.9	14.1	10.0
5. RH (per cent)	61	82	75
6. Sea salt in air ($\mu\mu$g/cm³)	19.3	13.5	6.1
7. Number salt particles cm³	0.94	0.49	0.34
8. Average covering fraction*	.011	.047	.020
9. No. of particles measured	775	777	694

* The covering fraction (Langmuir, 1944) equals a/a_r, where a = area covered by hemispheric droplets on slides, and a_r = total area over which droplet counts were made. Langmuir has indicated that this ratio should not exceed about 0.1. [It should be noted that Woodcock and Gifford (1949) have incorrectly referred to the covering fraction as "per cent of slide surface covered" (p. 189), though the actual values they give are the correct ratios.]

3. Vertical distribution of salt particles over the land

The distribution of salt particles sampled near the town of Everglades, Fla., is given in fig. 4, and table 4 gives other sampling data. The air at all sampling levels at this location had flowed from an easterly

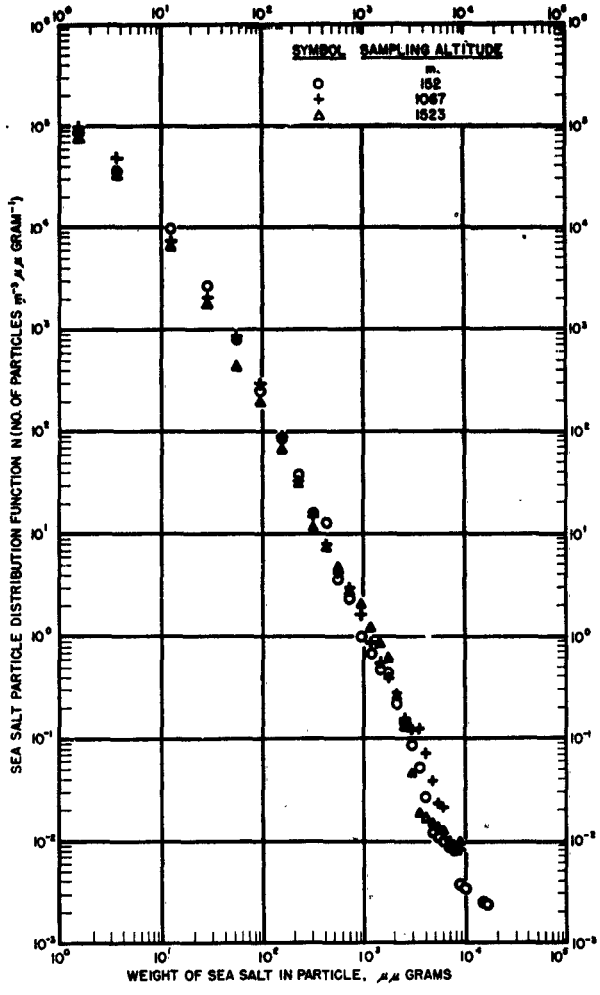

FIG. 4. Corrected distribution of sea-salt particles among samples taken in marine air after passage over about 110 km of land, 16 November 1950. For further information, see table 4 and text.

TABLE 3. Data concerning air-salt samples taken over the sea from an airplane near Miami, Fla., 14 November 1950 (lat. 25°51′N, long. 80°00′W). Surface wind: ENE, force 3 to 4. Scattered cu clouds, bases about 750 m. No rain showers visible during flight. Sampling speed: 26.8 m/sec.

	Height (m)		
	61	670	1370
1. Time (EST)	1055	1200	1316
2. Pressure (mb)	1016.5	946	870
3. T (deg C)	24.1	18.3	13.6
4. T_w (deg C)	19.8	16.6	10.7
5. RH (per cent)	67	85	71
6. Sea salt in air ($\mu\mu$g/cm³)	2.7	5	0.6
7. Number salt particles cm^{-3}	.44	.25	.08
8. Average covering fraction	.002	.008	.0004
9. No. of particles measured	602	493	433

TABLE 4. Data concerning air-salt samples taken over the land from an airplane near Everglades, Fla., 16 November 1950 (lat. 25°53'N, long. 81°21'W). Surface wind: E, 10 mi/hr. Scattered cu clouds, bases about 1150 m. Rain showers visible. Sampling speed: 26.8 m/sec.

	Height (m)		
	152	1067	1523
1. Time (EST)	1425	1530	1636
2. Pressure (mb)	1004.5	900	852
3. T (deg C)	24.3	15.3	11.6
4. T_w (deg C)	18.0	13.9	10.0
5. RH (per cent)	54	86	82
6. Sea salt in air ($\mu\mu g/cm^3$)	11.9	12.3	9.7
7. Number salt particles cm^{-3}	.694	.750	.557
8. Average covering fraction	<.008	<.04	<.03
9. No. of particles measured	716	715	734

direction and had traveled over about 110 km of land since crossing the Florida east coast. Passage over land seems to have thoroughly mixed the particles in the sub-cloud and cloud layers, presumably due to increased atmospheric turbulence. The cloud layer over the open sea usually contains fewer particles than the sub-cloud layer (for example, see fig. 2). Fig. 4 also shows that marine air, after flowing over many miles of land, still contains many large sea-salt particles. No evidence of other hygroscopic nuclei was found on the sampling slides exposed over land.

The presence of these large salt particles at cloud levels over land raised the question of their possible role in the processes of rain formation. This question is discussed below, and it is found that the weights and numbers of these particles, when related to the size and chlorinity of raindrops, are appropriate to and support the assumption that each salt particle becomes a raindrop.

4. Sizes of salt particles as droplets at high relative humidities

The approximate range of weights of the salt particles which have been sampled at cloud levels is shown in the first column of table 5. Column 2 shows the radii of these particles when assumed to be crystalline spheres, and columns 3 to 9 show their radii as liquid spheres having various concentrations of water and sea salt. The increases in radius shown in columns 3, 4 and 5 are observed to occur among atmospheric salt particles measured in the laboratory and are shown in major part by Woodcock and Gifford (figs. 4 and 8, 1949). It is assumed that similar changes in radius would occur with relative-humidity changes in the free air. Columns 6 through 10 give computed radii at decreasing concentrations, which have been measured in fog, cloud and rain waters.[2] Equation (2), below, was used in deriving the drop sizes given.

It is interesting that, within the usual range of salt concentration of rain waters, columns 7 through 10, all the particles become drops having a range of size comparable to that of raindrops (*i.e.*, radius about 150 to 3000μ, as shown in column 10). Expressing this in another way, the evaporation of raindrops having the usual range of salt concentration and size would result in a residue of individual salt particles having a weight range which falls within that actually observed among salt particles found in the air at cloud levels. Column 5 shows that, at the concentration and relative humidity presumed to exist as salt-laden air enters a cumulus cloud, the larger particles have already attained a size ample for growth by accretion[3] (see Langmuir, 1948, table 5, for critical drop-sizes at which accretion can occur).

Thus, table 5 gives evidence that the distribution of weight of the larger salt particles at cloud levels would favor their growth by accretion shortly after entering a cloud (this has been recently discussed by Ludlam, 1951). The table also lends support to the idea that individual salt particles become raindrops, since the distribution of weight among the particles is just appropriate for the formation of drops having rain-

[2] Salt in cloud, fog, and rain waters is usually expressed as weight of Cl. For convenience, these Cl values are, in this paper, converted to weight of sea salt by multiplying by 1.8, the ratio of total salts to Cl in sea water. Miyake (1948) has indicated that this ratio may be too low among salts in precipitation waters, but his results need not be considered here.

[3] Measurements of sea-salt particles suspended on spider webs have shown that a droplet formed on a particle weighing about 10,000$\mu\mu$g will grow to 70 per cent of the equilibrium size (40-μ radius) in about 200 sec, and 80 per cent of equilibrium size in 900 sec, when the relative humidity is rapidly changed from 70 to 98 per cent. These measurements, which were made in the laboratory, are to be extended and will be published later.

TABLE 5. Radii (microns) of sea-salt particles as crystalline spheres (column 2) and liquid spheres at the concentrations found with increasing relative humidity and at the concentrations found in fog, cloud and rain waters.

Weight of sea salt in nucleus	Salt-particle radius (μ) at various concentrations of sea salt and water*								
1	2	3	4	5	6	7	8	9	10
$\mu\mu$g	Crystalline spheres** RH <50%	17.9×10^4 mg/l RH: 90%	35.9×10^3 mg/l RH: 98%	17.8×10^3 mg/l RH: 99%	5×10^2 mg/l	10^2 mg/l	10 mg/l	1 mg/l	10^{-1} mg/l
1	0.5	1.1	1.88	2.4	7.8	13.4	28.8	61.9	134
10	1.0	2.5	4.05	5.1	16.8	28.8	61.9	134	288
10^2	2.2	5.1	8.7	11.0	36.2	61.9	134	288	619
10^3	4.8	11.0	18.8	23.7	78	134	288	619	1340
10^4	10.3	24.7	40.5	51.1	168	288	619	1340	2880
10^5	22.1	51.0	87.2	110	362	619	1340	2880	—

* The radius of some of the smaller droplets will actually be somewhat less, due to curvature effects upon vapor pressure. This fact does not, however, affect the present use of this table.

** Density about 2.2.

drop dimensions when their salt concentration is equal to the salt concentrations found in rain waters. This would mean, of course, that the process of growth of the salt particles to raindrop size would require the addition of relatively pure water. This point will be discussed later.

It has been shown that the weight of the salt particles found at cloud levels is appropriate to the assumption that individual salt particles are the nuclei upon which raindrops form. As indicated in the next section, the number as well as the weight of the salt particles in the air is appropriate and lends support to the idea that the individual particles become raindrops.

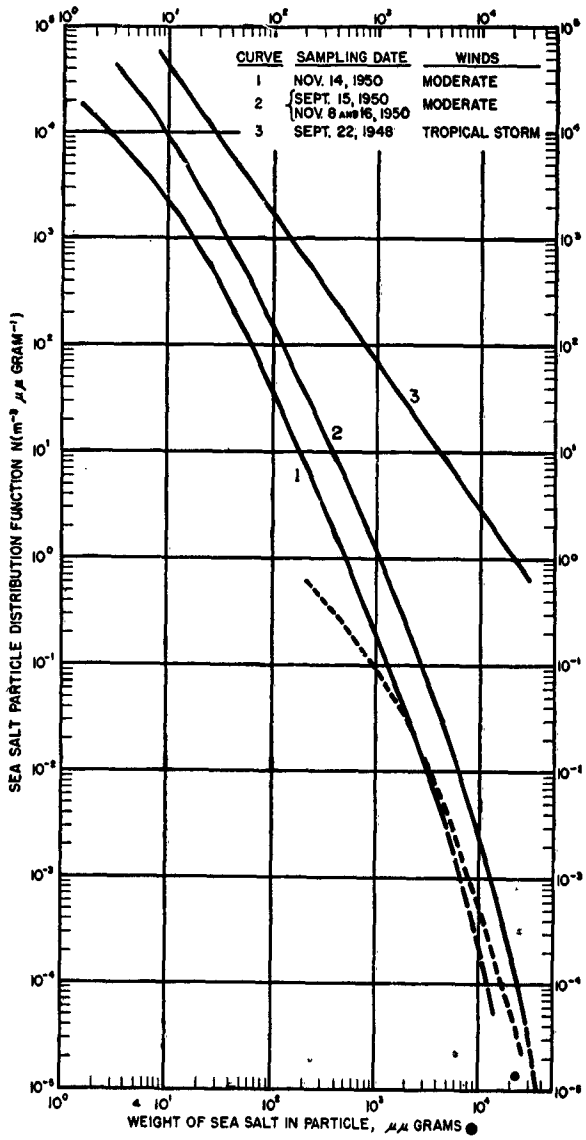

FIG. 5. Smoothed distribution-function curves (1 and 2), taken from observations of sea-salt particle-weight distributions shown in figs. 1–4. Line 3 shows smoothed distribution function of salt particles sampled in tropical storm. Dashed curve shows distribution of salt-particle weight and number, computed from distribution of raindrop number and diameter (fig. 6) and from assumed values for rain-water chlorinity (fig. 7).

5. Correlations of distribution curves for number and weight of salt particles in air, and drop size and salt content of rain waters

Salt particles.—Smoothed distribution curves for the salt particles sampled during moderate and strong winds are shown in fig. 5. Line 3 is based on measurements of particles sampled on 1-mm wide slides in a tropical storm (Woodcock, 1950a), and is included here to suggest the maximum weight and number which may be present in the lower air over the sea. Further measurements are needed during storm conditions, to extend observations to the larger particles. Lines 1 and 2 represent the minimum and average distribution of particles of salt sampled at sub-cloud levels during moderate winds.

Raindrop sizes.—Many observations have been made of the distribution of drop sizes in rains. Fig. 6 is, in part, a reproduction from a study by Marshall and Palmer (1948) and gives an average of the data obtained by these writers and by Laws and Parsons (1943). The dashed lines in this figure represent the distribution of raindrop size at rain intensities of 25, 5 and 1 mm/hr. The unbroken lines represent drop sizes computed from the weights of sea-salt particles and are discussed below.

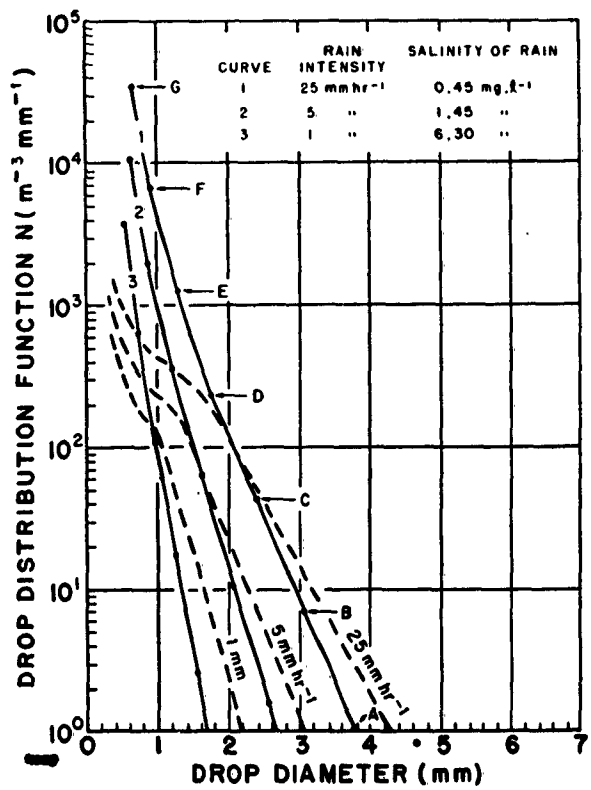

FIG. 6. Dashed lines show variations in distribution of raindrop size at rain intensities of 1, 5 and 25 mm/hr, as given by Marshall and Palmer (1948). Numbered unbroken lines 1, 2 and 3 represent drop-size distribution computed from weights of sea-salt particles given by smoothed distribution-function curve 1 of fig. 5, and from assumed distribution curve 1 for Cl of rains of varying intensity (se fig. 7).

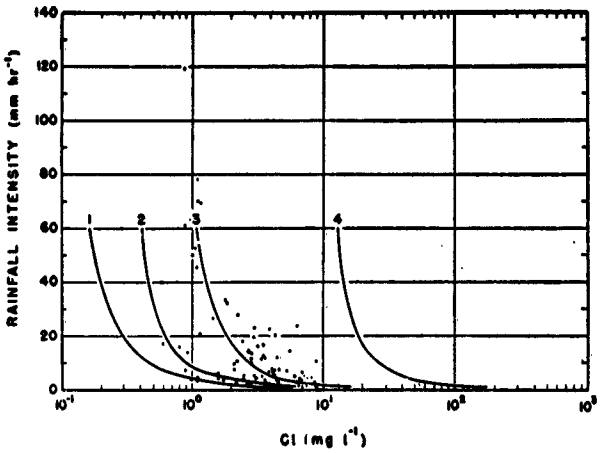

FIG. 7. Black dots represent observed chlorinities in rains of different intensities in southern Florida and Hawaii. Lines 1, 2, 3 and 4 are assumed variations which would be observed if all of chlorides present in rains were derived from larger salt particles found in atmosphere in one of these regions (see fig. 5).

Salt in rainwaters.—Many measurements have been made of the chlorides present in rainwaters, but none of them have clearly related chlorinity to rain intensity. The black dots in fig. 7 represent some measurements of chlorides in rains of various intensities. These preliminary data represent rains from shower clouds in the trade-wind regions of Florida and Hawaii. Many of the rain samples were taken within the clouds, at a height of 900 m, on the northeastern slope of Mt. Haleakala, T. H. A stainless steel funnel of 0.5 m^2 area was used to catch the rain, and sampling durations were from 15 to 300 sec.

A decrease in salt content of rain waters with increased rain intensity, barely suggested by the data in fig. 7, may be due to fundamental processes occurring within the rain-producing clouds. These processes are discussed in the following study of the distribution curves in figs. 5, 6 and 7. In fig. 7, the curved lines represent assumed distributions of chlorides in rains of various intensities. The observed points will be discussed again below

Correlations of salt particles, raindrops and rain salt.—Suppose that all the salt particles weighing $1.3 \times 10^4 \mu\mu g$ (see fig. 5, lower end of line 1)[4] are within a cloud and have become cloud droplets[5] due to condensation processes. Now suppose that more water is added to these droplets, for instance by coalescence with other numerous water droplets, until the concentration of salt in them is equal to that given by curve 1 in fig. 7 for a rate of rain of 25 mm/hr [*i.e.*, about 0.25 mg Cl/l (salinity 0.45 mg/l)]. At this concentration, the droplets become as large as raindrops and are similar in size and number to a value given by Marshall and Palmer (1948) for a rainfall rate of 25 mm/hr (see point A on distribution line 1 of fig. 6). Equation (2), below, was used in computing the drop sizes.

If the salt particles weighing $7 \times 10^3 \mu\mu g$ (see fig. 5, line 1, at the number 1×10^{-3} m^{-3}) become cloud droplets and are similarly diluted, the drop size and the number of these drops again falls near the distribution line given by Marshall and Palmer for a rain rate of 25 mm/hr (see point B, fig. 6). Other nuclei of decreasing weight and increasing number on line 1 (fig. 5) are similarly represented by the letters C, D, E, F and G on fig. 6.

When these salt particles are diluted within a cloud, to the average concentration given by curve 1 in fig. 7 for a rate of rainfall of 5 mm/hr [*i.e.*, a concentration of about 0.8 mg Cl/l (salinity 1.45 mg/l)], the resulting drops have a size distribution represented by the unbroken line 2 in fig. 6. Similarly, unbroken line 3 on this figure represents the sizes of the drops formed on these salt particles at the concentration obtaining in rain waters at a rain intensity of 1 mm/hr [*i.e.*, about 3.5 mg Cl/l (salinity 6.3 mg/l)].[6] If this process is reversed, and the salt particle weights are computed using the raindrop sizes of Marshall and Palmer and the Cl values assumed on curve 1, a distribution of salt particles represented by the dashed line in fig. 5 is obtained.

Thus, the distribution of sea-salt particles sampled in marine air, and represented by line 1 (fig. 5), suggests that the larger of these particles grow within clouds and eventually reach the earth as raindrops. To use line 2 of fig. 5 in a similar way, and to arrive at a drop-size distribution like that given by Marshall and Palmer, it is necessary to assume the higher concentrations shown by line 2 in fig. 7. Concerning curve 3 of fig. 5, a reasonable extrapolation in the direction of larger size indicates the presence in hurricane winds of 10^{-6} particles $m^{-3}\mu\mu g^{-1}$ as heavy as 2×10^{-6} g. This extrapolation suggests that the relationship of Cl and intensity in tropical-storm rains is approximately that of curve 4, fig. 7, assuming again rain-drop sizes near those of Marshall and Palmer. Curve 3 of fig. 7 would similarly represent rains from an atmosphere containing a distribution of salt particles which would fall between curves 2 and 3, fig. 5.

From the above results, it is suggested that a family of curves similar to those in fig. 7 eventually will be found, each representing rain from an atmosphere containing a characteristic distribution of sea-salt particles.[7]

[4] The extrapolation of lines 1 and 2 on fig. 5 in the direction of greater particle weight is a reasonable one, based upon the sampling experience discussed in section 9, below.

[5] No samples were taken within the clouds, because of the great difficulty of sampling sparsely distributed droplets in the presence of many other droplets.

[6] It may be significant that most of the range of concentration of sea salts and water necessarily assumed here (*i.e.*, about 0.5 to 6.3 mg/l) is similar to that used by Schaefer (1950) in obtaining large electrical potentials in the freezing of diluted sea-water. This electrical effect is suggested as a mechanism for the generation of thunderstorm electricity (Workman and Reynolds, 1948).

[7] In Japan, Takeuchi (1949) has found that rains from air masses of different origin contain greatly varying quantities of Cl.

The scatter of the observed points in 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 (see Miyake and Suguira, 1950). The writer is now prepared to measure from aircraft the three quantities, salt-particle size, raindrop size and rainwater chlorinity, near and under shower-producing clouds over the sea. Measurements so made are expected to eliminate much of the error in correlating the above interdependent quantities.

6. Discussion

The indicated distribution of raindrops computed from salt particles and those observed by Marshall and Palmer shows a wide divergence of number among the smaller diameter drops. This is assumed to be due, in part, to a low collection efficiency among the smaller particles as droplets entering a cloud, hence a decreased chance that they may grow to raindrop size by coalescence. This divergence may also be due to losses of the droplets formed on the smaller particles within clouds by coalescence with the larger droplets formed on the larger particles, or to errors in sampling the small raindrops.

In fig. 6, the divergence of the distribution of number of rain and salt drops occurs between the values 10^2 and 10^3 drops m^{-3} mm^{-1}. In fig. 5, the curve between 10^{-4} and 10^{-2} $m^{-3}\mu\mu g^{-1}$ represents salt particles weighing 13,000 to 3300$\mu\mu$g (see line 1). These particles will, shortly after entering a cloud as droplets, have radii ranging from 28 to 44μ and will fall fast enough to begin to grow by accretion in descending through a cloud containing droplets of the usual size-range (see Langmuir, 1948, p. 182). Other drops, represented by the curve in fig. 6 between about 2×10^2 and 3×10^4 m^{-3} mm^{-1}, were formed on salt particles weighing 1300 to 60$\mu\mu$g (see fig. 5, line 1). These particles, shortly after entering a cloud as droplets, will have radii ranging from 20 to 7μ and are less likely to grow significantly by accretion because their "collection efficiency" is low. In fact, many of them

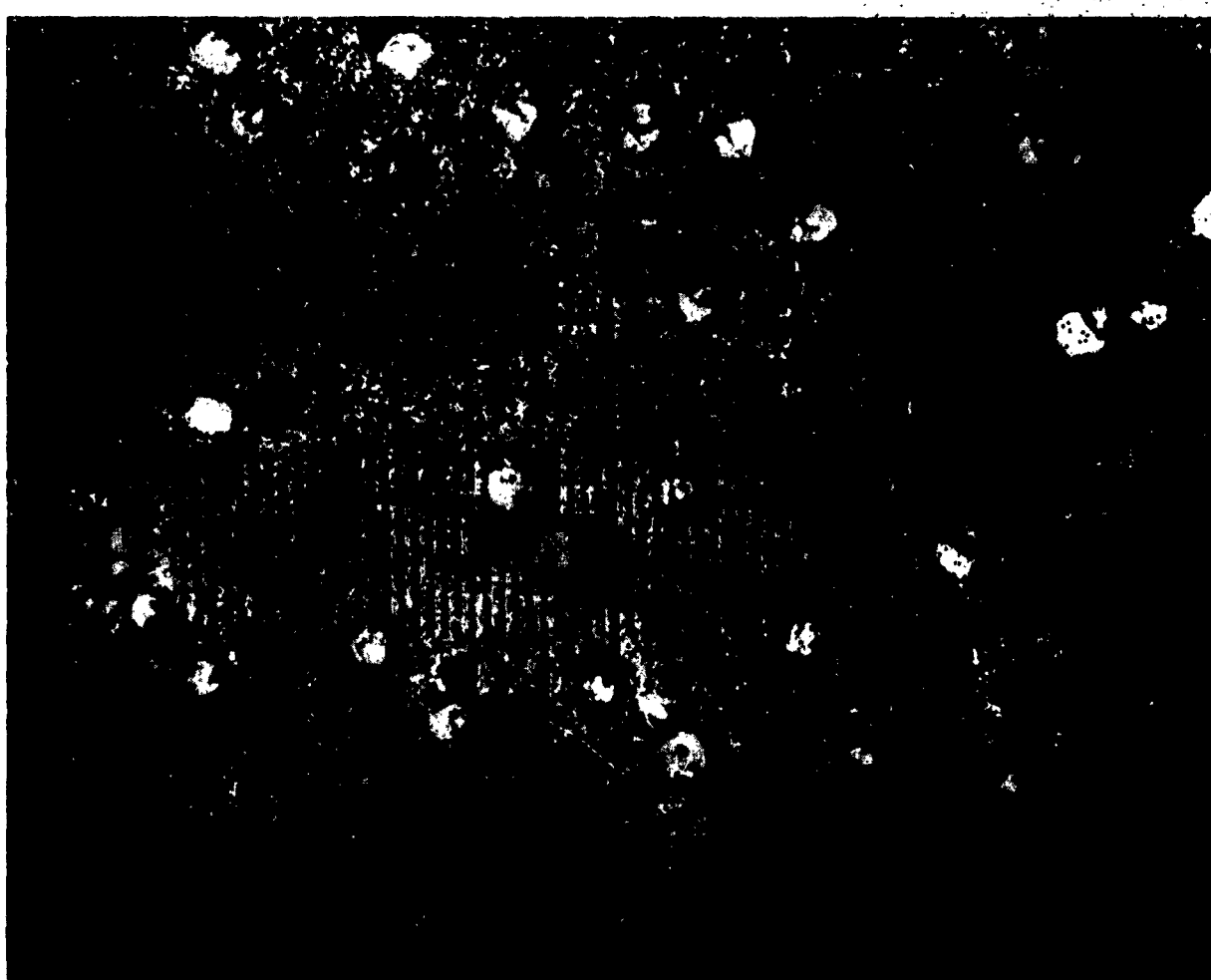

FIG. 8. Photomicrograph of impact discs formed by cloud droplets on surface of smoked glass slide. Slide was exposed 1 sec within cumulus cloud, 30 m above cloud base, over sea. Position: 26°N, 80°W. Time: 1040 EST 10 November 1950. Wind: E. Exposure speed: 26.8 m/sec. Altitude: 730 m. Scale: 1 division = 2.6μ.

TABLE 6. Comparison of the number of cloud droplets observed in a cumulus cloud (see fig. 8) with the number of condensation nuclei present near the sea surface (table 7), the number of large sea-salt particles present at cloud levels (tables 2, 3 and 4), and the estimated number of salt particles which may become raindrops.

	No. cm^{-3} of air
Condensation-nuclei counts near sea surface (average value)	145
Cloud-droplet counts on smoked slides (average value)	50
Total salt particles samples in clear air at cloud levels (average value)	0.4
Estimated salt particles which may become raindrops*	0.001 to 0.005

* Salt particles larger than about 100 $\mu\mu$g sampled in clear air at cloud levels.

are below the "critical radii" at which no accretion occurs (see Langmuir, 1948, table 5).

Thus, the apparent failure of many of the smaller salt particles to form raindrops may be due to their lower "collection efficiency" upon entering a cloud.

Growth of larger cloud elements to raindrop size by coalescence with smaller cloud elements seems generally accepted as a rain-producing mechanism. It is assumed here that the large cloud droplets, formed by condensation on the large salt particles,[8] subsequently coalesce with a great many smaller water droplets in the cloud which have formed on sub-microscopic condensation nuclei. The lower salt concentration associated with increased rain intensity may be due primarily to a greater "distance of fall" of the raindrops within the cloud (Langmuir, 1948), causing coalescence with greater numbers of cloud-water droplets. From Simpson's study (1941b), it would seem that most of the sub-microscopic condensation nuclei on which the numerous cloud droplets form are not sea salt, since an improbably high nuclei-production rate at the sea surface would be required (*i.e.*, about 5×10^4 cm^{-2} sec^{-1}). However, the work of Dessens (1946) suggests a mechanism for the production of great numbers of very small salt nuclei through the shattering of larger crystals attending phase changes from super-saturated droplets in the atmosphere.

The range of the average weight of condensation nuclei given by various writers is very great indeed (see summary by Howell, 1949, p. 138), making questionable the usefulness of estimates of how much salt may be contained in most cloud droplets in the air over the sea. Hence, no estimates are made here of the total weight of salt which might be added to an initially large salt particle due to its coalescence with a great many smaller cloud droplets containing salt. It should be pointed out, however, that 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. A further

[8] Growth to raindrop size by condensation processes is not considered here, because of the excessive time required.

TABLE 7. Measurements of condensation nuclei, with a Schultz counter, at increasing distances inland from the east coast of Florida.* Wind: ESE, force 3. 1430 to 1730 EST 11 November 1950.

Distance from windward shore (mi)	Number of counts	Condensation nuclei cm^{-3}
0.04	19	145
1.2	23	970
7.0	18	22,300**
16.5	20	5,985

* For further data from this area, see Woodcock (1950b).
** In the city of Coral Gables, Fla.

study of the correlation between the larger salt particles and the final raindrop size and salt concentration may give information about the salt present in the numerous cloud droplets.

Table 6 shows the numbers of salt particles sampled over the sea, compared to the number of condensation nuclei and cloud droplets present over the sea. The number of cloud droplets is an average of four samples taken on smoked glass slides (method from Schaefer, 1945) within the base of a cumulus cloud. Fig. 8 shows a photomicrograph of one of these slides.[9] The condensation-nuclei number shown in table 6 is an average of 19 counts made on the beach (see table 7). The total number of salt particles (table 6) is limited, by the impact method of sampling, to particles with weights greater than about 1 $\mu\mu$g. Hence, this quantity is not to be interpreted as indicating that there are not other smaller salt particles present. Table 6 indicates that the number of cloud droplets is much greater than the number of salt particles sampled in clear air at cloud levels, and is near the number of condensation nuclei found a few meters above sea level on the nearby shore.

It is supposed that the increase in condensation nuclei number over land (table 7) causes a greater number of droplets to form in clouds over land as compared to those present in clouds over the sea. The water in land clouds would thus be distributed among many more droplets, reducing the average drop size. This reduction in size may be a factor in accounting for the increase in the heights of rain-producing clouds as marine air moves inland from the Florida east coast to the inland city of Orlando (see Byers, 1949).

7. Conclusions

1. Many large salt particles are present at sub-cloud and cloud levels in marine air over the sea. Passage of this air over 110 km of land produced no great change in the size distribution of these salt particles. This fact suggests that marine winds probably carry these particles many hundreds of kilometers inland (see also Crozier and Seely, 1950).
2. The correlation of the weight distribution of salt par-

[9] The 1-sec timing of the exposure of these slides was manual, using the apparatus shown in fig. 11. Later tests with this apparatus showed that, in five trials, the writer's manual timing of 1 sec varied from 0.7 to 1.1 sec, with an average of about 0.8 sec.

ticles over Florida, the assumed distribution of salt in rains of varying intensity, and the distribution of drop size in rains of varying intensity at Ottawa, Canada and Washington, D. C., suggest that the salt particles play an important role in the formation of rain.

3. The similarity of the numbers of the larger salt particles of cloud levels and the numbers of raindrops of appropriate size and salt concentration suggests that the "chain reaction" process pictured by Langmuir (1948) does not necessarily occur in the formation of rain from warm clouds (T > OC). The effects of the presence of these large salt particles upon the production of rain from super-cooled clouds should also be considered, as was recently emphasized by Ludlam (1951).
4. The fact that the Cl content of rain waters may be accounted for through the presence of a relatively small number of large salt particles suggests that most of the cloud drops with which these large particles coalesce are formed on non-saline nuclei, or that they are formed on salt particles which are so small that most of the cloud droplets are a far more dilute salt solution than are the large droplets with which they coalesce.
5. The relationship shown between the weight and number of the larger salt particles and the number, size and chlorinity of rain suggests further that the failure of some large clouds to produce rain may be due to the absence in them of these salt particles. If such is the case, the possibility of inducing precipitation by seeding with appropriate numbers and sizes of sea salt or other particles becomes evident.

8. Methods

The general method used to sample airborne salt, and the laboratory technique which is used to count and weigh the individual salt particles, have already been described in some detail by Woodcock and Gifford (1949).

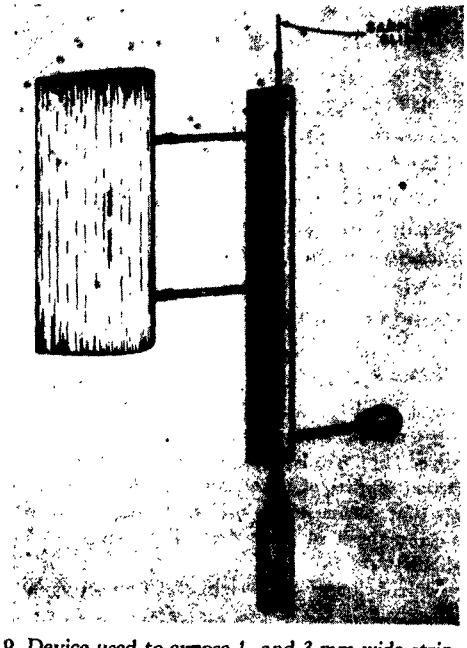

FIG. 9. Device used to expose 1- and 3-mm wide strips of glass at wind and aircraft speeds. Slide at top is extended from and can be drawn down into airfoil tube below.

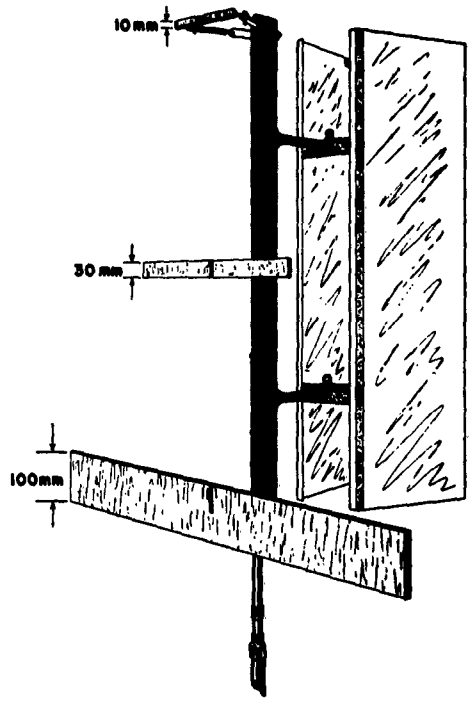

FIG. 10. Wind vane used to hold 10-, 30- and 100-mm wide formica strips normal to wind. Actual sampling surfaces were removable glass slides recessed in middle of strips.

A very brief description is given below of the fundamental sampling and measuring methods used. Some details are given of a modification of the sampling methods already described, which has made it possible to catch large particles which are present in the air in lower numbers. In addition, some methods and new

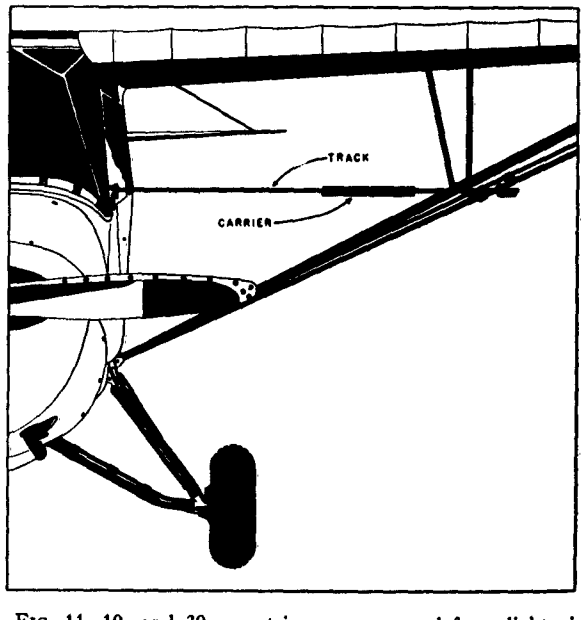

FIG. 11. 10- and 30-mm strips were exposed from light aircraft. On track extending from plane's cabin, carrier, with attached strip and recessed sampling slide, can be hauled out into free air stream and held normal to air flow for required exposure time.

results are given concerning the weight distribution of salt particles in the air, as determined by precipitation. These results are compared to those obtained by impact sampling.

Airborne particles of sea salt have usually been sampled by the writer by exposing 1-mm wide glass slides to the free air stream. The salt particles impinge upon and adhere to the slides and, if liquid, assume a hemispheric form, due to the presence of a hydrophobic film on the glass surface. These slides are then taken to the laboratory and placed in a chamber on a microscope stage, where counts and diameter measurements of droplets are made under controlled temperature and humidity conditions at values near 25C and 90 per cent. Usually at least 500 droplets are measured on each slide. Concentrations of salts in the droplets which form under these conditions were determined by the equal-pressure (isopiestic) method (Glasstone, 1940), and the weights of salt in individual hemispheric droplets were computed from diameter measurements, using the values of Higashi and others (1931) for the specific gravities and the vapor pressures of concentrated sea water.

9. Multi-strip sampling

The triangle symbols on fig. 13 show the approximate range of weight and number of particles sampled on the 1-mm wide glass strips, which had previously been used for almost all sampling (see Woodcock and Gifford, 1949). The slope of a smoothed distribution curve drawn through these points suggested, however, that larger, less numerous particles were also present in the air. The other symbols plotted on this figure, giving sampling results using a modified technique, indicate that there were indeed larger particles present in the air in relatively low numbers.

To catch these large, more sparsely distributed particles, the 1-mm sampling surfaces would have to be exposed, at a given air speed, for a much longer time than that used (see table 1, line 2). However, a prolonged exposure of these small slides is not, in this case, practical, since it will cause an excessive "covering fraction" (see footnote, table 2), and introduce

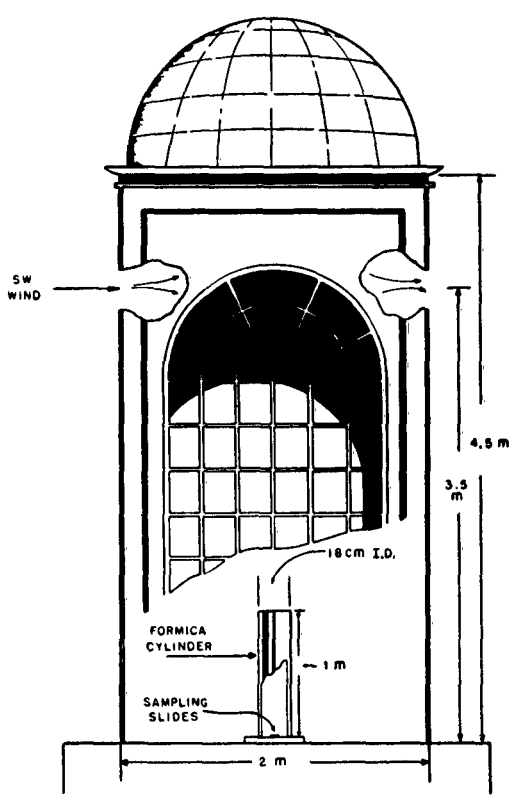

FIG. 12. Cupola of laboratory building was used as precipitation chamber. See text and table 1 for further information.

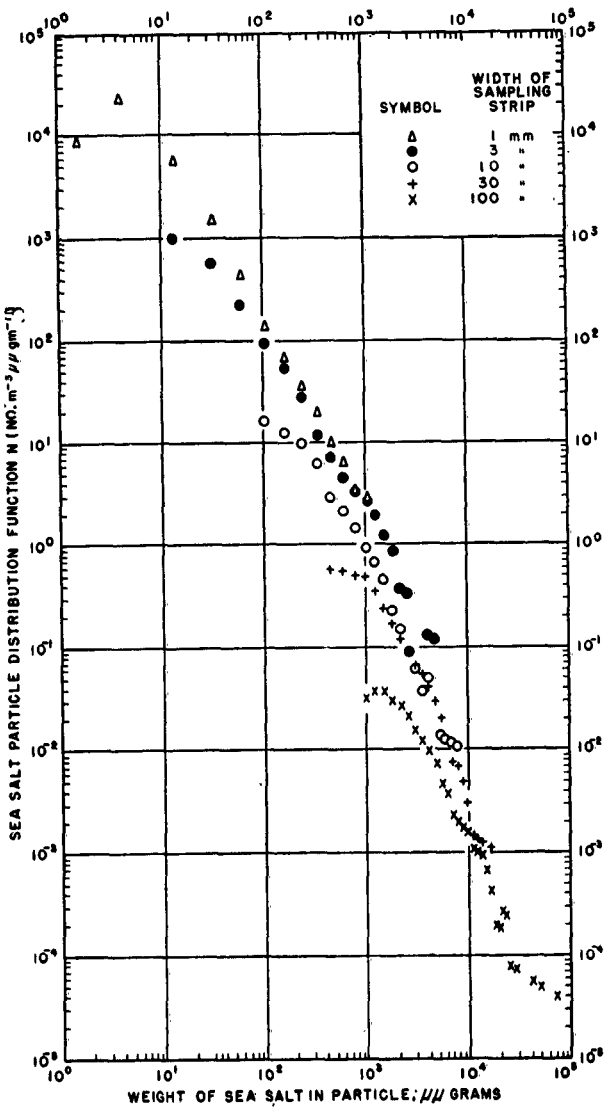

FIG. 13. Sea-salt particle distribution function, using uncorrected data from samples taken 15 September 1950, 5 m above sea surface. Symbols represent various widths of strips used to sample different particle sizes. See table 1, fig. 1 and text for further information.

serious errors in the size- or weight-distribution determinations (Langmuir, 1944, pp. 59–68).

In sampling more nearly the complete range of weight and number of salt particles, several widths of sampling strips have been used. Fig. 10 shows the mounting of the larger of these strips for sampling near the sea surface. In sampling from light aircraft, the 10- and 30-mm sampling strips were mounted on a small track, extending from the fuselage to one of the struts (see fig. 11). The actual sampling surfaces are removable glass slides, recessed near the centers of the formica strips. The apparatus shown in fig. 10 is exposed vertically and the wind vanes hold the sampling surfaces normal to the wind.

Fig. 13 shows the uncorrected distribution of weight and number of salt particles which were deposited on strips from 1 to 100 mm in width, exposed at the end of a dock at Woods Hole. Table 1 gives other information relative to the samples taken on these strips. Fig. 14 shows photomicrographs of some of the salt particles, as droplets (relative humidity: 91 per cent), which were sampled on the strips of various widths. Evaporation of the water in these droplets always reveals a crystalline residue or nucleus (for example, see fig. 16).

Fig. 1 shows the data on fig. 13 corrected for deposition error, according to Langmuir and Blodgett (1945). In fig. 1, it is seen that the smaller particles sampled on the various sizes of slides are in each case eliminated (compare figs. 1 and 13). This is done because the deposition error correction is inaccurate for these particles (see Langmuir and Blodgett, 1945, p. 25 and 25A).

The overlapping of counts on the slides of various widths has also been eliminated in fig. 1 and in subsequent sampling (see figs. 2, 3 and 4). In general, observations were not used unless the deposition efficiency for the smaller particles was 0.70 or more. Thus, a selection is made of that portion of the particle-weight spectrum which is most adequately sampled by each width of strip. Selection of sampling-strip widths thus makes possible a selection of the range of weight of aerosol particles to be sampled and avoids excessive covering fractions. The first numbers in table 1, lines 5, 6 and 8, are derived from the counts made on the 1-, 3-, 10-, 30- and 100-mm wide strips. The portion of the particle-weight spectrum selected from the particles measured on each strip is shown by the various symbols in fig. 1.

The addition of all of the observed weights shown in fig. 1 gives a value of 8.87 $\mu\mu$g cm^{-3} of air (see

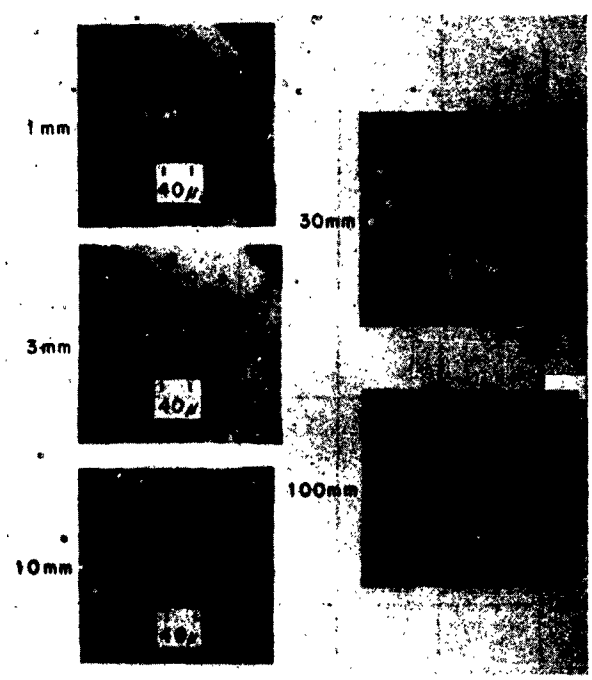

FIG. 14. Photomicrographs of salt particles as hemispheric droplets (relative humidity: 91 per cent) on glass surfaces exposed to wind 15 September 1950. 1- and 3-mm surfaces exposed with device shown in fig. 9; 10-, 30- and 100-mm surfaces exposed on strips shown in fig. 10. Photographed areas on slides selected to illustrate increase in size of particles sampled as width of sampling strips increased. See table 1 for further information about these samples.

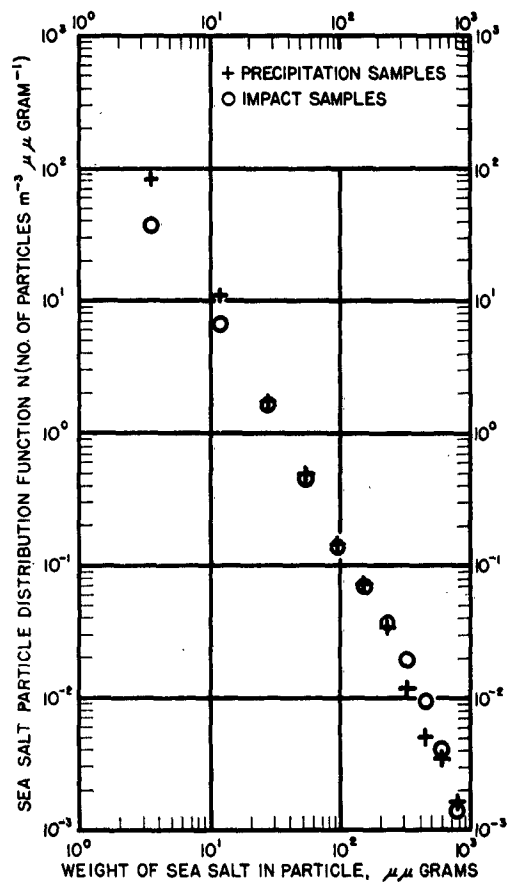

FIG. 15. Comparison of sea-salt distribution as sampled by precipitation and by impact on 15 September 1950. See table 1 and text for further discussion.

line 5, table 1). Titration, with a silver-nitrate solution, of the chlorides collected upon other surfaces exposed simultaneously, gave a value of 9.21 $\mu\mu$g cm^{-3}. Thus, the multi-slide, equal-pressure method of measuring the quantity of salt in the air gives a value very close to the value derived by direct titration. Titrations of total chlorides are occasionally used as a test of the validity of the equal-pressure method of determining the weight of the total salts per unit volume of air (see Woodcock and Gifford, 1949, p. 183). Determination of the weight of salt cm^{-3} of air was also made from precipitation counts, using estimates of the particle settling-speeds.

10. Precipitation samples

Precipitation samples of salt particles taken on 15 September 1950 show close agreement with the impact-sampling results. Fig. 15 shows the distribution of salt particles determined from precipitation samples (the +'s), compared to the distribution of particles sampled at the same time by impingement. The difference in the results is insignificant, with the exception of the smaller particles, where deposition-error corrections for impact sampling become questionable (Langmuir and Blodgett, 1945). However, we are concerned here primarily with nuclei larger than 10 $\mu\mu$g.[10] An adequate sampling of particles larger than 775 $\mu\mu$g was not taken by this precipitation sampling, because of insufficient exposure length and the small sampling surface used.

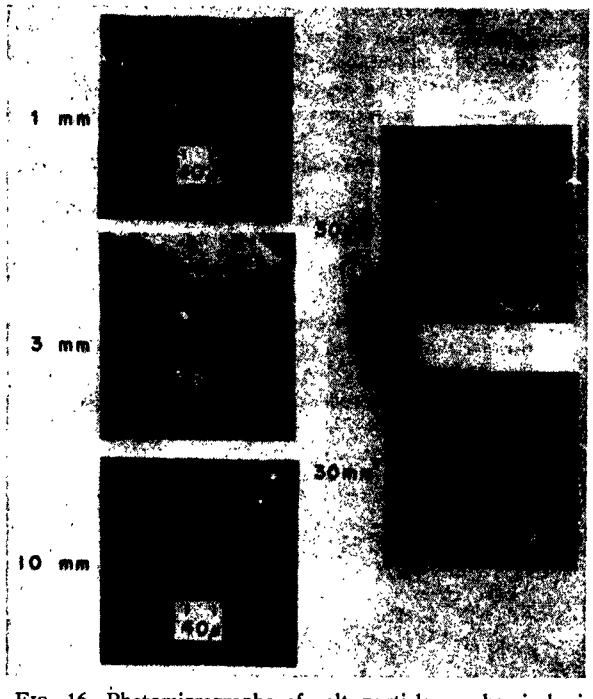

FIG. 16. Photomicrographs of salt particles as hemispheric droplets (relative humidity: 91 per cent) on glass surfaces exposed from aircraft at 1523 m. Table 4 gives further information concerning these samples. Surfaces 1 and 3 exposed with device shown in fig. 9; surfaces 10 and 30 exposed as in fig. 12. Photographed droplets selected to illustrate increase in size of particles sampled as width of sampling strip increased. Second photograph of 30-mm sample made at low relative humidity (40 per cent), to reveal crystalline nature of residue or nucleus.

The simple equation below was used in converting the precipitation counts to numbers cm^{-3}:

$$N = N_c/SR, \tag{1}$$

where N = number of particles cm^{-3} of air, N_c = number of particles counted cm^{-2} of slide surface; S = exposure duration (sec), and R = settling rate of particles.

Settling rate was computed from Stokes' law. Radii of the particles as spheres were from 1.5 to 8 μ, assuming equilibrium at the temperature and relative humidity of the ambient air in the precipitation chamber. Radii of the particles in the air were computed as follows:

$$R = [(W/CD)/(4\pi/3)]^{1/3}, \tag{2}$$

where R = particle radius (μ), W = weight of salt in particle ($\mu\mu$g), C = salt concentration by weight fraction, and D = particle density (the latter two from Higashi and others, 1931).

Precipitation samples were taken in the cupola on the roof of the laboratory, as suggested in fig. 12. This cupola is about 20 m above sea level and near the dock where samples were taken by the impact method. Drifilm-coated, clean glass slides (3 × 30 mm) were placed upon filter paper at the bottom of a cylinder of formica, which stood on the floor of the cupola. A continuous supply of airborne particles flowed through the upper part of the cupola and could rain down within the closed space, through the open end of the cylinder and onto the slides at the bottom. This cylinder, which was shielded from direct solar radiation, reduced the turbulence of the air within the building, so that relatively still air overlay the sampling slides. The relative humidity of the air over the cylinder in the cupola varied between 78 and 81 per cent during the sampling period, while that outside varied from 91 to 93 per cent. The weights of the particles which settled on the exposed slides were subsequently measured by the isopiestic method already described, and the results are given in fig. 15 and table 1 (last column).

Thus, the distribution of particle number and weight, sampled by impact on glass slides and measured by the isopiestic method, is in close agreement with these values determined by precipitation and the isopiestic method. The above agreement and the titration result already given clearly do not prove that fig. 1 represents precisely all the salt particles present

[10] The largest condensation nucleus considered by Howell (1949, p. 140), in his study of the growth of cloud drops, was 3 $\mu\mu$g (*i.e.*, 10^{-13} moles). The present study shows that this weight is far from the maximum actually present at cloud levels.

in the air during the sampling period. The results in fig. 1 are considered a useful approximation of the correct distribution of weight and number among salt particles, present in the lower air, which occupy the weight range of about 3 to 20,000 $\mu\mu$g. The results of the multi-strip sampling from aircraft (figs. 2, 3 and 4), which are based entirely on the equal-pressure method of measurement, are regarded as an equally useful approximation to the correct weight and number of salt particles present at higher levels.

Acknowledgments.—Instruments used in the work reported here were made by Kenneth G. McCasland, Donald Parsons, Robert G. Walden and A. T. Spencer. The writer is also indebted to H. Stommel and A. C. Redfield for helpful criticism in some phases of the work, and to Arnold Arons and D. C. Blanchard for their sustained interest, encouragement and help.

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Remarks on "Sea salt in a tropical storm"

By JAMES E. MCDONALD

Dept. of Physics, Iowa State College, Ames

4 January 1951 and 24 February 1951

In a recent paper in the JOURNAL, Woodcock[1] reported some measurements of concentrations of airborne sea salt in a Florida hurricane. I would like to call attention to what was for me a misleading mode of presentation of the magnitudes of latent heat released (and of associated temperature increases) in table 2 of Woodcock's paper. These magnitudes, as computed and tabulated, represent total amounts of heat and temperature increase that would be observed if the associated amounts of liquid water had been built up on initially dry salt nuclei entirely by condensation. Was it Mr. Woodcock's intention that the reader assume such a process to be operative?

As a second comment on the paper, it may be pointed out that the amount of heat released by condensation on the observed 0.837×10^{-6} g of salt per gram of air, as it ascends from a level of 82 per cent relative humidity near the surface to the lifting condensation level (about 1300 ft above the surface for Woodcock's data), is percentually so small compared to the magnitude of the internal-energy decrease of the associated air as to raise serious doubt concerning the significance of salt content of air as an additional stability criterion. The magnitude of the very adiabatic cooling effect required to realize the tabulated latent-heat release for 98 per cent relative humidity is, for Woodcock's data, about eighty times larger than the dropwise condensational heating effect, (0.055–0.006)C. Only if Gautier's[1] and not Woodcock's salt-content measurements approach the true upper limit of attainable salt concentrations could one expect appreciable effect of salt content on stability, in view of the above relative magnitudes. Hence, I would like to ask whether Mr. Woodcock feels that the experimental techniques employed by Gautier, when he measured a salt concentration some thirty times greater than the maximum of Mr. Woodcock's random samples in a hurricane, were such as to justify acceptance of Gautier's values as a more reliable indication of the upper limit of attainable salt content over stormy seas than Mr. Woodcock's own observations in this one Florida hurricane. And if so, what meteorological or oceanographic conditions prevailing during Gautier's observations might have been responsible for this thirty-fold excess of spray formation in the storm off France over that off Florida? In particular, one wonders if Gautier's measurements might have been made at a lower level or nearer the surf zone than were Mr. Woodcock's.

As a third point, I would like to ask whether it may not be possible that a locally more intense effect, opposite to that considered by Mr. Woodcock, occurs within a very shallow layer above an agitated sea surface? If one makes some rough calculations, which need not be reproduced here, he finds that newly formed droplets of sea water will evaporate so rapidly in coming to vapor equilibrium with their new environment that virtually all of the evaporational cooling effect may be confined to a thin surface layer over the sea. The actual depth of this evaporationally cooled layer should depend in part on the size distribution of the nascent droplets, but probably still more on the relative-humidity profile in the surface layer. Without here attempting to examine very closely the magnitudes involved, I would merely like to point out that, in contrast to the roughly 1300-ft depth of the layer through which is distributed the condensational heating effect envisaged by Mr. Woodcock, the layer within which occurs the equal and opposite evaporational cooling effect must surely be measured in feet or, at most, tens of feet. That the lower rather than the upper limit of this suggested range of cooling depths is probably the more nearly correct is suggested by the fact that a drop of sea water whose initial diameter is only one micron would require only about a millisecond to evaporate down to a concentration implying vapor equilibrium with air of 82 per cent relative humidity, if suddenly placed in air of that humidity. However, the steady-state relative humidity above an agitated sea surface would be expected to exceed this 82 per cent value up to a height of perhaps a few feet above the surface, so the depth of the cooled layer might be expected to be small and of that order most of the time. (A relative humidity of 82 per cent, incidentally, has been referred to for no other reason that that it was involved in the paper here under discussion.) In view of these arguments, might it not be more profitable to look for possible micrometeorological effects of this concentrated cooling process than to look for stability effects of its much more diffuse inverse?

It would be interesting to know if any such cool layer just above the sea surface has ever been observed instrumentally. It seems somewhat doubtful that it would have been detected by chance, because the very requirements for appreciable spray formation, strong winds and rough seas would render such observations difficult.

[1] A. H. Woodcock, "Sea salt in a tropical storm," *J. Meteor.*, **7**, 397–401, 1950.

A minor error, appearing in the second equation on page 401 of Mr. Woodcock's paper may be pointed out. The specific heat should be in the denominator, not in the numerator.

Reply

By ALFRED H. WOODCOCK

Woods Hole Oceanographic Institution, Woods Hole, Mass.

13 March 1951

Professor McDonald has made some useful comments concerning my paper. These comments are discussed below in the order of their occurrence.

Table 2 of my paper was intended to make it possible for the reader to see at a glance the total amounts of water present on the sea salt at different relative humidities, beginning at an observed value of 82 per cent. The table was also intended to give the reader, by a simple subtraction, the water condensed and the heat released through successive increases in relative humidity at a constant value for air salt, or through successive increases in air salt at a constant relative humidity. The fact that the first relative humidity in table 2 is 82 per cent (a value observed in the tropical storm) seems to have been misleading, causing the assumption that the values for water condensed and heat released were intended to represent only the amounts which would be released above a relative humidity of 82 per cent. Clearly this is not the case, since the tabulated values for water condensed and heat released are not zero at 82 per cent relative humidity.

As pointed out by Prof. McDonald, the quantity of heat released to a gram of air by condensation on 0.837×10^{-6} g of salt, as the relative humidity increases from 82 to 98 per cent, is very small. However, the purpose of this section of the paper was the introduction of the idea that small parcels of marine air may contain enough salt to cause the release of much larger quantities of heat. Gautier's high values for airborne sea salt were derived from samples taken by filtering the air at a lighthouse on the French coast. This method gives average values through long air trajectories of many thousands of meters. In some smaller parts of this trajectory the salt concentration would, of course, be even higher. Gautier's values, like those derived in the hurricane at Hillsboro Lighthouse, may have been affected by local surf. The need for reliable observations of airborne sea salt is clear. I now have an instrument with which samples may be taken continuously from aircraft and in parcels of air as small as 15 m in horizontal extent. This instrument should make possible the determination of the maximum quantities of salt present at various wind speeds and at locations free from the influence of shoreline surf.

As Prof. McDonald indicates, evaporation of the water from the original sea-water spray should cool a layer of air close to the sea surface. The heat released by subsequent condensation on the concentrated sea-water drops or particles will, of course, be equal to the heat utilized in evaporating the spray, under the assumption that the final salt concentration equals that of the original sea-water droplets. However, this subsequent condensation on the concentrated sea-water particles remaining in the air may occur remote from the area where heat was utilized in evaporation. Also, condensation will probably occur in different air parcels from those in which evaporation occurs, since the salt aerosols are continuously settling in the air. Thus heat which might otherwise appear as sensible heat in one area is utilized in evaporating spray drops, and may be subsequently released in the air (at appropriate relative-humidity values) over a different area. Hence a state of unstable equilibrium may exist in a parcel of salt-laden air which would not necessarily be indicated by a measure of dry-bulb temperatures, water-vapor content and air pressure.

The effects of airborne sea salt in liberating latent heat in the atmosphere at lower relative humidities are of interest to me because these effects may be an additional factor contributing to the vertical transport of water and water vapor in the sub-cloud layer over the oceans.

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