

OBSERVATIONS OF THE SALT CONTENT
OF THE LOWER MARINE ATMOSPHERE

by

Theodore Howard Benttinen Jr.

B.S., University of Washington

(1967)

SUBMITTED IN PARTIAL FULFILLMENT

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Department of Geology and Geophysics

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Certified by

Thesis Supervisor

Accepted by

Chairman, Departmental Committee

on Graduate Students

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Abstract

Measurements of the total sea-salt content of the air were made from shipboard at a height of 10 meters. The majority of the data were taken in the Indian Ocean, but data from the Mediterranean Sea, Red Sea and adjacent waters were included.

Small diameter wires were exposed to the wind. The sodium found on the wires was converted into total mass of sea-salt by assuming the ion ratios of sea water.

At winds above 8 to 10 knots, the sea-salt content increased logarithmically with increasing wind speed and an empirical formula was obtained. For the lighter winds no clear-cut relationship was found. The sea-salt content shows no obvious relationship to geographical position for maritime environments.

The effect of low-level instability on the sea-salt content was measured and found to be significant. For winds above 15 knots, greater instability resulted in a higher sea-salt content. The converse was true for low winds, i.e. those below 7 knots. In this case, greater instability was associated with a lowering of the sea-salt content.

The effects of washout and sedimentation on the sea-salt values were observed and qualitatively discussed.

Thesis Supervisor: Henry G. Houghton

Title: Professor of Meteorology, Head of the Department
of Meteorology

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My interest in this particular phase of oceanography was stimulated during many rewarding discussions in the fall of 1964 with Dr. Duncan C. Blanchard of the Woods Hole Oceanographic Institute. Mr. A.T. Spencer, also of that laboratory, contributed generously with advice about the methods of analysis of sea-salt.

I would also like to thank Mr. Paul Willis for his patient help during the various shipboard analyses.

Finally, I would like to thank Mr. A.R. Miller of the Woods Hole Oceanographic Institute for instilling the basic idea which led to my discussions with Dr. Blanchard. Mr. Miller was kind enough to supply me with the equipment and funds necessary for the shipboard segment of this investigation through National Science Foundation grants G.P. 821 and G.P. 870.

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I. INTRODUCTION

Interest in natural aerosols was initiated by the classic papers of John Aitken. These were published in the latter quarter of the nineteenth century and subsequently collected in a convenient volume (Aitken, 1923). Aitken was able to show that the condensation of vapours depended upon the presence of free surfaces in the form of tiny particles of "dust." In one of his major papers, "On Dust, Fogs, and Cloud," published in 1881, Aitken realized the important role which the oceans could play in producing the "dust" particles so necessary for condensation. "In all probability the spray from the ocean, after it is dried and nothing but a fine salt-dust left, is perhaps one of the most important sources of cloud producing dust." These particles having their origin in the sea have since become known as sea-salt particles or more commonly, sea-salt nuclei.

With the invention of the Aitken nuclei counter, a device which works on the general principles of the Wilson cloud chamber, many observations were made of the number of "dust" particles in the air. However, only the total number of particles in a particular sample was determinable, and because of the very great differences in concentration, the presence of the larger particles was masked by the much more numerous small particles. Hereafter, in general accordance with the

literature, those particles with a radius of one-tenth micron or less will be referred to as Aitken particles, those with a radius between one-tenth and one micron as large particles and those with a radius of one micron or more as giant particles.

Because of the widespread use of the Aitken counter very little work was done on the large and giant nuclei during the first part of this century. Various observers came to realize that particles could be produced by the sea and that these particles would probably consist of sea-salt but data pertaining to the actual measurement of these particles were virtually nonexistent until the work of Owens (1926). He measured the sizes of some of the largest salt particles found in clear air near the sea surface. Houghton (1932) felt that at least the smaller droplets in the fogs which he investigated were the result of condensation onto sea-salt particles. After measuring the size spectrum of fog droplets, Köhler (1936) was convinced that sea-salt particles were a major contributor to the supply of cloud condensation nuclei. Working with visibility measurements, Wright (1940) was able to confirm the hypothesis that the opacity of marine air varies with humidity in such a way as to indicate that condensation on hygroscopic particles is the cause. Those particles responsible for this effect were found to lie mainly in the so-called large category.

Simpson (1941a) stated that breaking waves were the source

of the sea-salt nuclei. He also argued (Simpson, 1941b) that the sea-salt particles were far too few in number to account for normal cloud formation. He was perhaps the first to actively further this latter point and more recent work has affirmed his belief. It was not until the detailed work of Woodcock and Gifford (1949) and Woodcock (1950a, 1950b, 1952, 1953) that significant amounts of data on the size (and weight) distributions of sea-salt particles were obtained. Even though those particles in the giant range were found in very small numbers in comparison to the Aitken nuclei, it soon became evident (Woodcock, 1950b) that they were important for the formation of rain in clouds above freezing.

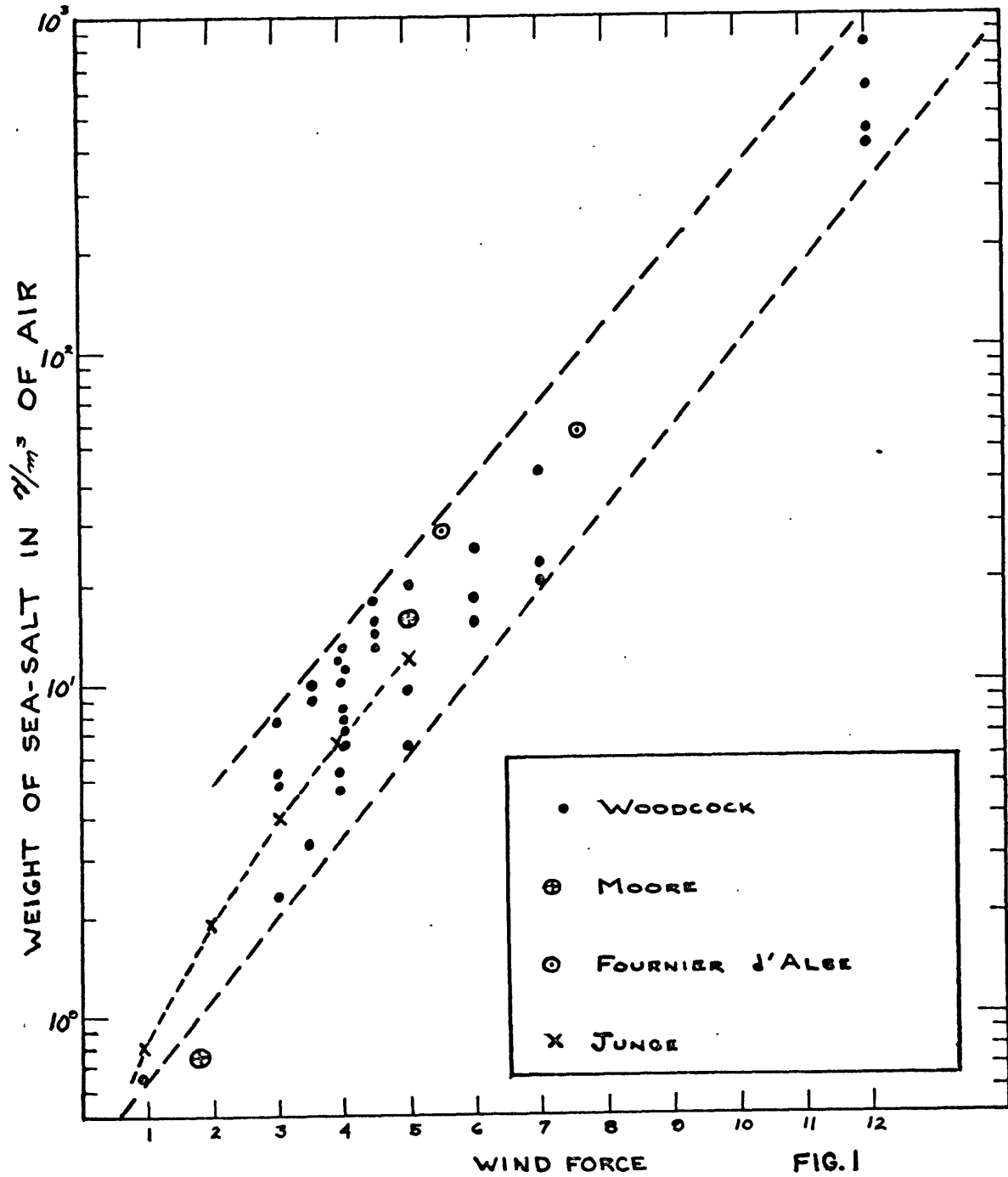
Since the publication of Woodcock's work, many other investigators have become active in the field. Lodge (1955) investigated the concentrations of sea-salt nuclei over the island of Puerto Rico. His work tended to support Woodcock's earlier endeavors. Moore (1952) and Moore and Mason (1954) made measurements of the size distribution of these nuclei over the North Atlantic at a height of about ten meters. Twomey (1954) and Junge (1954, 1957a) have also been quite active in the field.

More recently, Woodcock et al (1963) suggested that the latent heat released by the condensation of water vapor onto the surfaces of airborne sea-salt particles may be an additional stability factor, particularly during periods of high salt production. Isono et al (1966) found these giant parti-

cles important in the formation of snowfall from convective cells over the sea. Toba (1965a, 1965b, 1966) presented a detailed account of the world-wide distribution and production rate of the giant sea-salt nuclei. Reviews of the subject of sea-salt nuclei may be found in Mason (1957), Junge (1963) and Roll (1965).

During the investigations of the size distribution of these sea-salt nuclei the question of their origin constantly arose. Many early investigators felt that the spray generated by breaking waves at sea (as well as by coastal surf) presented the most obvious source. However, Jacobs (1937) theorized that perhaps bubbles and not wind blown spray were the primary source of the sea-salt nuclei. Kientzler et al (1954) Knelman et al (1954) and Blanchard and Woodcock (1957) did detailed work on the problem, often employing high speed photographic techniques. In the latter paper, the authors were able to show that droplets from the jets of bursting bubbles could account for the majority of the giant sea-salt particles found in marine air.

It thus appears that the production of these giant particles will be linked quite closely to the white cap coverage and hence the wind speed near the surface of the ocean. Such a correlation has indeed been found by several observers for the total amount of sea-salt in the marine atmosphere (Fig.1) (Junge, 1958). While information on the size and weight distributions of the individual particles is perhaps more valu-



able, knowledge of the total amount of sea-salt in the air is important for a complete understanding of the air-sea coupling process. Therefore, after some preliminary discussions with Dr. D.C. Blanchard, then of the Woods Hole Oceanographic Institute, I decided to attempt to determine the variations (at a height of about 10 meters) of the total sea-salt content with various meteorological parameters as well as geographical position. These parameters were wind speed and direction, relative humidity, present meteorological conditions (e.g. were showers present or not), cloud type, low level instability and the like. This paper is a presentation of the methods and results of my experimentation on board the R.V. Atlantis II during cruise 15 which took place in 1965. The relevant part of the cruise track is presented in Fig. 2.

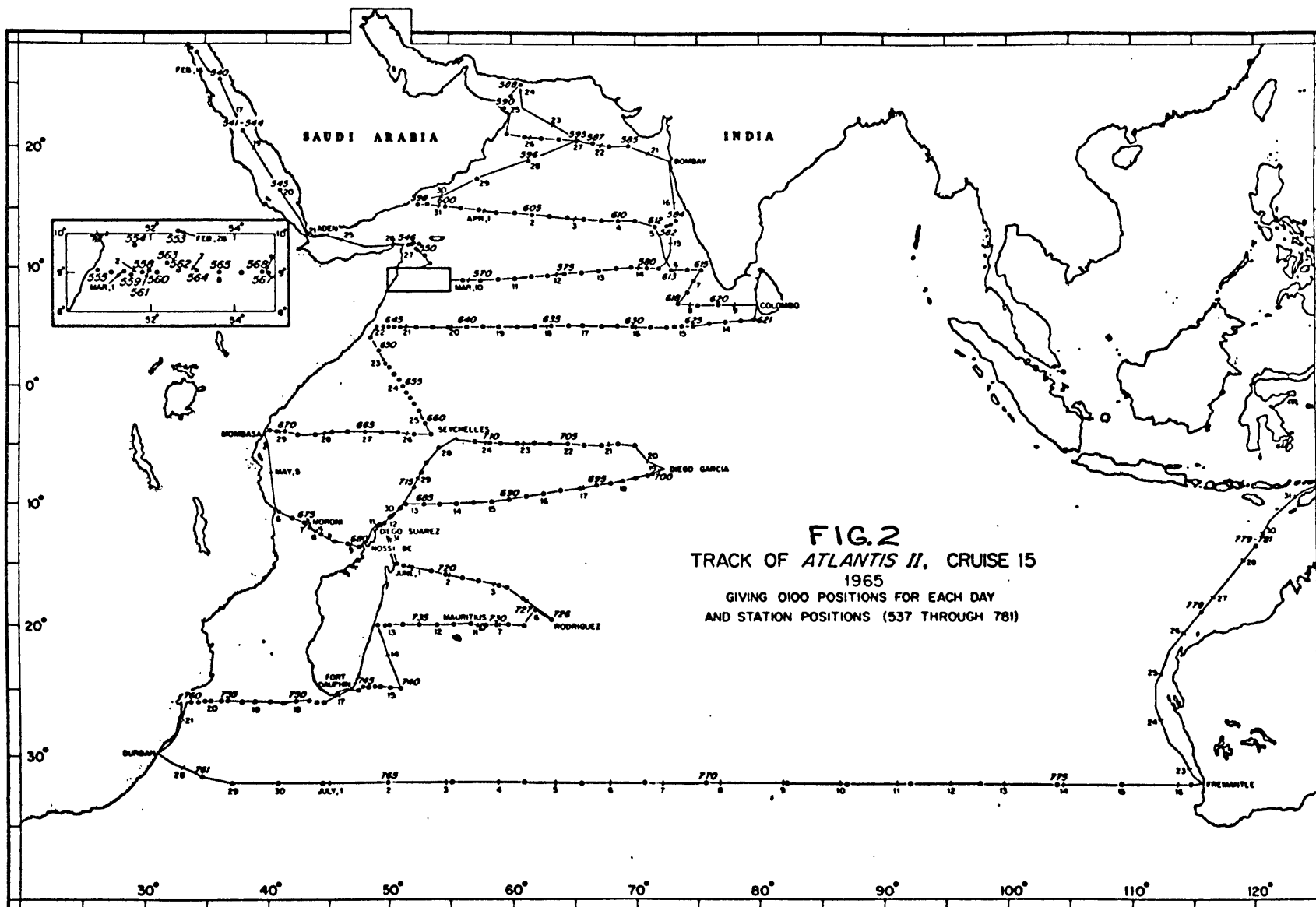
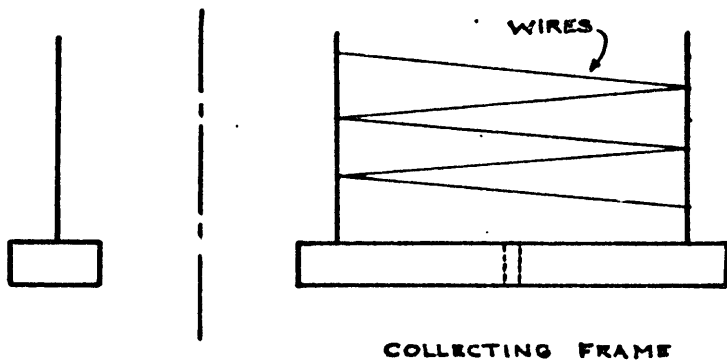


FIG. 2
 TRACK OF *ATLANTIS II*, CRUISE 15
 1965
 GIVING 0100 POSITIONS FOR EACH DAY
 AND STATION POSITIONS (537 THROUGH 781)

II. COLLECTION METHODS

The salt was collected on 152μ diameter stainless steel wire which was attached to a frame consisting simply of a base and two upright metal dowels (Fig.3). These frames (and hence the wires) were kept normal to the wind by mounting them on a wind vane built for that purpose. The vanes and their frames were secured onto pipes and the latter were extended forward from the railing above the bridge of the R.V. Atlantis II. This procedure insured that when the resultant wind was anywhere forward of abeam, an unobstructed flow of uncontaminated air would pass by the collecting wires. The collection height was approximately 10 meters above the sea surface. One set of pipes was fastened on the port side, and another on the starboard side in order to facilitate the taking of double samples. The collecting wires were approximately 10 m. apart in the horizontal.

In order to obtain the flow of air past the wires, a standard four-cup anemometer system which drove a simple digital counter was employed. The counter and the necessary batteries were placed in a waterproof PVC container with one end made of clear plastic. This insured that the counter and batteries would remain free from corrosion while still permitting the counter to be read with ease. An insulated set of leads ran from the container along a set of pipes on which the anemometer was mounted. It was mounted amidships,



COLLECTING FRAME

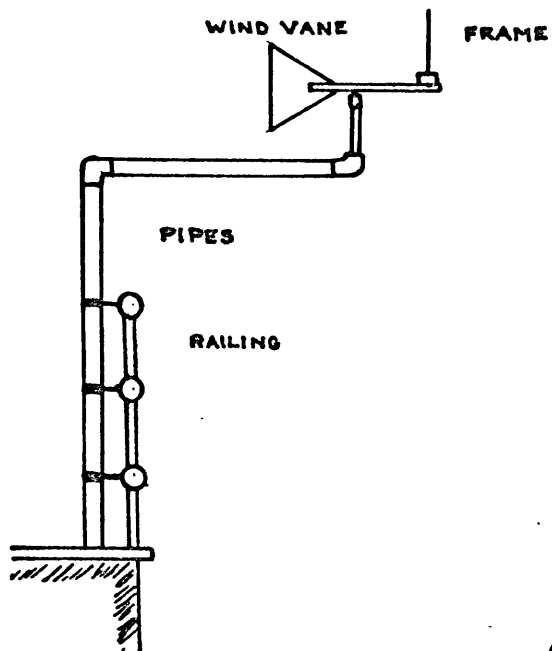
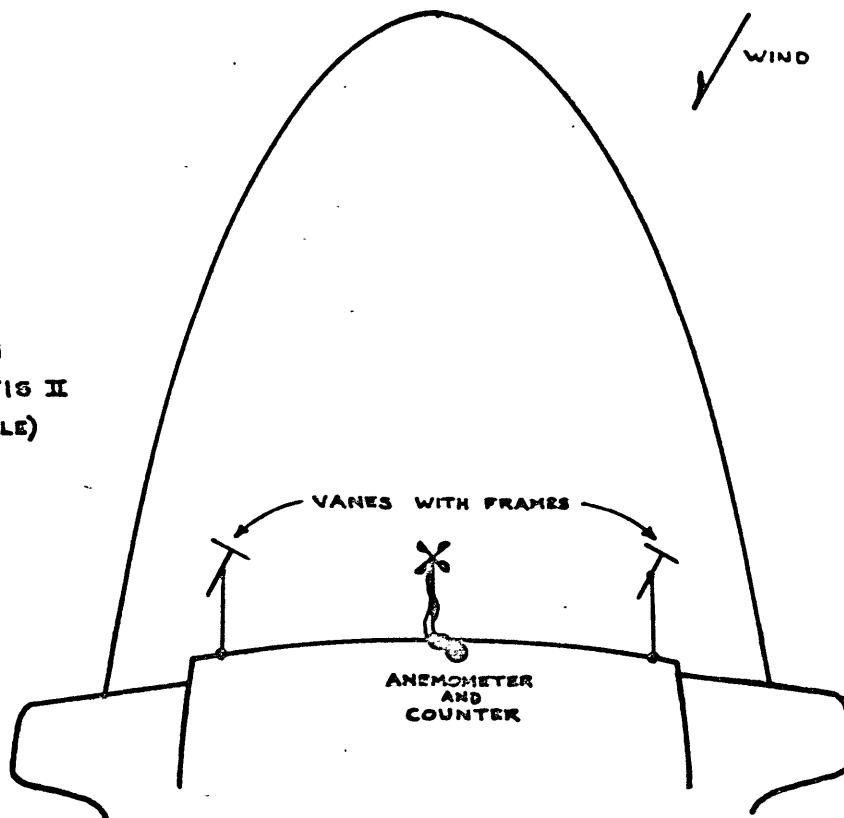


FIG. 3

BOW REGION
R.V. ATLANTIS II
(NOT TO SCALE)



i.e. halfway between the two sets of exposed wires. Several anemometers were checked against one another and since they read the same, only one was employed for most of the observations. Before starting on the cruise, I calibrated the four anemometers along with their counters in a wind tunnel at the Woods Hole Oceanographic Institution. Therefore by noting the counter readings before and after exposure of the wires, I was able to determine the number of meters of air which had passed.

A typical collection procedure would consist of the following. First of all, I would wash the wires (which were permanently attached to the frames) in distilled water and then attach the frames to the wind vanes by means of a single central bolt. The pipes were attached to the railing in such a way that they could be swung in for this procedure. I would then note the time, switch on the counter, and record the pertinent meteorological information. The latter consisted of: vector wind and direction, (note: vector wind is the resultant of the actual wind and the ship's velocity), actual wind and direction, sea state, wet and dry bulb temperature readings, sea surface temperature, and a verbal description which consisted of cloud type and amount as well as the present and past "weather." Particular attention was given to noting the number of showers visible and to changes which occurred in the actual wind velocity during a collection period.

The collecting wires were exposed for varying periods, depending on the wind velocity. At very low wind speeds they were not exposed unless the ship was underway. This was done in order to keep the collection efficiency of the wires reasonably high. During high winds when there was a chance of spray contamination from the bow waves, or when the wind was aft of the beam, the samples were taken during the various oceanographic stations. At these times, the ship would be headed into the wind and sea with little or no forward progress, a procedure known as "steaming on the wire." Thus by varying the manner of collection I was able to obtain samples during all the various winds encountered without either spray contamination or the obstruction of the ship's superstructure.

Generally there were no significant changes in the meteorological conditions during the one-half to two hour collection periods, but when there were, these were noted at the end of the collection period. When there had been a sufficient passage of air to insure a significant sample the frames were removed from the vanes, and the counter reading noted. The frames were then carefully clamped to the inside of a box especially made for this purpose and stored in a safe place until there were enough of them to warrant a run on the flame spectrophotometer.

III. ANALYSIS METHODS

When I obtained from 10 to 20 exposed frames, the material collected on the wires was analyzed for its sodium content. This was done by means of a standard Beckman DU flame spectrophotometer with a special small bore atomizer. The instrument was calibrated and tested according to the accompanying Beckman manual and no difficulties worthy of mention were encountered. Two large batches of standard solution were made up by mixing appropriate volumes of Copenhagen standard sea water and double distilled water. These were both 100 ppm (parts per million) by weight sodium ion. By pipetting various amounts of double distilled water into a known volume of the 100 ppm solution, standards from 0-100 ppm could be easily obtained.

These standards were run through the atomizer and a plot of net luminosity (transmittance minus a blank) versus the sodium concentration in ppm was obtained (Fig. 4). The standardization procedures always gave smooth curves and the results were repeatable at the end of each analysis. Over the months new standards were made up to check for any deterioration of the original ones but no significant changes were observed.

The collecting wires were washed in a large drop of double distilled water whose volume (1 ml) was known. In order to facilitate the ensuing calculations exactly one meter

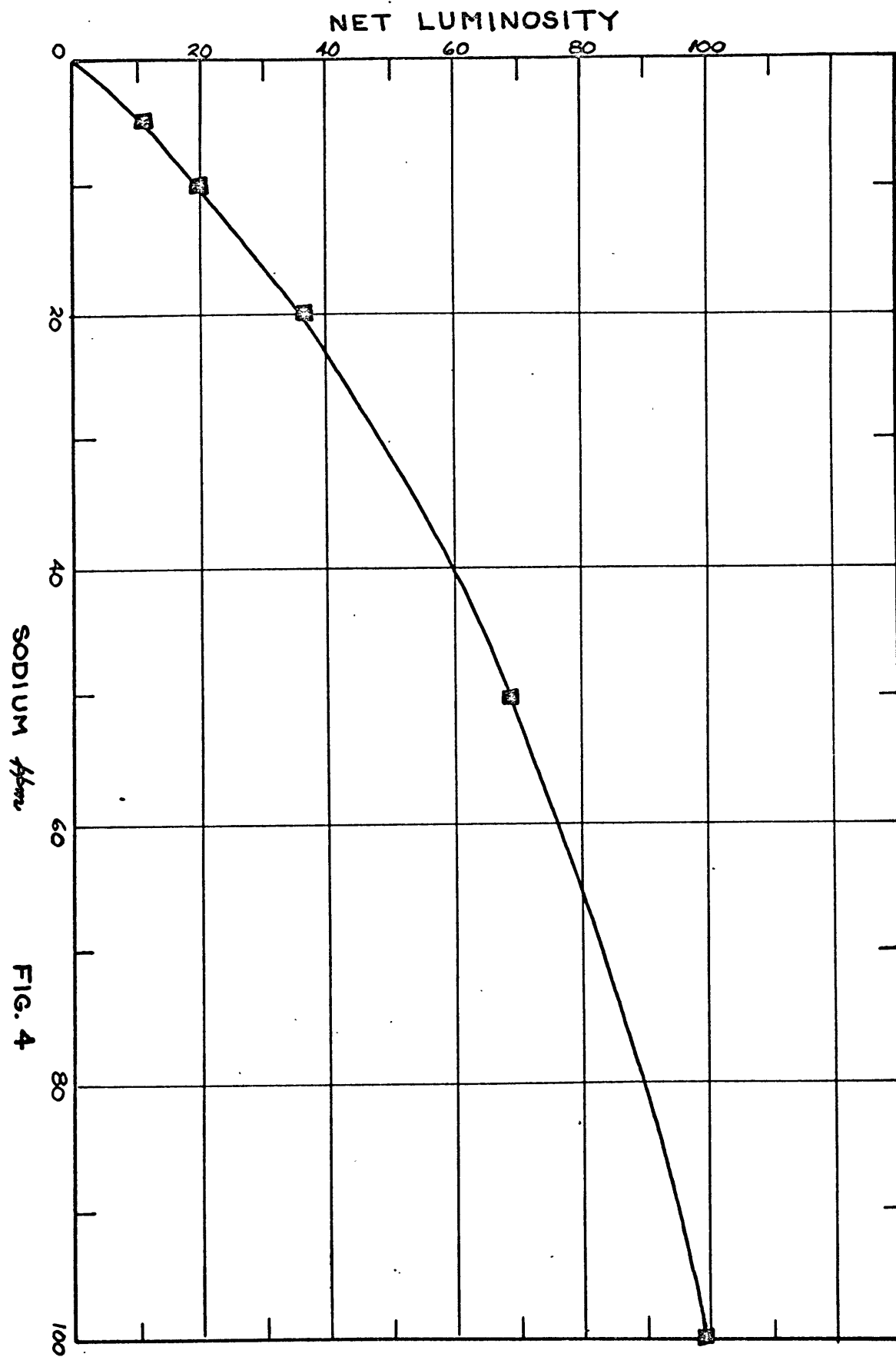


FIG. 4

of wire was washed. This was a simple matter because the dowels on the frames (Fig. 3) were 25 cm apart. By using five sections of wire, each consisting of a 20 cm central section and two borders I was able to accurately wash the desired length of wire. The drops of wash water were pipetted onto biological slides which had a depressed central region. These slides were treated with Dri-Film¹ which caused the drops to become hemispherical and have a contact angle with the glass of nearly 90°. This shape made the drops handy for the washing procedure. Each 20 cm section of wire was washed by gently pulling it through the drop. After all five sections were washed, the drop was analyzed for its sodium content.

There were no problems with surface tension as might be expected, or with any significant amount of salt or solution remaining on the wires. The wires were washed twice before analysis. As a control test, I washed several frames again and was unable to find even a trace of salt in these drops. Clean wires which were left in the box with the exposed frames were tested for any salt contamination with negative results each time. On several occasions when double samples were taken I analyzed one immediately and left the other in the sample box for a month or so before analyzing it. This was meant as a check against the possibility of some of the salt falling off the wires due to jarring, lower humidity inside the ship, etc. Fortunately,

¹ Dri-Film no. 9987, General Electric Co., Silicone Prod. Div., Boston, Massachusetts.

these tests were negative each time. This meant I could store a number of frames for several weeks before analyzing them on the spectrophotometer. This resulted in a significant saving of time and energy.

If the duration of the collection periods was properly planned, the samples gave a transmittance (essentially the brightness of the flame caused by the sodium ions) between 10 and 100. After subtracting the blank which was usually about 1, the sodium content of the wash drop could be read directly from the standard curves. For example, in figure 4 which is an actual standard curve used on one of the spectrophotometer runs, a net luminosity of 50 corresponds to a sodium content of 31.5 ppm. Knowing the volume of the drop and its sodium concentration the total amount of sodium on the wire could be calculated. Assuming that all the salts which were present in any significant amount were sea-salts, the total amount of sea-salt on the wires could be calculated by multiplying the value for sodium by a factor of 3.25. (Svedrup et al, 1942).

For a discussion of the reasonableness of this method refer to the following section. The number of meters of air which passed the wires was determined from the anemometer counter readings. Multiplied by the cross sectional area of the wires, this gave the volume of air swept out by the wires. The amount of sea-salt found on the wires was divided by the volume of air swept out. This gave the total sea-salt

content of the air in $\mu\text{g}/\text{m}^3$ (micrograms per cubic meter)
averaged over the exposure interval:

IV. CHEMICAL COMPOSITION OF THE PARTICLES

It will be seen in the next section that the collecting wires acted as a filter which essentially sampled only the so-called giant particles, i.e. those with a radius $> 1 \mu$. One of the main points of contention in the literature has centered around the actual chemical composition of the individual particles. Recent authors do not seem to doubt that the sea surface acts as an enormous source of both large and giant nuclei but there has been a vigorous debate as to whether these nuclei have the same chemical composition as sea water.

Woodcock and Gifford (1949) sampled those particles with (approximately) at 80% R.H. They found that the changes in radii with relative humidity which they observed with natural particles were very similar to those observed with known sea-salt nuclei. They also found that within reasonable limits the chlorinities obtained by their isopiestic method agreed with those obtained by titration of the salts on a collecting slide. Junge (1954) was able to show that the sodium and chloride contents of a coastal aerosol were limited almost entirely to the giant particles, i.e. that the sea-salt particles have a sharp limit somewhere around 0.8. In flights over southeastern Australia Twomey (1954) found that the vast majority of these particles in the giant size range were hygroscopic sea-salt nuclei. Some authors had predicted that the chemical composition of

these particles would change with time, the main effect being a loss of chloride. Twomey found no measurable change with time of the chloride content of the particles he sampled. Within the limits of his experimental method, Junge (1956) was able to establish that the sodium and chloride contents of individual giant particles were related as in sea water. Since NaCl is by far the main constituent of sea water (Svedrup et al, 1942), this seems to indicate that the giant particles are indeed sea-salt.

Junge (1957b) found that the giant particles collected in Hawaii consisted almost entirely of sea-salt nuclei and that only about 1% of the total mass of sea-salt in the air resided in those particles with radius less than 0.8μ . Wilson (1959) claimed that in freshly fallen New Zealand snows the K/Na ratio was about ten times its value in sea water. Even if his conclusions are valid, and there is reason to believe that they might not be, the error in the present calculations would be of the order of 10% if I assumed the sea-salt nuclei were of the same chemical composition as sea water. Oddie (1960) does feel there is some enrichment of K with respect to Na, but that the factor should be nearer 4 than 10. This would mean only a small error in the total salt content as calculated from ion ratios. In an interesting experimental investigation, Kombayasi (1964) determined the chemical composition of spray drops originating from various saline solutions. He found that for drops larger than 0.4μ

radius there was no difference between the drop concentrations and that of the parent solution.

Thus there is much evidence in the literature to support the assumption that those particles in the giant size range are composed of sea-salts in approximately the same ratios as in sea water. This is particularly true when the situation is thoroughly maritime and that this was the case in the present investigation is easily ascertained by examining the wind direction arrows in figure 5. Also, the vast majority of the total mass of sea-salt is found in the giant particles. Since the wires collected only the giant particles, I can compute the total amount of sea-salt in the air from the quantity of sodium found on the wires by means of the relevant ion ratio. This latter is about 3.25 for total salt to sodium. Therefore by multiplying the quantity of sodium found by 3.25 I obtain approximately the total amount of sea-salt in the air.

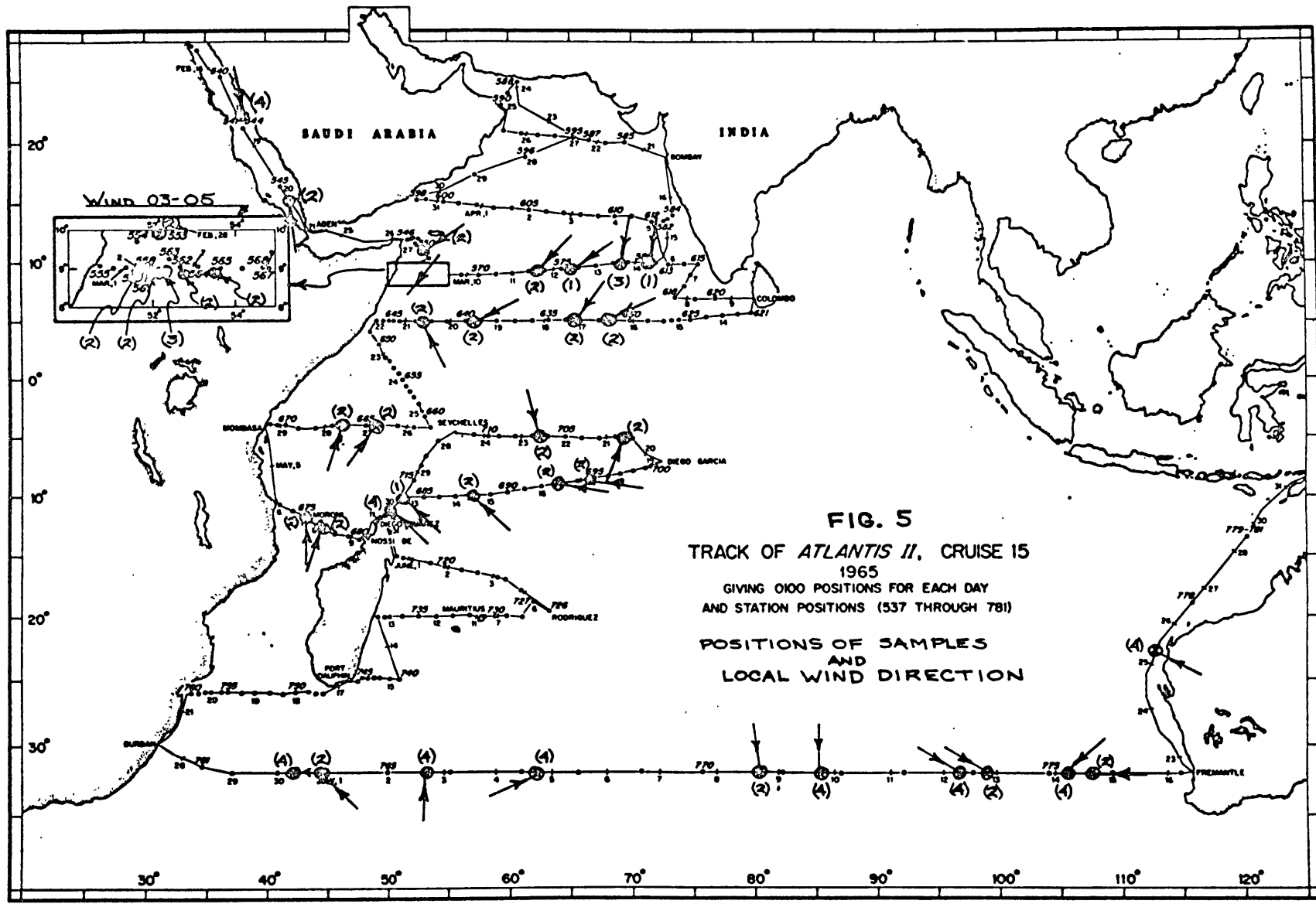


FIG. 5
TRACK OF ATLANTIS II, CRUISE 15
1965
 GIVING 0100 POSITIONS FOR EACH DAY
 AND STATION POSITIONS (537 THROUGH 781)
 POSITIONS OF SAMPLES
 AND
 LOCAL WIND DIRECTION

V. COLLECTION EFFICIENCIES AND OTHER SOURCES OF ERROR

The collection efficiencies of the wires were calculated according to the work of Langmuir and Boldgett (1946). Their work pertains to wind-driven particles which will stick to the wires upon contact. The question arises as to whether or not the sea-salt nuclei fulfill this latter criterion at the humidities observed during the collection periods.

It is well known that a crystal of pure NaCl (which makes up roughly 80% of the total sea-salt) undergoes a phase transition as the humidity is increased above approximately 78%. However, an interesting hysteresis was observed by Junge (1952). As the relative humidity was lowered he found that the solution remained supersaturated until the surprisingly low relative humidity of 30%. Woodcock and Gifford (1949) noted that in natural sea-salt particles there exist certain highly soluble salts such as $MgCl_2$ and $CaCl_2$ which remain in solution at humidities as low as 30-35%. They concluded that sea-salt nuclei would adhere to their collecting slides at humidities far below 70%. They also conducted shattering tests to see if any of the particles broke up and were consequently lost during impact. At humidities as low as 43% and wind speeds of up to 42 m/sec no shattering was observed. Junge (1954) was also able to show that the particles remained "sticky" down to humidities of 40-50%. Blanchard and Spencer (1964) observed that the more soluble salts formed a solution which appeared to envelop the NaCl "crystal." They also found (at least under laboratory conditions) that the supersaturation of saline drops was commonplace and extreme. Since the humidities I observed were all over 55% and the vast majority were between 65-80% there should be no difficulty in assuming that the sea-salt nuclei would stick to the wires upon impact. This allows the use of the Langmuir and Boldgett (1946) collection efficiencies for small cylinders, which is what the wires are.

In their paper Langmuir and Blodgett presented the collection efficiencies as a function of two dimensionless parameters, ϕ and K , which are defined by:

$$\phi = 18 \left[\frac{\rho_a^2 C U}{\gamma \rho_s} \right] \quad K = \frac{2}{9} \left[\frac{\rho_s a^2 U}{\gamma C} \right]$$

The following list defines the symbols in the above expressions and gives the values used in the calculations.

- (1) ρ_a air density. Since the air temperature was often 20-25°C and the atmospheric pressure was approximately standard (760 mm Hg) a value of $1.2 \times 10^{-3} \text{ g/cm}^3$ was used for ρ_a
- (2) C radius of the collecting wire. In the present case this was $7.6 \times 10^{-3} \text{ cm}$
- (3) U wind speed. Almost all readings were made with a vector wind speed of at least 8 m/sec. Since a few were lower the collection efficiencies were evaluated for 4 m/sec and 8 m/sec.
- (4) γ air viscosity. With conditions as stated in (1), taken as $1.85 \times 10^{-4} \text{ g/cm sec}$
- (5) a radius of the impinging sea-salt nuclei. Plotted as the abscissa with collection efficiency E as the ordinate (Fig. 7). Values of $0.5 - 30.0 \mu$ were used in order to cover the giant particles.
- (6) ρ_s density of the impinging sea-salt nuclei. This will vary with the humidity. Two extreme values corresponding to relative humidities of 60% and 90% were used. These are 1.27 and 1.10 g/cm^3 respectively for NaCl solutions which will be

close to the values for a sea water solution.

Reference to table 2 and figure 6 will quickly show that for practical purposes ϕ may be considered zero throughout the determination. With this assumption, the values of E (collection efficiency) were determined as a function of drop size, relative humidity, and wind speed. They were read from figure 6 for $\phi = 0$ and variable K, the latter calculated by the equation above. Since the effect of relative humidity is very small, a single value of 80% is used because of its frequent occurrence in the literature. In figure 7 the collection efficiency is plotted as a function of the droplet radius for the two different wind velocities employed in the calculations. As was noted earlier, at low wind speeds the salt was collected while the ship was underway in order to increase the collection efficiency, the result being that nearly all the samples were obtained with a relative (vector) wind velocity of 7-8 m/sec or higher. The curve for a relative wind of 4 m/sec is included to indicate a lower limit of collection efficiency although very few samples were taken at this lower speed. In general, reference should be made to the 8 m/sec curve.

Since I am concerned only with the total sea-salt content of the air and the mass of the sea-salt nuclei is almost exclusively concentrated in the giant particles (Junge, 1957), I need to calculate the efficiency of mass collection only for those particles with a radius $> 1.0 \mu$. This will

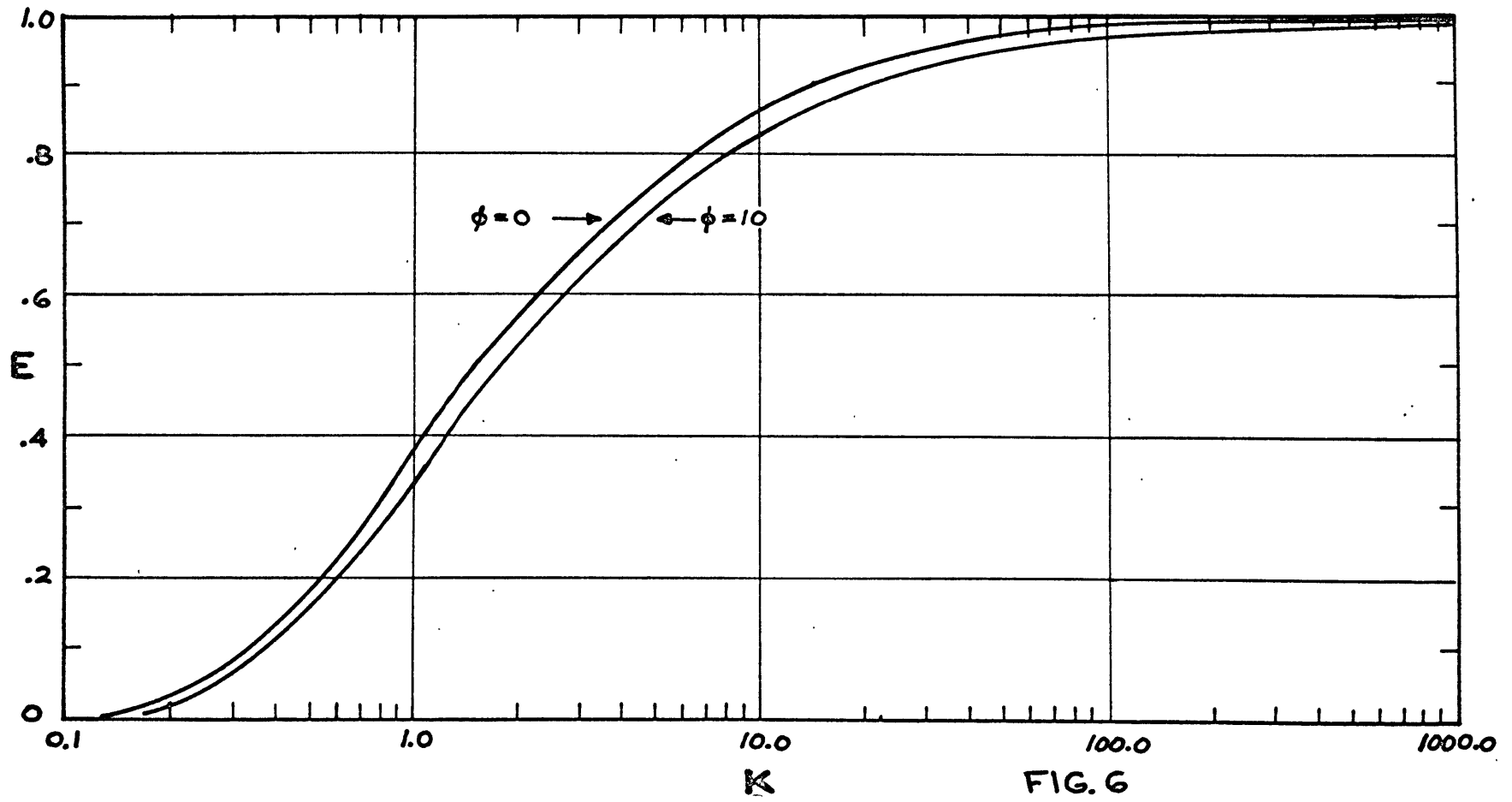


FIG. 6

TABLE 1

a, DROP RADIUS, μ	U = 4 m/sec.				U = 8 m/sec.			
	R.H. = 60%		R.H. = 90%		R.H. = 60%		R.H. = 90%	
	K	E	K	E	K	E	K	E
0.5	0.20	.02	0.18	.02	0.40	.13	0.36	.10
1.0	0.80	.33	0.69	.29	1.6	.51	1.4	.47
2.0	3.2	.67	2.7	.64	6.4	.80	5.4	.77
3.0	7.1	.83	6.2	.80	14.0	.91	12.0	.89
4.0	13.0	.90	11.0	.88	26.0	.95	22.0	.94
5.0	20.0	.93	18.0	.92	40.0	.96	36.0	.96
6.0	29.0	.95	25.0	.94	58.0	.97	50.0	.97
8.0	51.0	.97	44.0	.97	100.0	.98	88.0	.98
10.0	80.0	.98	69.0	.98	160.0	.99	140.0	.99
20.0	320.0	.99	270.0	.99	640.0	~1.00	270.0	~1.00
30.0	710.0	~1.00	630.0	~1.00	1400.0	~1.00	1200.0	~1.00

TABLE 2

R.H. %	U = 4 m/sec.	U = 8 m/sec.
60	0.33	0.66
90	0.38	0.76

} ϕ

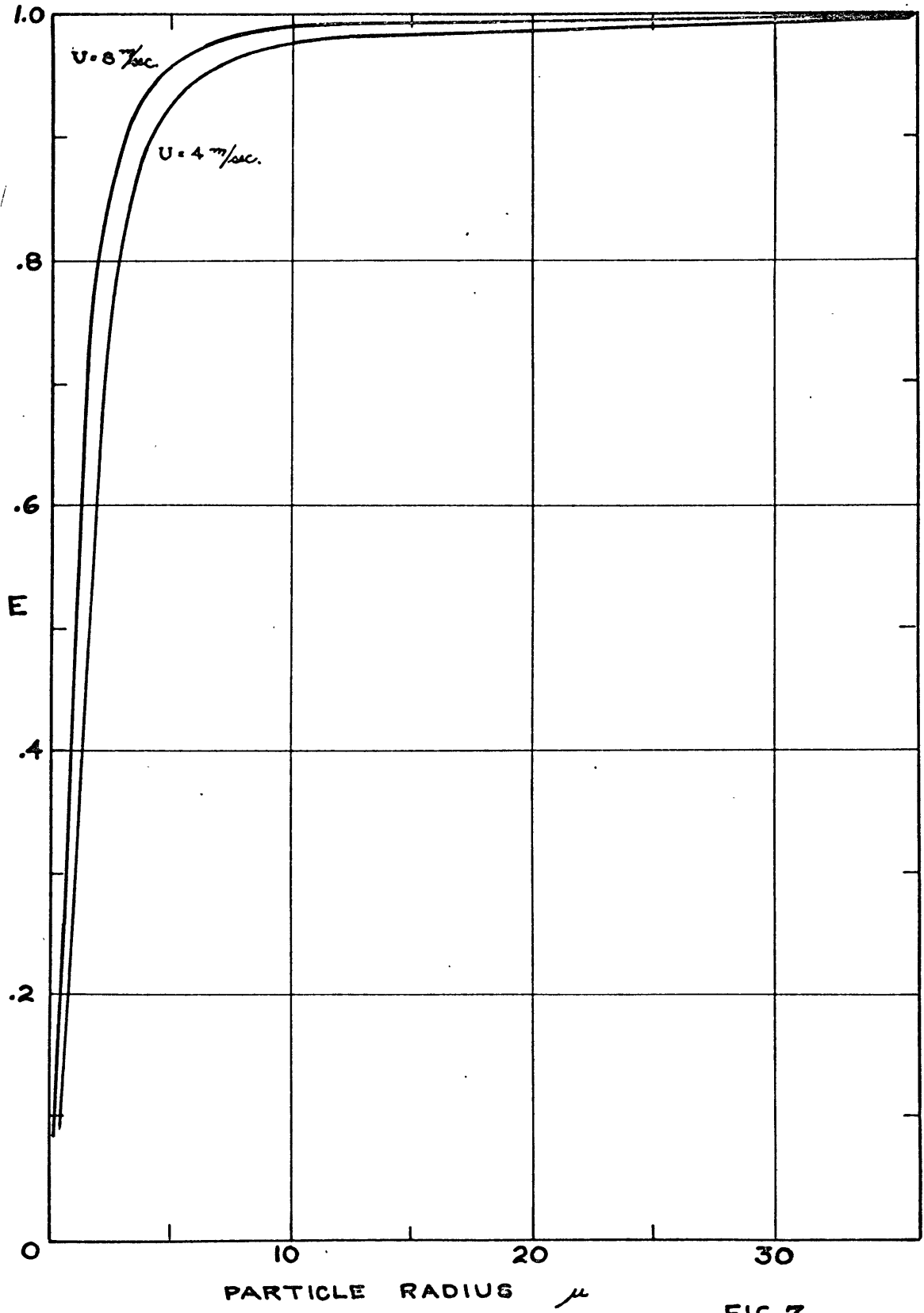


FIG. 7

essentially give the efficiency with which the total mass of sea-salt is sampled. The mass distribution curves of Woodcock and Gifford (1949) and Woodcock (1953) were used in conjunction with the efficiencies plotted in figure 7. Numerical integrations showed that for all wind speeds encountered the wires collected 90% (+10%) of the total amount of sea salt in the air. Except for the very low wind speeds, i.e. those below 5 kts, the percent error does not exceed 5 or 6%. Thus +10% is a conservative, maximum figure and only applies to certain limited cases whereas +5% is in general more realistic. Multiplying all the observed values of sea-salt by the factor 1.1 will give the actual total sea-salt content of the air corrected for collection efficiency. The maximum error in this result is +10%.

There are of course a number of sources of error in addition to the collection efficiencies. The possibility that salt fell off the wires while they were being stored prior to analysis was previously discussed and shown to be unimportant. Errors in the anemometer counter readings before and after a collection period combined with uncertainties in the anemometer calibrations are not expected to exceed 1%. After washing, no salt remained on the wires, but an error of roughly 1% is to be expected in the washing procedure due mostly to the impossibility of washing exactly 1 meter of wire. The total error resulting from the standardization procedure, the operation of the spectrophotometer, and the

graphical conversion method is estimated to not exceed 3%.

(The percentages apply to the actual total sea-salt content as measured in $\mu\text{g}/\text{m}^3$.) Therefore for winds greater than 5 kts. the total error from all sources will not exceed 10% while the total error for lighter winds may be as high as 15%.

VI. RESULTS AND DISCUSSIONS

(a) The Relationship Between Sea-Salt Content and Wind Velocity

As a result of the preceding arguments, it can be stated that the data I collected do indeed represent the total sea-salt concentration of the marine atmosphere at the collecting level of 10 meters. The maximum errors are small enough to be disregarded in the following discussion. The vast majority of the data were collected in the western and southern portions of the Indian Ocean. The rest are from the Mediterranean Sea, Suez Canal and Red Sea. The Suez Canal data are interesting in that they indicate there is significant inland transport of the sea-salt even at the lower levels. Certain data taken in the North Atlantic while the equipment was being tested and a few values (3) which showed wide discrepancies between two simultaneous observations are not included due to the possibility of error.

The main purpose of the investigation was to attempt a confirmation of the general relationship thought to exist between the total amount of sea-salt in the maritime atmosphere and the near-surface wind velocities. This relationship has been discussed by several authors, among them Woodcock (1953) and Junge (1958).

To this end all data have been plotted in figures 8-11 irregardless of geographical position. The first graph (Fig. 8) gives the amount of sea-salt in $\mu\text{g}/\text{m}^3$ (or equivalently g/m^3)

versus the wind velocity at the time of observation. The next three graphs (Figs. 9-11) give the sea-salt content versus the average wind velocities for periods of six, twelve, and twenty-four hours preceeding the observations. During certain intervals the ship travelled a considerable distance, but because of the spatial uniformity of maritime conditions the effect on the average winds for the longer time intervals is not considered to be great.

Upon studying the graphs it becomes immediately obvious that there are two different regimes. Above a wind speed of 8-10 kts the values show a very definite increase with increasing wind speed. At a particular wind speed they vary by a factor of two to three. Below 8-10 kts where some sort of discontinuity is in evidence, the relationship of the total sea-salt to the wind velocity becomes uncertain. Here values range over an order of magnitude at a given wind speed and some very low values occur.

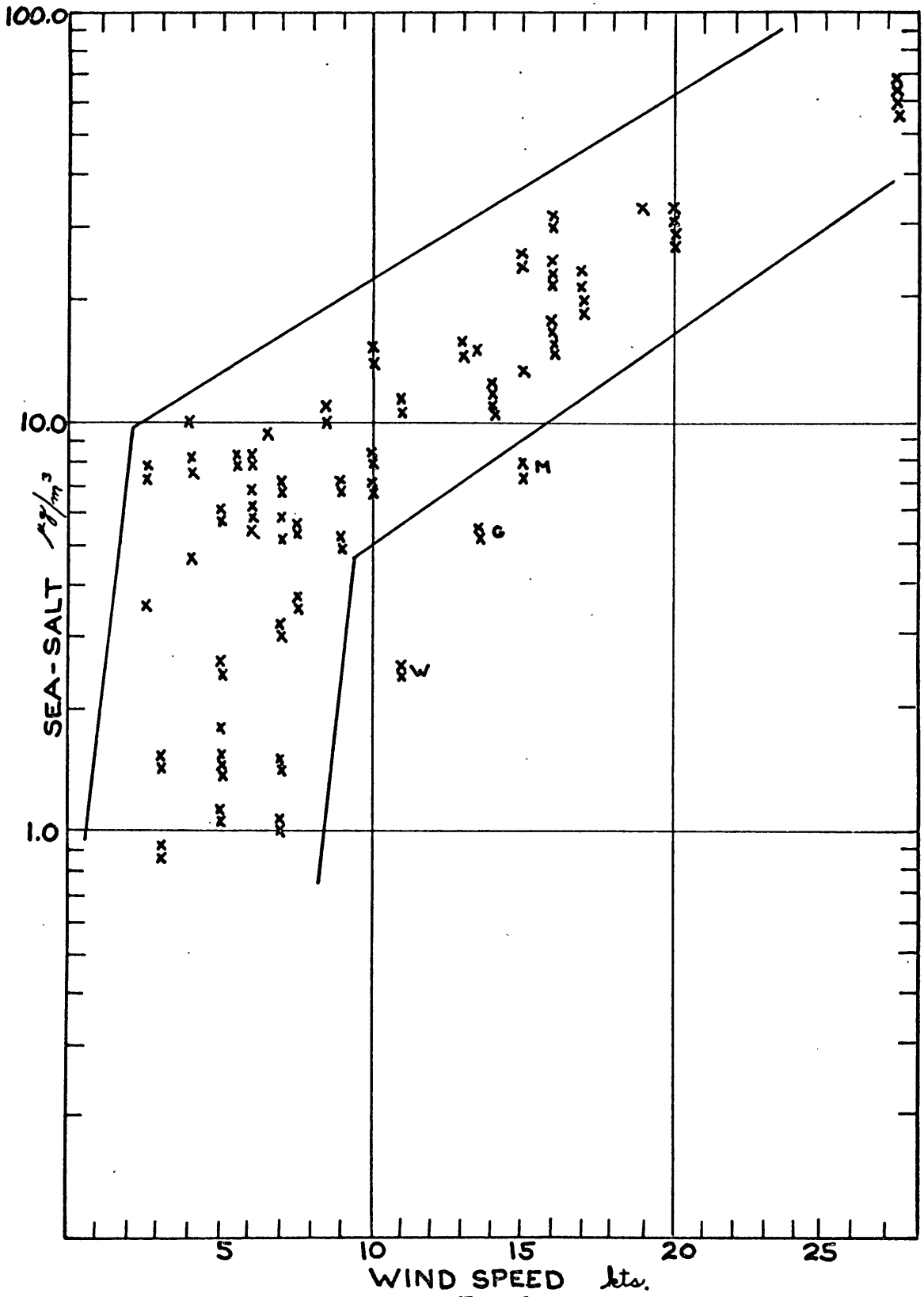
The data present yet another piece of evidence for the apparent discontinuity in sea-surface and low-level phenomenon noted by Woodcock (1940), Svedrup (1946) and Munk (1947), among others. These authors found apparently discontinuous changes in various parameters as the wind speed increased above force 3, Beaufort, i.e. above approximately 10-12 kts. Since it is quite apparent that the sea-salt results from breaking waves and more particularly from the bubbles produced by the "whitecaps," (Keentzler et al, 1954; Blanchard

and Woodcock, 1957) it is to be expected that local production of sea-salt will begin when whitecaps are first formed. This production occurs whenever the wind exceeds a certain critical value. Munk (1947) found no foam patches at a wind speed of 9-10 kts and 10-15 per 10^4 m^2 for winds exceeding 13 kts. Blanchard (1963) found that whitecaps began to form when the wind reached 7-8 kts. On the basis of these results, local production of sea salt should begin somewhere in the vicinity of 8-10 kts. This appears to be just the case in the data presented in figures 8-11. An important conclusion is that when no local production occurs (wind speed below 8-10 kts) the factors of sedimentation, advection (from areas of active production), instability (see later) and washout become much more determinative on the sea-salt concentration than the local wind speed. This accounts for the wide range of values which occurs below the discontinuity.

Note that several pairs of values are not included within the lines which I drew to emphasize the two drastically different regimes. The first pair, labelled "M," were taken in the harbor of Moroni island (fig. 2) where a local effect caused high gusty winds which were not indicative of the general conditions at the time. Since the fetch was but a mile or so, the values represent those found over the adjacent waters with their lower wind speeds. The pair labelled "G" were taken in the Great Bitter Lake area of the

Suez Canal (just off the map north of the Red Sea) immediately following a local increase in wind which again resulted in no significant production. The wind near Port Said had been onshore and fairly strong the previous day. The sea-salt in the air over the canal and the lake was probably blown inland from the Mediterranean Sea and not locally produced. The third pair of points, labelled W, represents a case where the wind had just risen to about 11 kts after 18 hours of 6-7 kts. This becomes apparent in the graphs representing the salt content versus the average winds over the various time intervals.

In figure 12 I have compared the boundary lines which I felt were applicable to the data with those which were used by Woodcock (1953) and cited in Junge (1958). (Also see Fig. 1) These latter boundaries represent the range of data gathered by Fournier d'Albe (1951), Moore (1952), Junge (1953), and Woodcock (1953). Woodcock's data were generally taken at or near cloud base level, that of the other observers essentially a few meters above the sea surface. The general agreement in slope above the discontinuity is quite close and shows the same logarithmic increase of salt content with wind speed. The values of Fournier d'Abbe and Moore which occur above the discontinuity are found in or near the lower part of the range I observed. This is significant because they were also taken near sea level. Junge's data were collected at the seashore in southeastern Massachusetts



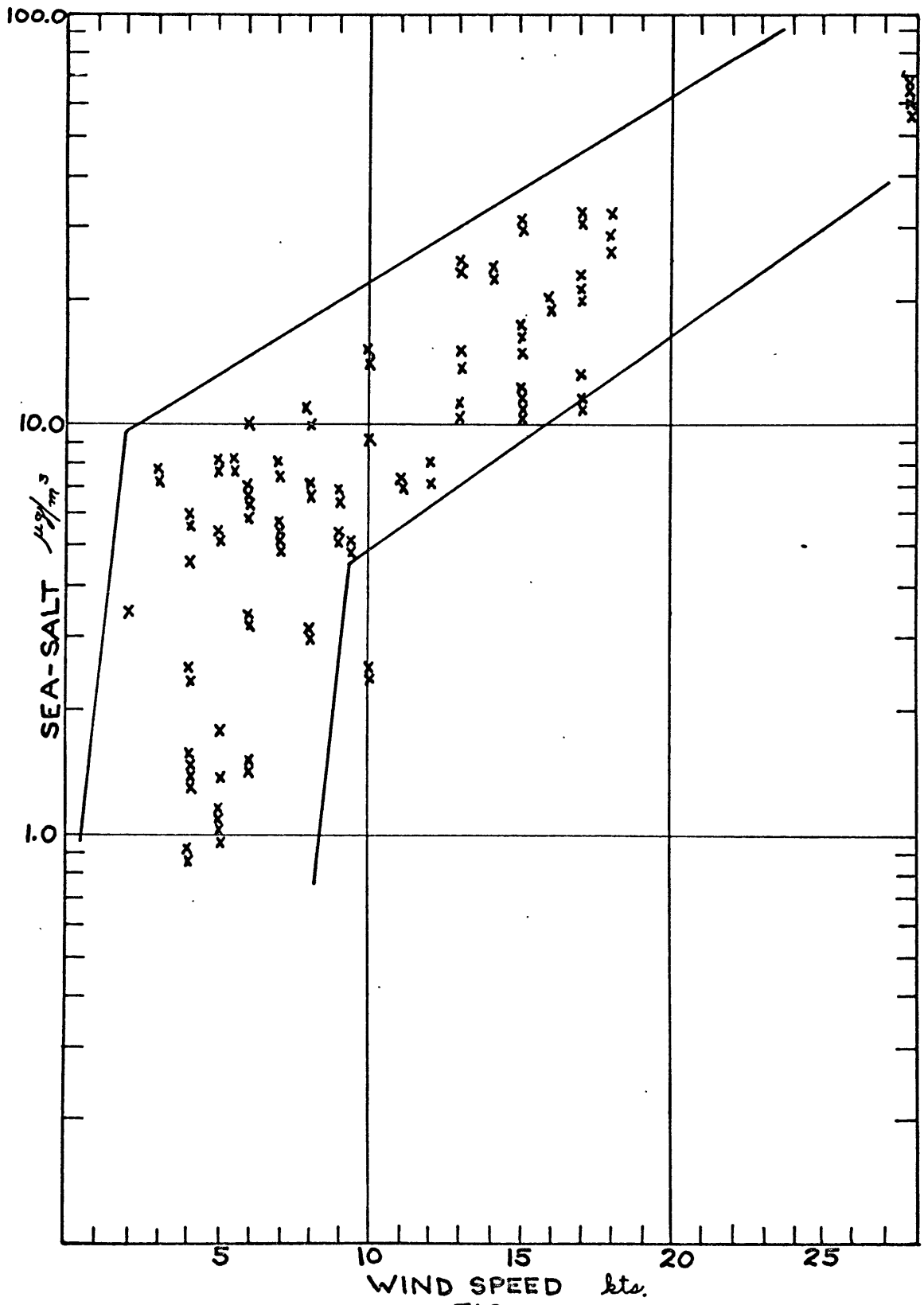


FIG. 9

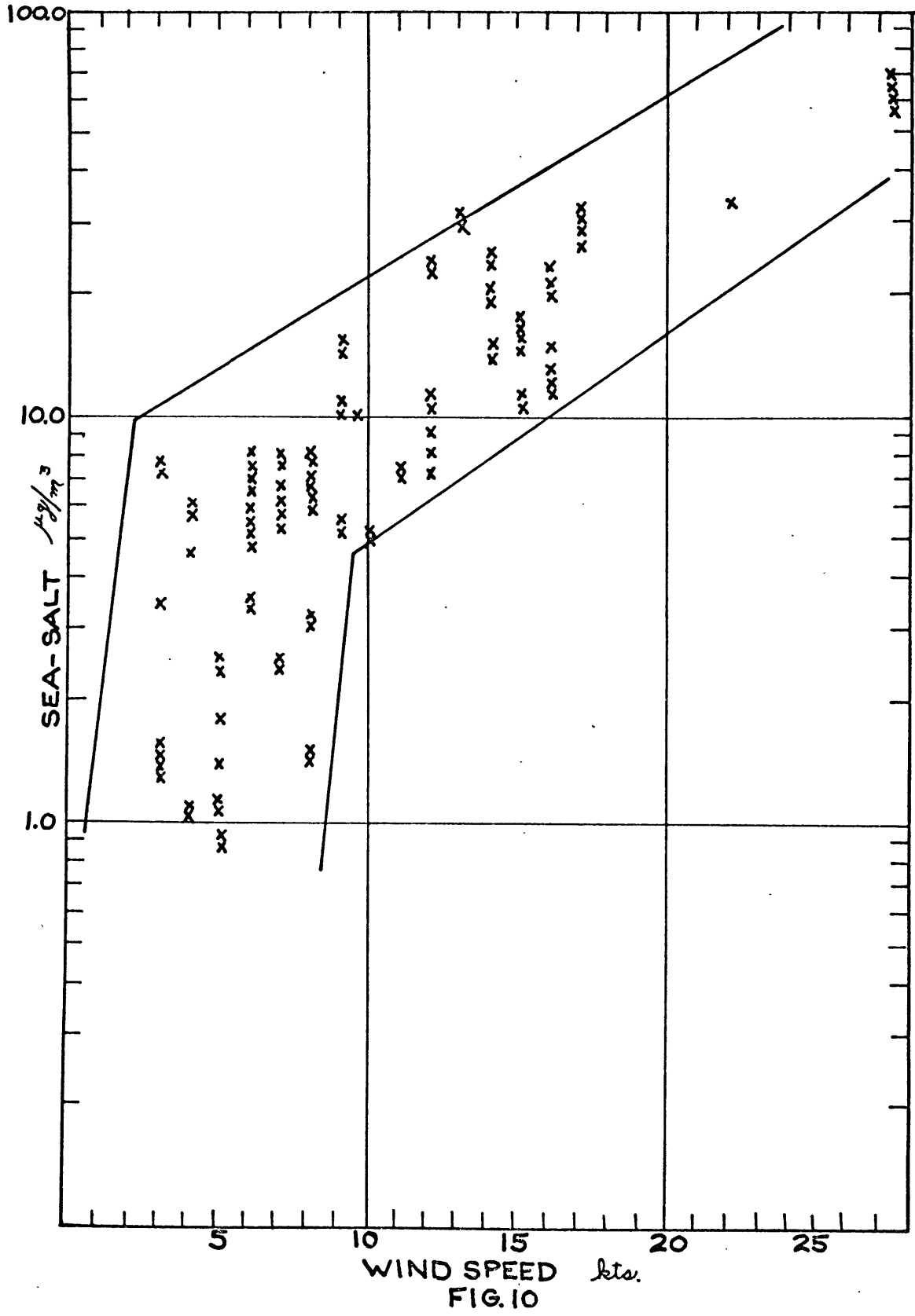


FIG. 10

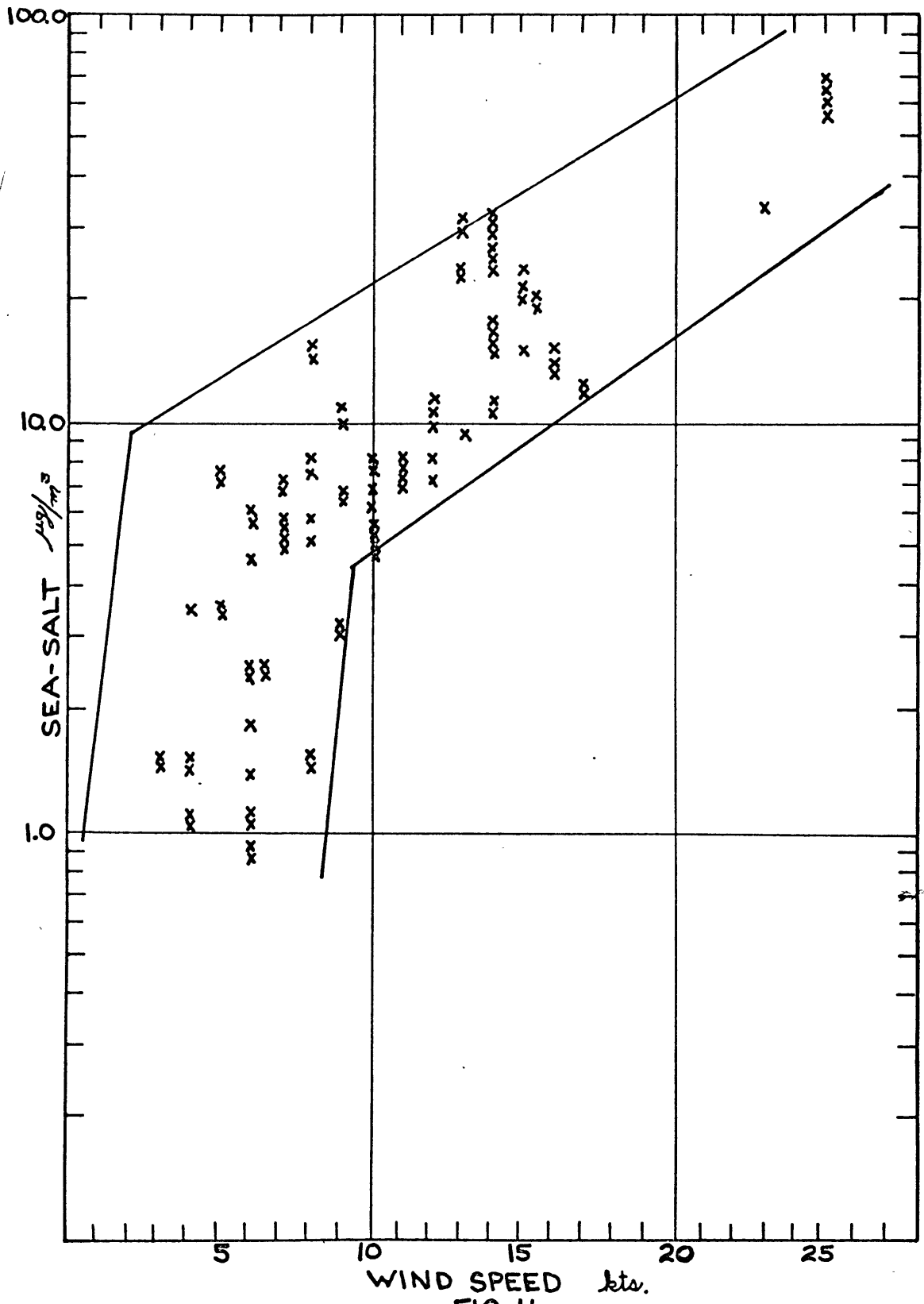


FIG. 11

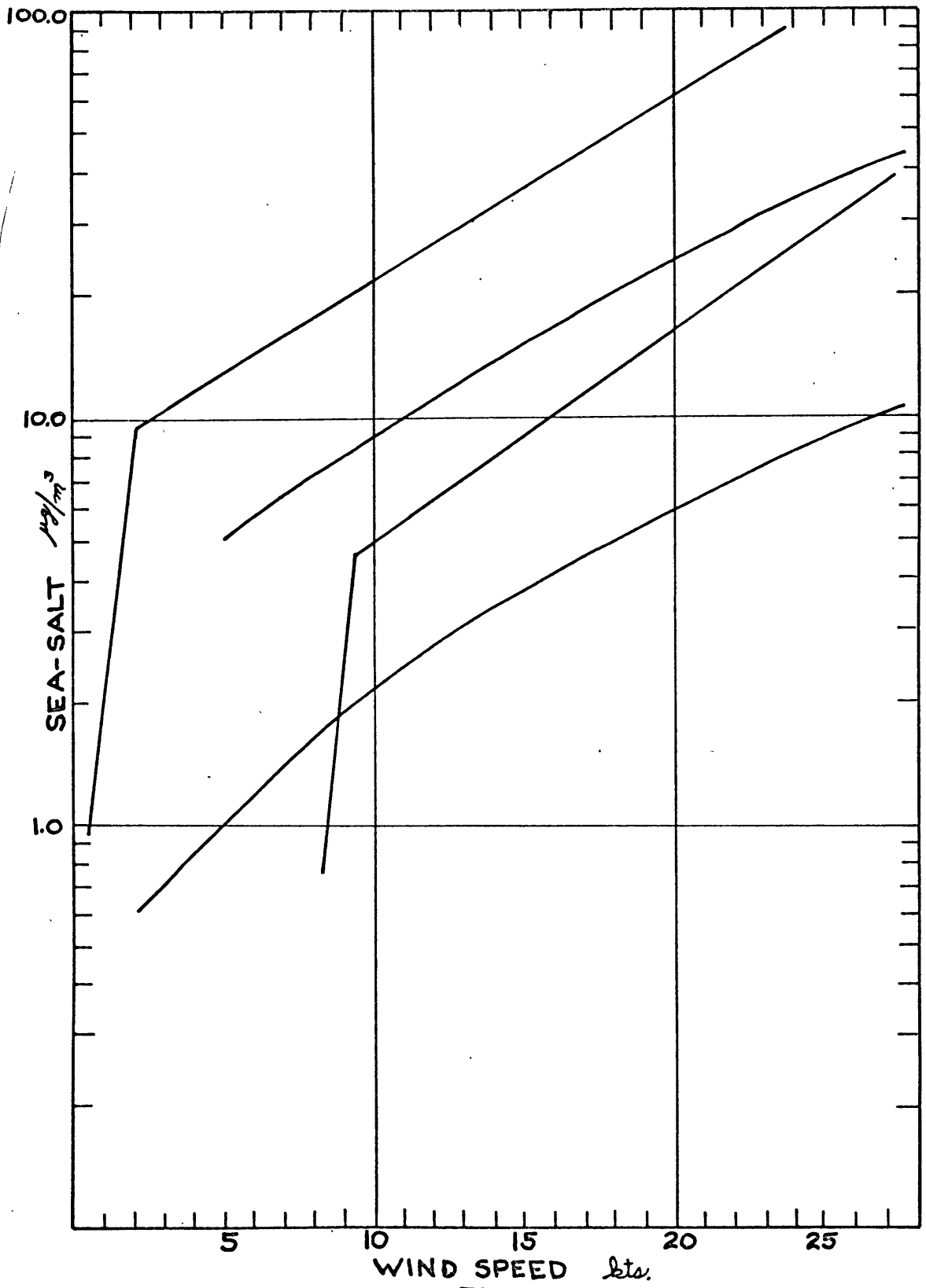


FIG.12

and cannot be considered typical of wholly maritime conditions although he was careful enough to only cite his higher values (Fig. 1). These were certainly closer to what would be expected under maritime conditions. As mentioned above, Woodcock's data was generally taken just below cloud base. Since he finds (Woodcock and Gifford, 1949) that at a level of 15 m the total salt content is roughly double that at cloud base the two "curves" representing the ranges are seen to be in excellent agreement above the discontinuity at 8-10 kts. More than likely, the presence of a few very large particles which never get carried aloft due to prohibitive sedimentation rates causes the increase in the total sea-salt content at the lowest levels. For winds below 10 kts I have many more data points than were presented by the other authors and therefore feel that the range curves I drew in figures 8-12 are more indicative of the real situation than those presented in figure 1 and repeated with a change of scale in figure 12 (Junge, 1958). There just is no very clear relationship between sea-salt and wind speed for the lighter winds. An upper limit is perhaps feasible for the values of sea-salt in light winds but no empirical expression would be valid or useful.

This is not the case with the higher winds, i.e. those greater than 8-10 kts. For this case an empirical formula can readily be derived which will give the range of salt content at a given wind speed. This formula can be obtained

from the range "curves" (Fig. 12). Thus as an approximation to the total sea-salt content I obtain:

$$\text{Sea-Salt}(\mu\text{g}/\text{m}^3) = (2 \text{ to } 8) \times e^{0.1W}$$

where W is the wind speed in knots. For more limited geographical areas the range of (2 to 8) can be narrowed somewhat.

During the period February 27 to March 14, 1965 the R.V. Atlantis II was occupying hydrographic and current stations between the Somali Coast and southern India (Fig. 2). Since the passage was roughly along the latitude circle 10°N , and because of the season, the northeast monsoon winds were present (Fig. 5). During the entire period no showers were observed either at the ship or on the horizon. The typical cloud cover was several tenths of undeveloped cumulus, the so-called "fair weather cumulus." Because of these steady conditions, the sea-salt data fall within a narrower range, as may be noticed by referring to figure 13. Values for the period fall within the limits drawn and increase with increasing wind speed according to the empirical relation derived above. The difference is that the relationship may now be written with a narrower range. Therefore, approximately

$$\text{Sea-Salt}(\mu\text{g}/\text{m}^3) = (3 \text{ to } 7) \times e^{0.1W}$$

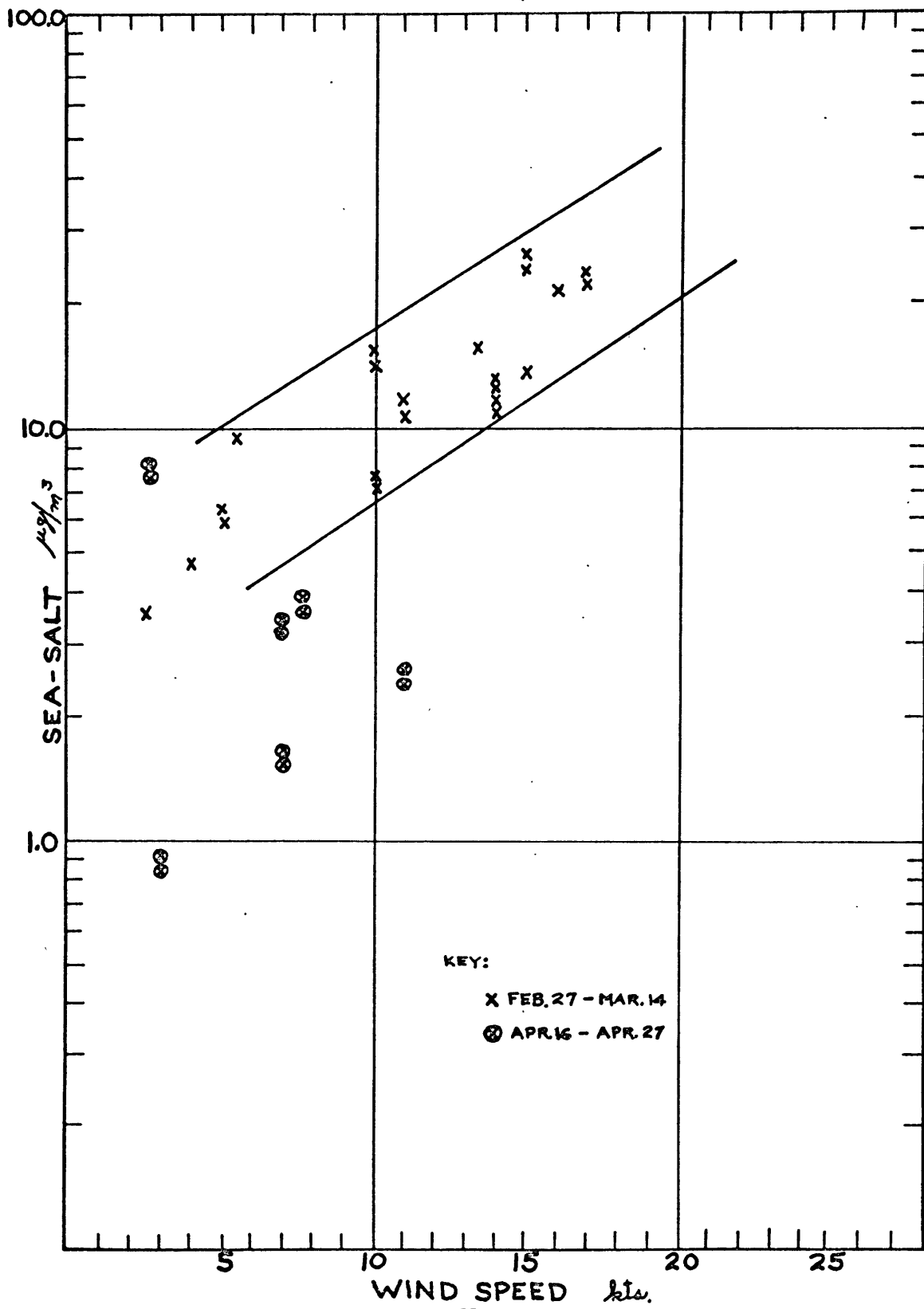


FIG. 13

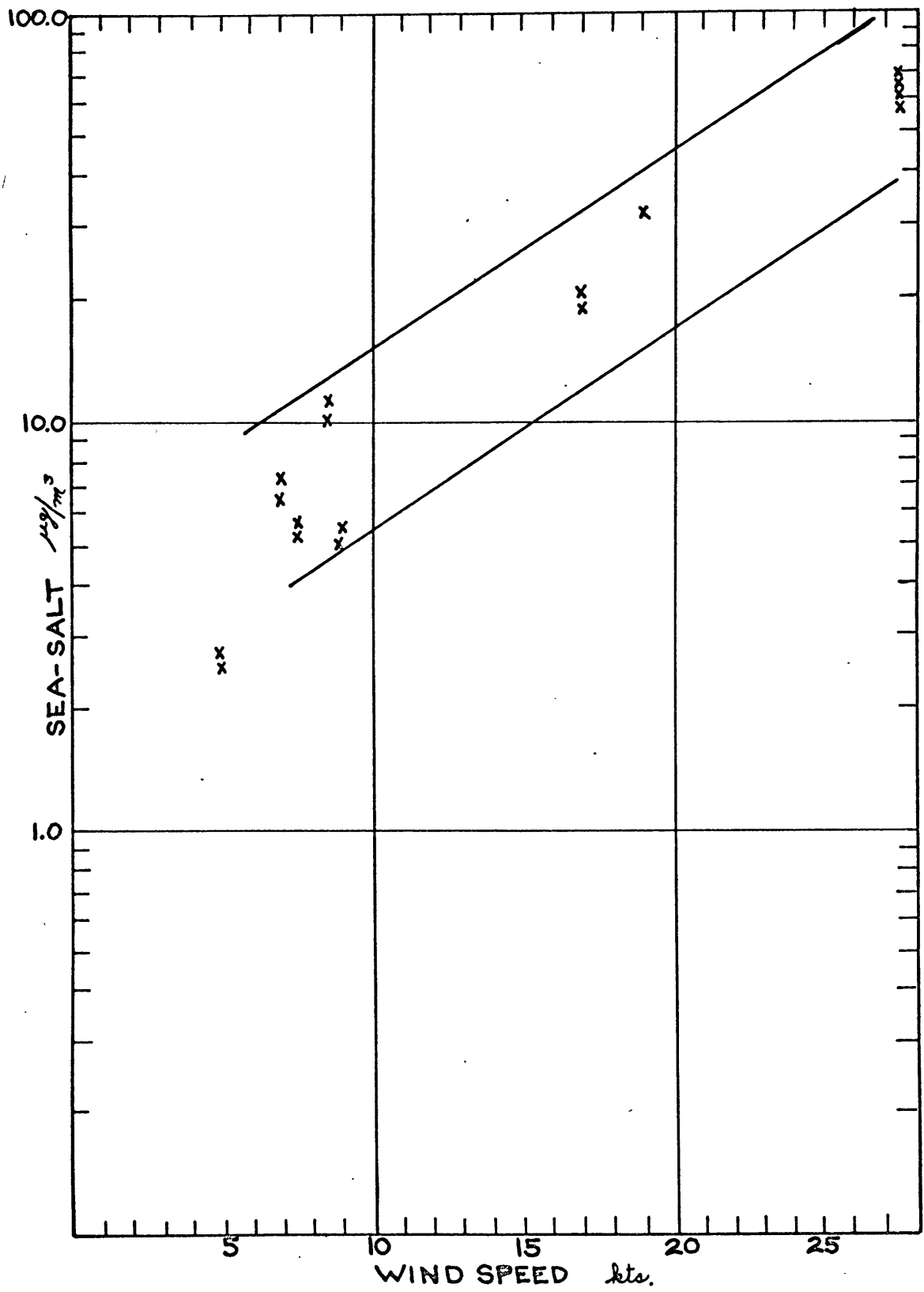
where as before, W is the wind speed in knots. It becomes apparent that even with quite steady conditions and the absence of washout values may vary by a factor of two between one observation and the next, even though the local wind speed may remain unchanged. Since the collection method results in an averaging over a fairly large parcel of air, often ten to fifteen kilometers in length, this means the lower atmosphere must be less homogeneous with respect to large parcels than one might intuitively expect. This same result was mentioned by Toba (1965a).

Also plotted in figure 13 are six pairs of values obtained during the period April 16 to April 27, 1965. Although two pairs were taken in an area somewhat remote from the others, they are meteorologically similar and hence grouped together. This was the period when the monsoon was in the process of changing from northeast to southwest (Fig. 5) and the weather was hot, humid and squally with generally light winds. There were many showers both at the ship and on the horizon for most of the period. The effect of low production and washout is immediately obvious. The values are much more scattered, and except for one aberrant pair for which no explanation is available, all are significantly (by a factor of 2-4) lower than the values recorded during the dry northeast monsoon. Although it would be unwise to draw any strict quantitative conclusions from this small amount of data, the effect of washout is qualitatively well illustrated, and it

is seen to significantly effect the total sea-salt budget. This process was noted by Lodge (1955) although he found a stronger decrease, nearly an order of magnitude, for the giant particles he measured before and after a local shower.

The period May 8 to May 22, 1965 was marked by generally south or southeast winds with only minor exceptions (Fig. 5). Several days of relatively high winds were experienced. Although there were some rain squalls, with the exception of one day they occurred for quite limited periods and were nowhere near as prevalent as during the monsoon reversal discussed above. The range is slightly lower overall (Fig. 14) but the slope of the sea-salt "curves" is essentially identical to that found during the dry northeast monsoon (Fig. 13). It is possible that the slightly lower values do represent a small washout effect, but the data are not numerous enough to quantitatively support this idea. For the data garnered during both monsoons an empirical expression of the form discussed above in relation to figure 13. gives a reasonable approximation to the total sea-salt content.

During the latter part of June and the first two weeks of July, 1965, the R.V. Atlantis II made a crossing of the southern Indian Ocean along the latitude circle 32°S . Data gathered on this crossing as well as four points from the vicinity of the western coast of Australia (Fig. 5) are presented in figure 15. The winds were on the whole anomalously



light for a winter crossing at that latitude. The barometer was high (1030-1035 mb) for almost the entire crossing and the only rain consisted of a few light showers on several days during the middle of the passage. It appears that a high pressure cell was drifting across the ocean at roughly the same rate as the ship. The cloud cover was often stratocumulous with some undeveloped cumulus and the sea was remarkably still. The situation was the anti-thesis of the stormy conditions expected. Notwithstanding these exceptional conditions, all data collected with wind above 8 kts (and some below this) fit within the range "curves" sketched for the dry northeast monsoon and the same empirical expression appears valid.

Since there were many days of low winds, the discontinuous nature of the data is particularly obvious in figure 15. Below 8-10 kts the sea-salt content is not related in any obvious way to the wind speed. At 4 kts (local wind speed) the sea-salt content is more than an order of magnitude higher than it is at 7 kts.

The nature of the "discontinuity" must not be misconstrued. It is not particularly sharp and the lines drawn on the earlier plots (Figs. 8-12) are meant as aids to the eye and are suggestive rather than definitive. There is, however, no doubt that there are two different "regimes," as it were, in the data collected. These regimes are closely related to the presence or absence of local production. When there

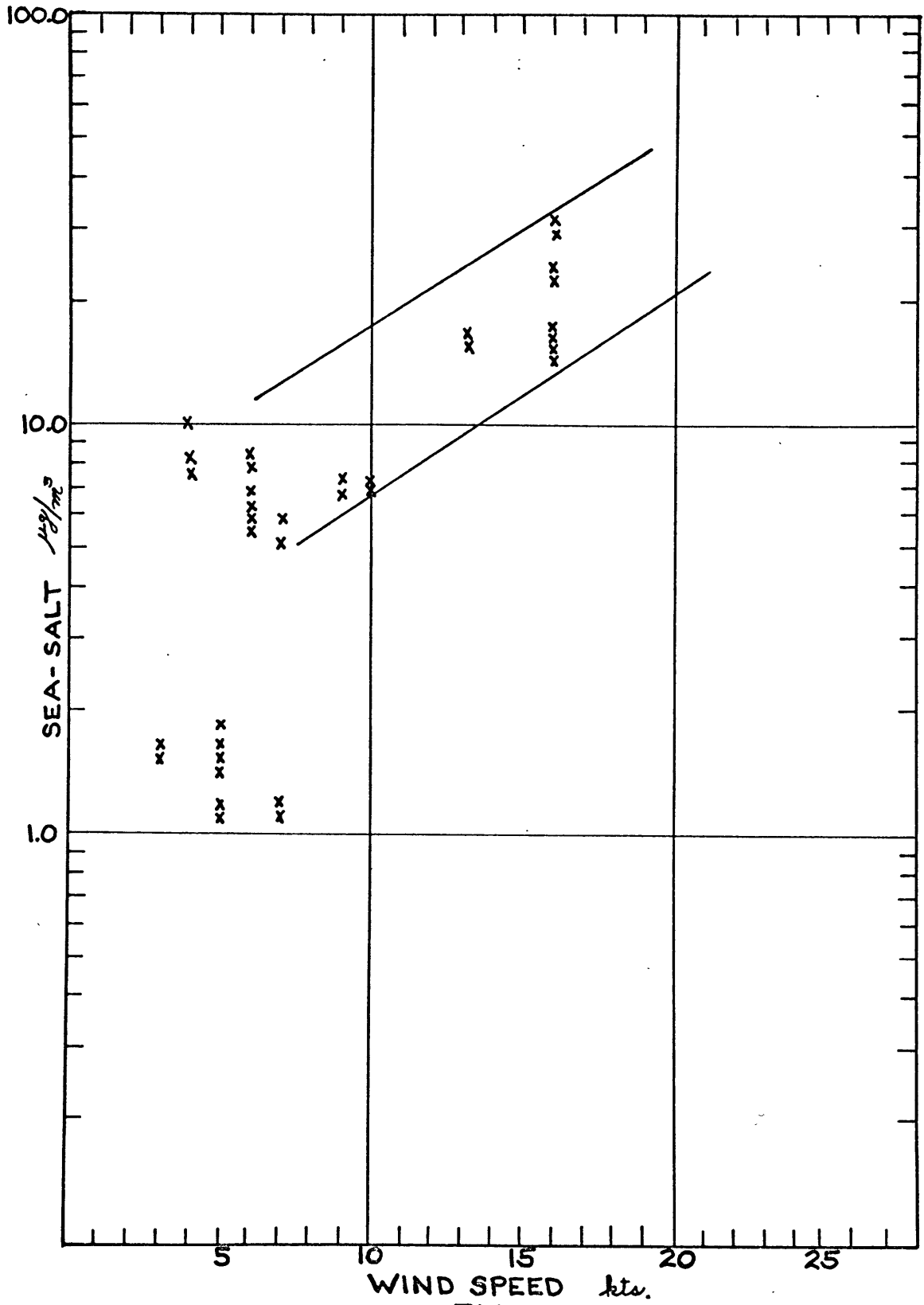


FIG. 15

is no local production, the combined effects of advection, sedimentation, washout and instability are decisive in determining the sea-salt content of the air. Very scattered values are noted during showery weather, due no doubt to the patchy nature of the washout. During general rains I would expect the depletion of the sea-salt to be more consistent over a large area.

Advection brings in at least the remnants of aerosols formed in areas of higher production and may cause values which seem inordinate for the local wind speed. Sedimentation like washout, tends to lower the salt content, although it acts preferentially on the larger particles where the greater amount of mass is found. Instability, which is rarely considered a significant factor in reference to salt content, will be shown to have a strong influence in a following section.

As a result of the not well understood interactions of these four parameters the salt content at wind speeds below those necessary for local production is distributed chaotically with respect to wind speed. When local production begins, and this occurs when the wind reaches about 8 kts., the more random effects of the four parameters are masked and the sea-salt content becomes a fairly clear-cut function of the wind speed. This then is the nature of the discontinuity: below it several factors act in a manner which is poorly understood quantitatively and scatter the sea-salt

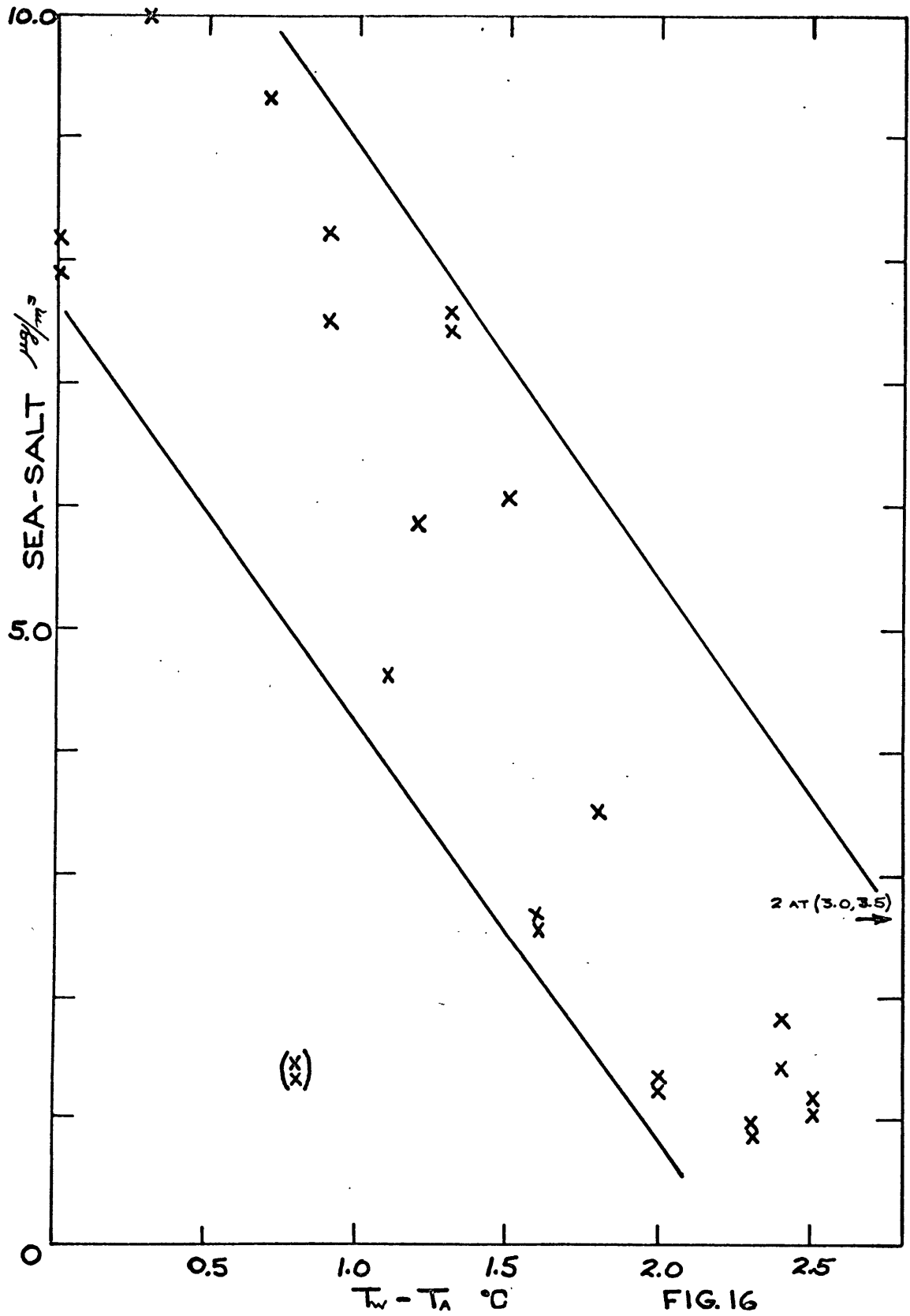
content; above it a single parameter, wind speed, becomes determinative, at least when general rains are absent.

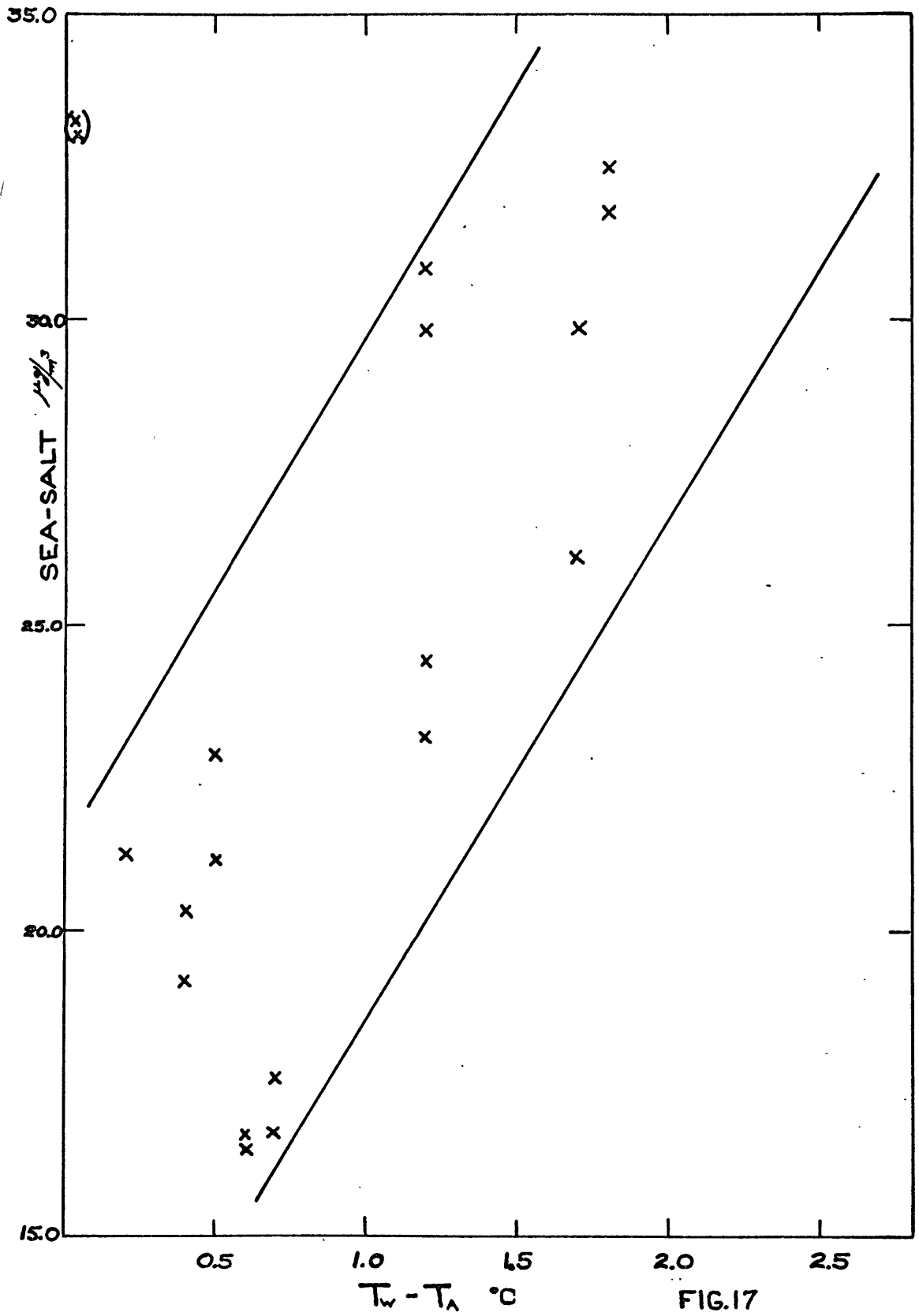
RESULTS AND DISCUSSION

b. The Relationship Between Sea Salt Content
and Low Level Instability

Over the oceans the difference between the surface temperature of the water and that of the overlying air can be conveniently used as a first-order indication of stability. The larger the difference ($T_W - T_A$) the greater the instability. Negative values of the difference indicate stable conditions. Two strong correlations were found between the total amount of sea-salt and the degree of instability. For light winds (Fig. 16) when there was no local production the mass of salt in the air at the 10 m level decreased quite rapidly with increasing instability. For strong winds (Fig. 17) the converse proved to be the case. As the instability increases, the amount of salt increases.

The higher winds consist of those over 15 kts. All data points are included with the sole exception of those taken on May 11, 1965 when stable conditions were recorded along with high winds and high local production. This was one of the very few instances on the whole cruise where stable conditions were noted. For the wind speeds of 7 to 15 knots no correlations with instability were found and the data tended to be quite scattered. For the light winds, those of less than 7 knots, all data points without exception are included.





During the higher winds fairly intense local production is taking place and the dependency of sea-salt mass on the instability is rather straight forward. As the instability increases, the probability that parcels of air originating immediately over the sea surface will reach the 10 m collecting level within a fixed time interval also increases. Since these parcels will contain some of those larger particles whose size generally relegates them to the air immediately over the sea surface due to high sedimentation rates, there will be a significant increase in mass per cubic meter of air at the collecting level. In this case physical intuition is closely matched by the data. As the instability increases the larger particles are on the average lifted up higher against the settling force of gravity. Even those particles with high fall velocities were able to reach the 10 m level where observations were taken. To avoid confusion, it should be mentioned that the increases noted in sea-salt content due to increased instability were in addition to those related to increasing wind speed. For example, if two samples were taken at 20 kts on different occasions, the higher value would be found on the occasion with the greater instability.

The light wind case (Fig. 16) is at first somewhat less obvious, although it too is best explained by instability and consequent mixing. It is well documented that over the continents the heating of the surface and the resulting instability creates a nearly uniform distribution of sea-salt nuclei with height, at least in the lower several kilometers. (Junge, 1958). This is in sharp contrast to the maritime situation where the distribution generally falls off quite rapidly with increasing altitude, although certain secondary maxima may occur (Woodcock, 1953; Lodge, 1955). Particularly with some of the larger values of $T_W - T_A$ it is to be expected that mixing will occur from the sea surface up to an altitude of several thousand feet (Houghton, 1968).

With no local production to complicate matters, what occurs is a simple redistribution of the particles and hence the mass. Instead of having greater concentrations right over the sea surface (Woodcock and Gifford, 1949) which fall off with height, a more nearly uniform vertical distribution results from the presence of the relatively deep mixed layer. This in turn will cause the values near the sea surface to be lower as a consequence of the approximate conservation of mass in a particular column of air. This result was observed and the correlation between low values of salt and greater instability was quite strong. Instability is therefore another significant factor which must be considered in the overall salt distribution in addition to production, sedimentation, wash-out and advection.

RESULTS AND DISCUSSION

c. Fallout Rate and the Particle Lifetimes

On March 10 and 11, 1965, (Fig. 5) there were typical north-east monsoon winds of approximately 15-17 knots causing local production of sea-salt nuclei. Quite suddenly, around 2400 h. on March 11, the wind dropped to about 6 knots. Between that time and March 15, the winds were very light, reaching 9 or 10 kts for several hours only and often being under 4 knots. During this period the wind turned from northeast to north and remained from that direction. Thus there was essentially no production after 2400 h. March 11 and the direction and speed of the wind were such that no continental influence would be felt due to advection. Even though the ship travelled several hundred miles, it is not inconceivable that the same conditions prevailed over a fairly broad area. Keeping the limitations of the argument in mind, it is possible to obtain at least a rough idea of the fallout rate. Figure 18 shows that after approximately 2 1/2 days the salt content has fallen to one-third its value before production ceased. This means that roughly 60-70% of the mass fell out during the interim. Referring to Woodcock's (1953) distribution curves, we see that for a wind between force 4 and 5 Beaufort (about 16 kts), approximately two-thirds of the mass is found in particles larger than 10^{-10} gm. mass. It can be concluded as a limited approximation that those particles larger than 10^{-10} gm. all had lifetimes of 2 1/2 days or less. Considering the somewhat qualitative arguments involved, this finding is remarkably consistent with the estimates of 2.6 and 2.9 days given for particles of 10^{-10} gm. and larger by Eriksson (1959) and Toba (1965a) respectively.

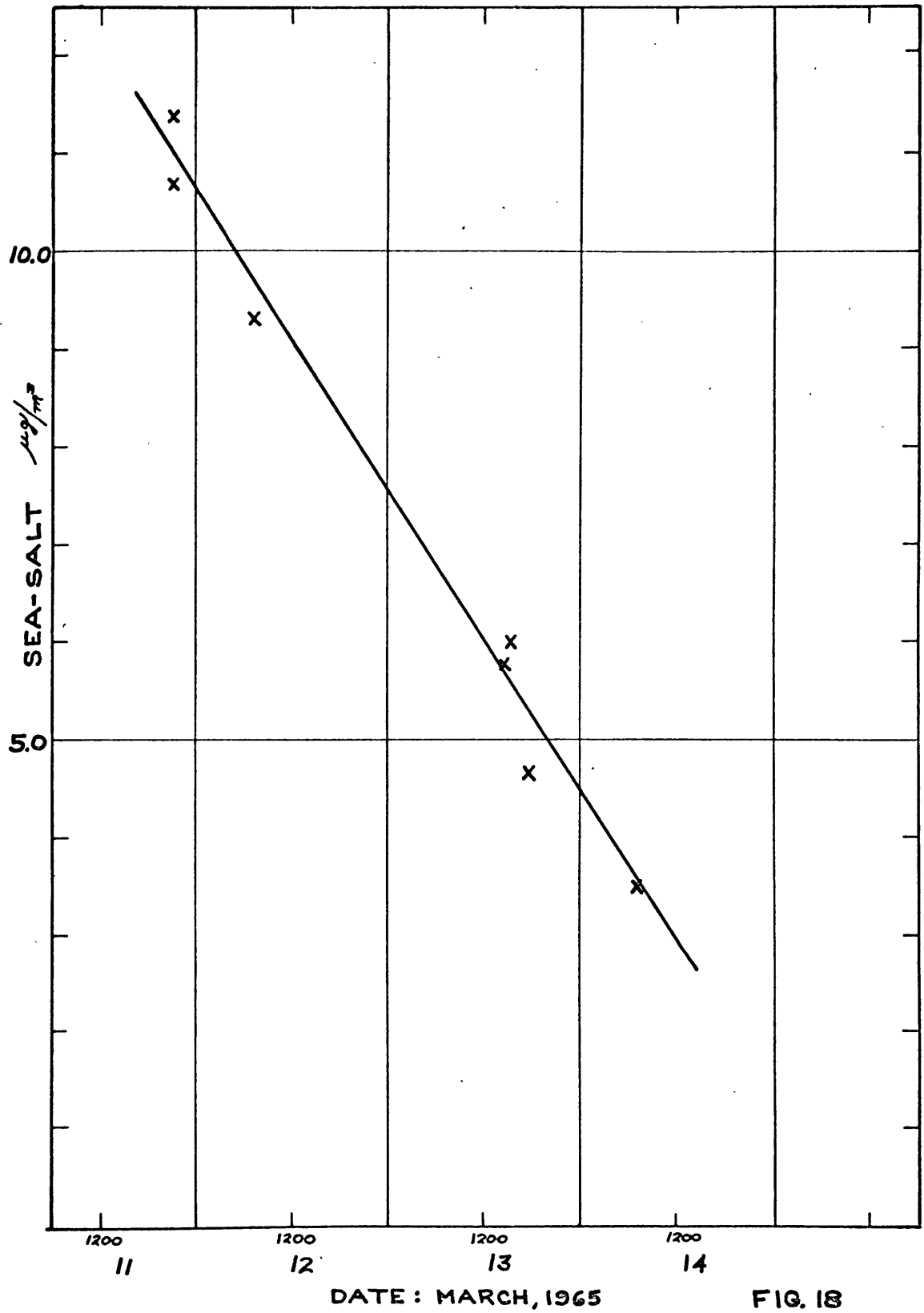


FIG. 18

VII. SUMMARY

- (1) Sea-salt nuclei may be collected on small diameter wires at efficiencies of approximately 90% for those particles whose radius is greater than one micron. Almost all of the total mass of sea-salt in the marine atmosphere is to be found in these so-called giant particles.
- (2) At wind speeds above 8 to 10 kts there is local production of sea-salt nuclei and the total sea-salt content is a logarithmic function of the wind speed.
- (3) The production rate appears unrelated to geographical position so long as the situation is truly maritime.
- (4) At wind speeds below 8 to 10 kts the logarithmic relationship breaks down due to the highly variable effects of advection, sedimentation, instability, and washout. The last two factors qualitatively appear to be the most important at the sampling height of 10 meters.
- (5) Even with fairly uniform conditions as during the dry monsoons, the salt content varies from one large parcel of air to the next at the same wind speed. Values may differ by factors of two to three in the same day, all other conditions seeming equal.
- (6) The effect of convection as measured by the degree of low-level instability is found to be quite significant. Vertical mixing decreases the salt content at the 10 meter level for light winds, those less than 7 knots and it increases the salt content for higher winds, those greater than 15 knots.

- (7) Based on a somewhat qualitative argument it appears that the larger particles (mass $\gt 10^{-10}$ gram) settle out quite rapidly when production ceases. A reasonable estimate of their lifetime is one to three days.

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