TEKNISK NOTAT NR 7/76

AIRBORNE SEA SALT MEASUREMENTS
A LITERATURE SURVEY

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ABSTRACT

The work of this survey was directed toward a thorough evaluation of the current state-of-art of airborne sea salt measurements. Literature references surveyed were mainly those available through Norwegian library services. Aspects of airborne sea salt generation, distribution, transport, and removal processes were also examined, as well as the special sea salt-related problems encountered in the use of gas turbines in marine environments.

A variety of sea salt measurement methods is available to suit different objectives of measurement programs. Chemical analysis techniques for sea salt sample evaluation are more advanced than techniques for obtaining representative samples of airborne sea salt. The main factor determining sea salt particle production and their airborne concentrations and size distributions is wind speed, but local influences in coastal zones can have substantial impact.
From the information available on sea salt, only range estimates of airborne sea salt concentrations for a given locations can be made.

1 INTRODUCTION

The enigmatic role of sea salt, in that it can be both benefical and detrimental, has been recognized for some time. Beck (1829) more than 150 years ago described the destructive effects of airborne sea salt on coastal vegetation. Aitken (1881) was the first to point out that hygroscopic sea salt particles are essential in the atmospheric processes of condensation and precipitation.

The oceans are the primary sources of airborne sea salt. When transported by winds over the land, the salt particles are subsequently removed from the air by fallout and precipitation, and carried back to the oceans by rivers. This cyclic salt phenomenon, which appears to be one of the numerous processes on which the present status of the biosphere depends, was first recognized by Conway (1942/43).

Thus, airborne sea salt is virtually ubiquitous and exerts its influence in a variety of ways, ranging from therapeutic effects on the human respiratory tract (Schmidt, 1972), to the generation of atmospheric electricity (Blanchard, 1963), to serving as carrier of organic matter (Neuman et al. 1959, Barger and Garrett 1970) and viable organism (Aubert and Aubert, 1973), to determining the physical and chemical characteristics of coastal soils and plants (El Swaify et al., 1968), to the deterioration and corrosion of materials, structures and machinery, and to missile failures (Brierly, 1965). Accordingly, airborne sea salt has received increasing attention of many diverse scientific disciplines, as its role in natural processes and environmental problems has become apparent. Although unevenly distributed among the various disciplines, a substantial amount of scientific literature exists today on the different aspects of airborne sea salt. Oceanographers, geochemists, and cloud physicists, for example, have a considerable interest in the processes of sea salt generation, transport, chemistry, residence time in the air and removal mechanisms, and have provided by far the largest and continuing source of scientific information on the subject.

Of more recent origin are the engineering applications-oriented investigations, which have focussed on the damaging aspects of airborne sea salt in the use of machinery in marine environments, and cooling towers for electric power generating plants. Gas turbines, used for marine propulsion, or for shore- and over-the-ocean installations, suffer fouling and corrosion damage from sea salt ingestion (Kaufman, 1969). Seawater cooling towers disseminate salt particles in the surroundings (Rofman, 1973), with potentially detrimental impact on the environment.

2 BACKGROUND FOR THE SURVEY

2.1 Purpose of the survey

To understand the phenomenon of airborne sea salt and to assess or estimate the effects it may produce, quantitative data, derived from field and laboratory measurements, are essential. Because the occurence and quantities of sea salt are highly variable in space and time, available data may not be adequate or of sufficient resolution to make such an assessment for given geographical areas.

The impetus for the survey was supplied by an inquiry from the Kongsberg Våpenfabrikk A/S, Gas Turbine and Power System Division*, requesting NILU's assistance in the procurement of airborne sea salt measurement data relevant to the North Sea oil platform environment. The parameters of interest were:

a) sea salt particle size
b) distribution (wet and dry)
c) concentration
d) temperature spreads
e) relative humidity
f) wind velocity

An earlier NILU investigation by Larssen (1974), consisting of onboard ship measurements of sea salt during a cruise on the Oslo fjord and a limited literature search, did not provide adequate answers to the above. It was then decided that NILU should conduct a thorough literature survey to gather information as complete as possible on the over-the-ocean sea salt particle concentrations and size distributions, and to assess the current state-of-art of measurement techniques and instrumentation.

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* Letter to NILU from F. Erikson, A/S Kongsberg Våpenfabrikk, Gass Turbin division, 1.Dec.1975.

2.2 Scope of the survey

It was thought reasonable not to restrict the scope of the survey to the conditions encountered only on oil platforms, because in the future requirements might arise for broader-based information. Furthermore, there was also an early impression of a complete lack of information on the off-shore platform environment, and data even from onboard ships measurements appeared to be sparse.

Thus, for the sake of completeness, available literature on all types of airborne sea salt measurements was examined, including studies on the spatial variations of sea salt above the ocean and in transport over land.
In the majority of studies of geochemical and cloud physics significance, sea salt sampling has been done in such a way as to exclude the contributions of direct spray droplets. Generally then these measurements will not reflect adequately the more severe conditions near the ocean surface. It is also common, that he results are given in terms of particle number concentrations and number distributions, instead of the corresponding particle mass parameters, of primary interest to engineering designers. Although, when using certain assumptions conversion is possible, it was not attempted here.

In the survey, literature available principally from Norwegian library sources was examined, unless relevant materials could also be obtained from outside Norway within the time period available.

2.3 Method of the survey

The literature searches for the survey were conducted through NILU's Bibliotek. An APTIC (United States) search was initiated by supplying a list of key words. The libraries of Det Norske Veritas, Norsk Skipsforskning Institutt, NTH (Trondheim), and FFI (Kjeller) were contacted and literature listings requested, but only NTH had relevant materials.

Bibliographies of certain publications proved valuable as starting points for expanded searches, among which were Junge (1963), Junge (1972), Brierly (1970), and Larssen (1974). Once the literature listings from the various sources had been received, relevant books, reports, and journal articles were selected for detailed examination. For example, out of 176 citations selected by APTIC, 85 were deemed relevant to the survey. If the materials were unavailable at NILU's Bibliotek, they were ordered from other sources, if available on relatively short notice.
Several personal request for clarifying details were sent to authors, some of which resulted in additional up-to-date information.

During the course of the survey, 23 books and reports, as well as some 160 journal articles were selected and scanned for information. Relevant information on sea salt was distributed among the various scientific periodicals, examined in this survey, as shown in Table 1. The remaining articles were distributed among various other journals. Additionally, numerous articles, cited in the Reference section, deal with the over-all methods and problems of suspended particulate matter sampling, and, although relevant, are not specific to airborne sea salt measurements. These were not listed in Table 1. The most important single source of information on gas turbine-related studies was the papers (9) presented at various gas turbine conferences, and published by the Americal Society of Mechanical Engineers (New York).

Table 1. Distribution of information on airborne sea salt among scientific journals, selected for the survey.

Name of journal	Number of articles	
	selected for review	cited in References
Journal of Geophysical Research	28	18
Tellus	23	19
Journal de Recherches Atmospheriques	18	12
Journal of Applied Meteorology	12	6
Journal of Meteorology	12	12
Quarterly Journal of the Royal Meteorological Society	10	4
Journal of Aerosol Science	9	6
Nature (London)	8	2
Atmospheric Environment	7	4
Environmental Science and Technology	6	4
Geofisica Pura e Applicata	4	4
Special Contributions of Kyoto University	4	4
Bulletin of American Meteorological Society	4	4
Journal of the Oceanographical Society of Japan	3	3
Journal of the Meteorological Society of Japan	3	3
Journal of Atmospheric Sciences	3	3
Analytical Chemistry	2	2
Journal of Colloid and Interface Science	2	2
Science	2	2
Journal of Engineering for Power; Transactions of the American Society of Mechanical Engineers	2	2
Total	162	115

3 THE NATURE OF AIRBORNE SEA SALT

3.1 Composition af sea water

Sea water, the source of airborne sea salt, has a salt content, i.e., salinity in the open ocean (away from inshore influences) from about 34 $^{o}/oo$ to 38 $^{o}/oo$* (Neumann and Pierson, 1966). This means that the "average" sea water has about 35 grams of various salts dissolved in 1 kilogram of water. Land-locked waters, influenced by melting ice and fresh-water river run-off from land, may have much lower salinities. Examples are the inner parts of the Baltic Sea and some fjords, where the salinity may be as small as 0.5 $^{o}/oo$ to 1 $^{o}/oo$. The revers is true in land-locked arid regions where evaporation exceeds precipitation.
It has been found that regardless of the absolute concentrations of salt, the relative proportions of the different major constituents in sea water remain remarkably constant. Table 2 shows representative average values.

Table 2: Average ionic composition of sea water for total salinity of 35 $^{o}/oo$ (after Neumann and Pierson, 1966)

Cations	g/kg	Anions	g/kg
Sodium	10.752	Chlorine	19.345
Potassium	0.39	Bromine	0.066
Magnesium	1.295	Fluorine	0.0013
Calcium	0.416	Sulphate	2.701
Strontium	0.013	Bicarbonate	0.145
		Boric acid	0.027
Total	12.866	Total	22.2853

---

*The symbol $^{o}/oo$ is read "per mille" or "parts per thousand" (by weight).

About 80% (by weight) of the dissolved salts in sea water is NaCl, followed by $MgCl_2$ and $MgSO_4$ in abundance. As a consequence of the slight excess of cations (2.38 meq/kg), sea water is weakly alkaline (pH from 8.1 to 8.2).

3.2 Properties of sea water particles

A droplet of sea water introduced into the air will undergo evaporation, because of the low concentration of salts in sea water. The resulting sea salt particle will then assume a size at which the concentration of the salt solution is in vapour pressure equilibrium with the surrounding air (Koehler, 1936). Thus, a droplet may shrink by a factor of 2 or 3, depending on the relative humidity (R.H.) of the air before reaching equilibrium and consist of a concentrated salt solution, or is a dry crystal. Correspondingly, its density can change from near 1 g/cm^3 to about 2.2 g/cm^3 for a dry sea salt particle.

The various dissolved salts in sea water are hygroscopic and deliquescent. While pure NaCl crystals deliquesce (i.e. form droplets) at a little below 75% R.H., phase transition of the mixed-composition sea salt appears to occur at somewhat lower relative humidities (Winkler and Junge, 1972). However, it is generally necessary for the relative humidity to decrease considerable below these R.H.'s to ca. 40%, before solid particles separate again. This supersaturation hysteresis phenomenon has been experimentally varified (Orr *et al.*, 1958; Junge, 1963). At the relative humidities normally encountered over ocean surfaces, sea salt particles will almost always be in the liquid droplet state, with their size being a function of the salt content (i.e., the size of the original sea water droplet) and the ambient R.H.

Table 3 shows sea salt particle sizes at equilibrium conditions for selected salt content and relative humidities.

Table 3: Computed sizes of sea salt particles as a function of salt content and relative humidity (after Woodcock, 1950a).

Weight of total sea salt in particle,* grams	size of particle		
	radius of crystalline sphere, μm	radius of liquid sphere at 80% R.H., μm	radius of liquid sphere at 95% R.H., μm
1.5×10^{-12}	0.55	1.1	1.7
1.2×10^{-11}	1.1	2.0	3.2
1.5×10^{-10}	2.6	4.7	7.5
1.2×10^{-9}	5.2	9.4	15.1
1.2×10^{-8}	11.0	20.1	32.2
1.2×10^{-7}	24	44	70
5.0×10^{-7}	38	70	112

In our constantly changing atmosphere, however, equilibrium conditions are probably seldom actually reached. Furthermore, sea salt phase transition and droplet growth have been found to be affected by the presence of organic substances (of natural or pollution origin) dissolved or dispersed in sea water (Pueschel and Van Valing, 1974), which exhibit geographical variations.

*Also called the "nuclear mass".

3.3 Production processes of airborne sea salt

Aitken (1881) was the first to suggest that airborne particles might be produced by the ocean, stating that "In all probability the spray from the oceans, after it is dried and nothing but a fine salt-dust left, is perhaps one of the most important sources of cloud forming dust". Following this idea, it was thought for some time, that sea spray produced in foam of breaking waves was the main source of airborne sea salt particles. However, apart from those droplets that originate at the coastline in the surf, such particles are not very numerous and are generally too large to remain airborne for very long.

Melander (1897) had proposed, that salt particles may escape from warm salt solutions in the course of the evaporation of the solutions. This hypothesis remained unchallenged for over half a century, until Lodge et al. (1954) rejected it both on theoretical and experimental grounds.

Although the formation of jets from collapsing cavities at the surface of water had been studied and photographed earlier (Worthington and Cole, 1897), no other production mechanisms were advanced until Jacobs (1937) recognized the possible significance of the upward-moving jet, associated with the breaking of an air bubble at the surface of water. The subsequent theorotial and experimental research efforts devoted to the investigation of this process have been reviewed (up to about 1962) by Blanchard (1963) and (up to about 1970) by Mason (1971). Bubble bursting over the vast expanses of the open oceans is now accepted as the most important process in the production of sea salt particles. Various other mechanisms, such as direct shearing of droplets from the wave crests, and the aerodynamic suction of droplets from the crests of capillary waves, undoubtedly contribute a share of the total production (Lai and Shemdin, 1974).

Figure 1 shows diagrammatically the processes involved in the bubble bursting mechanism.
Bubble bursting ejects two types of sea water droplets from the water surface into the air; small droplets, representing the remains of the actual film of the bubble exposed to the atmosphere, and larger jet drops, which rise rapidly from the bottom of the collapsing bubble.

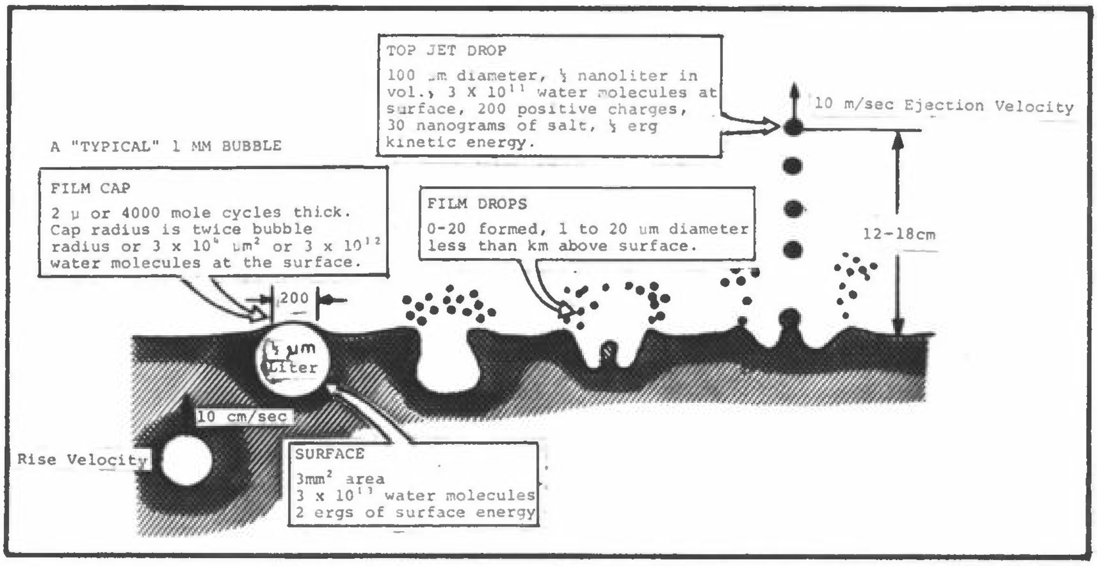

Fig. 1. Diagrammatic representation of bubble bursting and film and jet particle production. (after Horne, 1969).

It can be expected then, that the resulting particles will also have two distinct, if somewhat overlapping, size ranges.

The bubbles themselves have been observed to be generated primarily at wave crests from air entrapment (whitecaps), but falling rain and snowflakes, impacting on water surface, are also producing air bubbles (Blanchard and Woodcock, 1957). In the absence of sufficiently strong winds, the majority of bubbles might result from the growth of the so-called "microbubbles" of gas, already existing in the water column (Horne, 1969). Sofar no direct proof of the existence of such microbubbles is available, but speculation seems to centre on their formation by gas sorption on the surface on small particles suspended in sea water, or by biological processes, e.g., photosynthesis and respiration of marine organisms.

Several secondary production processes, that is the production of a number of particles from a single sea salt particle after it has become airborne, have also been suggested. Aitken (1911) observed large numbers of condensation nuclei at the inter-tidal zone along a beach when the sun was shining on it. The literature shows no evidence, however, that this lead has been followed up (Blanchard, 1963), Dessens (1946), and later Twomey and McMaster (1955) reported production of secondary particles when sea salt droplets were allowed to suddenly crystallize. In follow-up experiments, Junge (1958), and Blanchard and Spencer (1964) found no evidence of this, and concluded that phase changes associated with sea salt droplets in the atmosphere were an unlikely source of secondary particles.

Numerous difficulties hamper making reliable estimates of airborne sea salt production and distributions on a global basis. Published data on the film particle production are scarse (Tomaides and Whitby, 1976), but it has been established that the generation of film particles is critically dependent on such factors as the presence of organic surface films and the clustering of bubbles, and is very sensitive to the sixe of the bursting bubbles (Blanchard, 1961; Day, 1964). Considering all these uncertainties, reliable estimates of global production rates of film particles are virtually impossible to make. Estimating the production rates of the larger jet particles is also a complex task, because here it is the <u>net</u> production (that is, that proportion of the total production which remains airborne for sufficiently long periods to participate in the various atmospheric processes) which is of importance. This net production is a function of many factors, e.g., wind speed, the size spectrum of the air bubbles, the ejection heights and size of the jet droplets, the relative humidity of the ambient air, and eddy mixing of air above the water surface.

Eriksson (1959), using whitecap bubble size distribution spectra of Blanchard and Woodcock (1957) and jet droplet ejection heights, obtained a sea salt particle flux spectrum, expected to approximate the net production per unit area of whitcaps. In the steady state, this input, or production rate, must equal the output, or fallout rate. The product of the production rate and an estimated total area of whitecap coverage of the world oceans (about 0.3-3% of the total surface area of the oceans) would not, however, give a reliable global net production rate. The reason for this is, that the jet particle size distributions tend toward larger particles with increasing wind speeds, which would not be accounted for when simply a given spectrum of particles, weighted only with the percentage of whitecap area, is used.
Toba (1965 a), from measurement data of Woodcock (1953, 1957), Lodge (1955), Hayami and Toba (1958), and Durbin and White (1961), extrapolated average vertical sea salt profiles down to 10 m level above sea surface, and then calculated the average concentration of sea salt at the water surface (Toba, 1965 b). From this, he calculated the net production as a function of particle mass (in the range $10^{-11.5}$ to 10^{-8} grams) and wind speed (force 1-12 on the Beaufort scale). Equilibrium was assumed to exist between the sedimentation loss of particles from the air and the introduction of new ones at the sea surface.

These finding, together with marine climatological data on wind velocities, enabled Toba (1966 a) to present estimates of the average number concentrations of airborne sea salt (at the sea surface and at 1 km height) for the world's oceans in the form of isopleth maps for various seasonal periods of the year.

From all measurement data available up to about 1970, Mason (1971) estimated an average sea salt particle production rate of 100 $cm^{-2}sec^{-1}$, from all the production processes and for the oceans as a whole.
There is clear evidence, that measurements of sea salt production rates are subject to considerable local influences in coastal areas. For example, Blanchard (1969) found production rates as high as 4000 $cm^{-2}sec^{-1}$ at a shoreline site with heavy surf and breaking waves. In terms of mass concentration (e.g., $\mu g/m^3$ of air), particles produced under such conditions far surpass (by a factor of 50 or more, cf. Fig. 4) those particles which are already present in the air, i.e., blowing over the surf zone from the open sea (Duce and Woodcock, 1971).

3.4 The size of airborne sea salt particles

Experimental measurements have indicated that, in general, the atmospheric aerosol is highly variable from place to place, depending on local sources and the extent of "aging" of particles in the air. However, a pattern of regularity, at least over a portion of the aerosol spectrum, has emerged regardless of the location of the measurements. This feature was first noted by Junge (1963) from plots of "log radius-number" distributions. This distribution is defined by:

$$n(r) = \frac{dN}{d(\log r)} \qquad (1)$$

where N is the total concentration (number per cm^3) of particles of radius smaller than r.

Figure 2 shows such a composite plot for airborne particles in the undisturbed marine environment. The number of particles, ΔN, between the limits of the interval $\Delta(\log r)$ can be obtained from the plots by $\Delta N = n(r)\Delta(\log r)$.
It is seen, that in the undisturbed marine environment (i.e., remote from direct shore and continental influences) particulate matter consists of essentially two components:

the sea salt component (lines 'a' and 'b'), and the superimposed tropospheric background aerosol (line 'c').

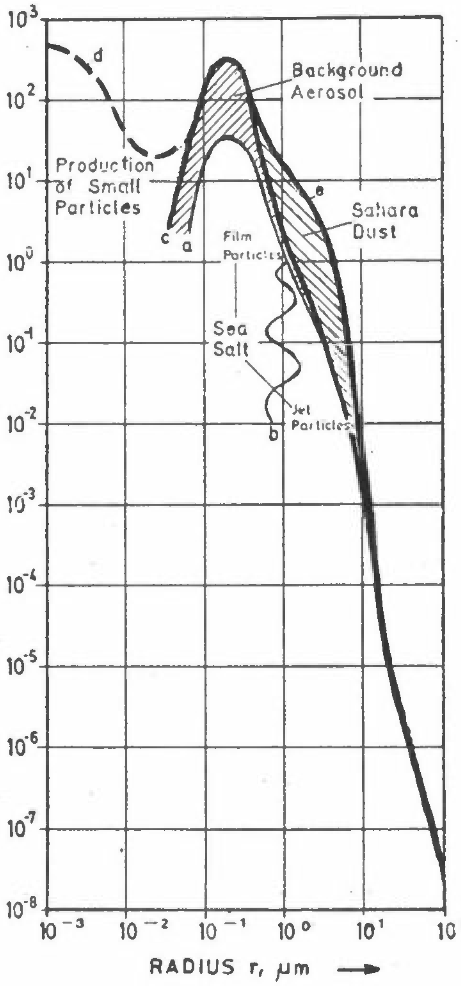

Fig. 2.

Idealized representation of the various components of the undisturbed marine aerosol, according to Junge (1972).

The sea salt component is below the line 'a' and separated by line 'b', which demonstrates the uncertain information on the size and overlap of film and jet particles.

The background aerosol particles (between lines 'c' and 'a', and less than 1.0 μm in radius) consist most likely of sulphates, e.g., $(NH_4)_2SO_4$.

Both components, however, are probably modified by antropogenic pollutants, particularly over the North Atlantic (Junge, 1972). Oceanic areas, influenced by air masses originating in the Sahara desert region, may experience an added contribution, as indicated by line 'c' in Fig. 2. Line 'd' represents a still tentative component of very small particles (Junge and Jaenicke, 1971), for which no definite source has been yet confirmed.

Junge (1963) proposed that a portion of the distribution could be approximated by the power law relationship:

$$\frac{dN}{d(\log r)} = cr^{-\beta} \qquad (2)$$

where c is a constant and β is about 3. This has become known as the "Junge subrange" of the aerosol spectrum.

The sea salt component, in the distribution shown in Fig. 2, is contained mainly in the "large" and "giant" particle range. In the nomenclature system suggested by Junge (1963), the "large" particles have radii between 0.1 and 1 μm, and the "giant" particles have radii greater than 1 μm. Although for individual observations the exponent β in Eqn. (2) may deviate considerable from 3 (Metnieks, 1958; Junge, 1963), for the average sea salt particle size distribution, shown in Fig. 2, it appears to be reasonably close to that value in the giant particle range. Similar agreement for sea salt particles in the lowest atmospheric layer above the sea surface was found by Chaen (1973), from the results of extensive observations on various cruises aboard oceanographic vessels.

In view of the hygroscopic nature of sea salt particles, size distributions are frequently represented in terms of the so-called "nuclear mass" or "weight of sea salt in particle" (in grams), instead of radius or diameter (cf. Table 3). This size of particles, however, is comparable to others only when related to the same relative humidity or air, or when representing the size of spherical particles of equivalent dry mass. In Junge's subrange, the replacement of r in Eqn. (2) by the nuclear mass corresponds to $\beta=1$.

The size distinction between the bubble film (large) and the jet (giant) particles, as well as the lower and upper size limits of these particles are still uncertain at this time (Junge, 1972). Recent laboratory studies (Tomaides and Whitby, 1976) showed that the size of the bursting bubbles determines the relative proportions of film and jet particles produced.

Within a certain range of bubble sizes, the number distribution was clearly bimodal, that is, both film and jet particles were produced, although the jet particles showed much better monodispersity. The size of the bubble also determined the

relative numbers of film and jet particles generated. The bursting of larger bubbles outside this range resulted in the production of film particles only, the monodispersity of which was improved with larger bubble sizes.
Woodcock's (1972) observations of the smaller sea salt particles in the air (over Hawaii and the Alaskan sea) suggest, that particles of a mass of 10^{-14} to 10^{-13} gram represent the transition from bubble film to jet particles. This is in the size range of 0.1 to 1 µm in radius depending on the relative humidity.

Most accounts in the literature, however, seem to indicate that it is only the size range between 1 and 20 µm in radius, that is sufficiently well known from field studies (Junge, 1972). Most of the size distribution measurements have been done in the giant particle range, but data by Metnieks (1958) and extrapolation of Woodcock's (1953) results place the lower size limit of airborne sea salt particles at about 0.1 µm radius. The numbers' maximum in the distribution (cf. Fig. 2) is in the vicinity of about 0.3 µm radius. Meszaros and Vissy (1974) concluded from the results of their measurements in South Atlantic and Indian oceans, that sea salt particle number concentrations were an order of magnitude smaller in these water, than those observed in the North Atlantic (Junge and Jaenicke, 1971). The southern hemisphere measurements also place the transition between film and jet particles at 0.2 to 0.4 µm radius size, with the number conccentration maximum at approximately 0.1 µm radius.

From his evaluation of giant sea salt particle distributions, Toba (1965a) concluded that, although the production of particles of radii 20 µm was indicated, turbulent eddy diffusion above the sea surface was insufficient to transport the particles upward against gravity beyond this size.

Monahan (1968) observed sea salt droplets of 75 μm radius and larger at a height of 1.2 m above sea surface, but also reasoned that atmospheric turbulence would not carry these particles to a sufficient height for them to remain airborne for long enough period to evaporate significantly. And yet, more recent measurements with improved methods for the detection of the more massive sea salt particles over the open ocean (Junge and Jaenecke, 1971) revealed, that (at least at the 10 m level above sea surface) sea salt particles with radii 100 μm were present. Ship-produced spray particles were reportedly excluded from these measurements. It is not clear at this time, whether these larger particles are due to droplet coalescence at cloud level, followed by evaporation and sedimentation, or to higher than previously estimated production rate at the sea surface.

While the film particles are the numerically more abundant group, the jet particles account for most of the mass in airborne sea salt particle distributions. The log radius-number distribution (Eqn. 1) may be converted to a log radius-volume distribution, v(r), by:

$$v(r) = \frac{dV}{d(\log r)} = \frac{(4/3)\pi r^3 dN}{d(\log r)} \qquad (3)$$

Furthermore, if an average density, D, can be assumed for the particle population, a log radius-mass distribution, m(r), results:

$$m(r) = Dv(r) \qquad (4)$$

Where Junge's subrange relationship holds, the mass concentration distribution should then be constant over the range of the radii.

In the distribution shown in Fig. 2, sea salt particles larger than 1 μm in radius carry about 90% or more of the total sea salt mass. Fig. 3 shows examples of two typical

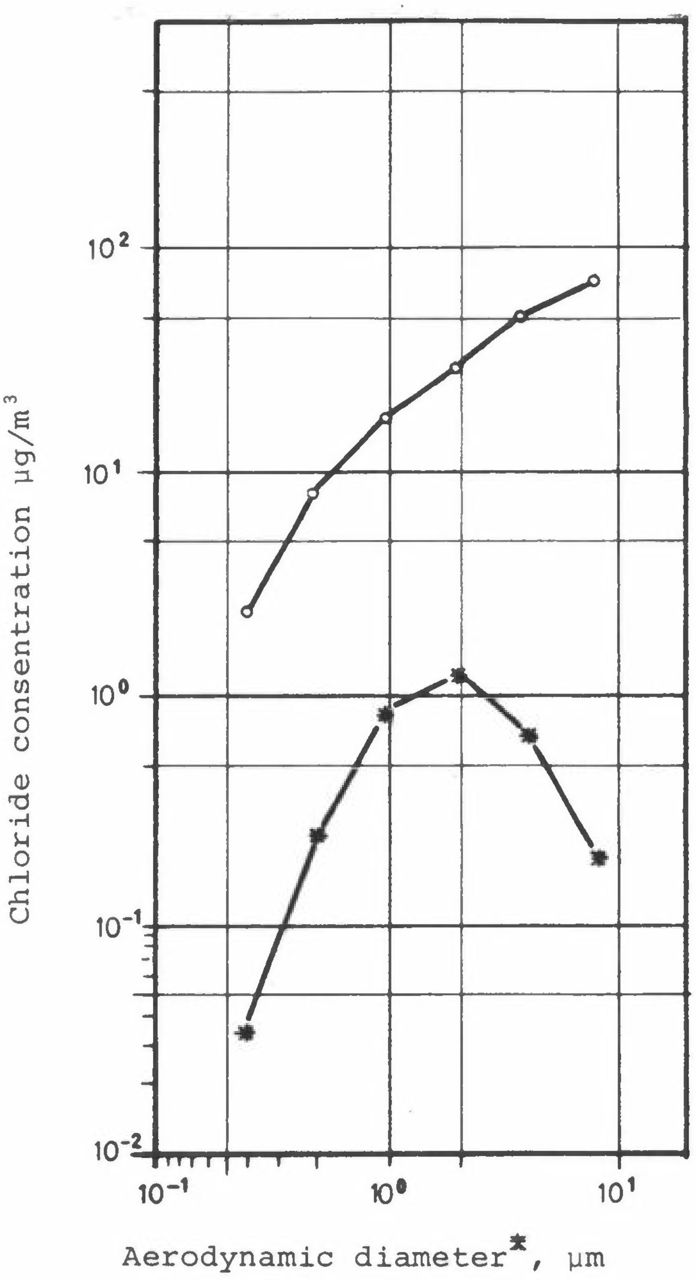

Fig. 3. Typical size distributions for mid-ocean and surf zone airborne chloride in sea salt:

Crosses - mid-ocean (Duce <u>et al</u>., 1967)

Circles - surf zone (Duce and Woodcock, 1971)

*cf. p. 50 for definition.

measured mass concentration distributions. The maximum in mass distribution of mid-ocean chloride in salt particles is in the vicinity of 1 to 3 μm radius (Duce et al., 1967). For mixed surf zone and open ocean particles, the maximum shifts toward considerably larger sizes, depending on the local conditions (Duce and Woodcock, 1971).

3.5 Chemical composition of airborne sea salt

At first glance the chemical nature of particles of sea water origin would appear straight-forward, due to the constancy of chemical components in sea water. Thus, if the sea surface is the source of all the chemical elements and ions in airborne sea salt by a process of sea water dispersion, e.g., bubble bursting, it could be expected that all elements and ions would also have the same size distribution as, say, sodium or chloride. The fact is, however, that the chemistry of sea-air interface is probably the most poorly understood of the three boundaries separating the sea-air-land system (Duce et al., 1972). Discussions and controversies surrounding the chemical composition of airborne sea salt have gone on for more than 30 years (MacIntyre, 1970; Chesselet et al., 1972a; Junge, 1972). The ratios of the various chemical elements and ions can, however, differ considerably from those found in sea water, and yet the particles can have their origin in the sea. Particularly the bubble film particles, which come from the thin surface layer of sea water, could well be different from that of the bulk sea water.
The reasons postulated for the experimentally found "non sea water ratios" have included:

ionic fractionation during particle formation;
modification of ionic composition due to organic surface films;
modification of particles in the atmosphere subsequent to their formation;

non-marine origin;
experimental errors;
sample contamination due to the very low amounts of material involved.

One of the major controversies has revolved around the "enrichment"* of the cations Mg, Ca, K, and Sr relative to Na, for which preferential fractionation seems to exist for the smaller size particles (Komabayasi, 1964). Recent experimention (Hoffman _et al_., 1974) with careful separation of continental dust from the collected samples, appears to indicate that there is little major cation fractionation in airborne sea salt, relative to sea water, at least in the morthern hemisphere oceans. Particulate matter of mineral origin from arid areas can penetrate unexpectedly far over the oceans and have a considerable influence on the overall composition of marine particles.

The halides, Br and I, also show size-dependent enrichment. Both Br and I fractionations again appear to be associated with the smaller particles (i.e. of bubble film origin), which have longer residence times in the atmosphere, than the giant particles containing primarily chloride. The particularly high iodine content in the smaller particles is thought to be most likely due to absorption of gaseous iodine (Junge, 1972).

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* Enrichment E is defined as:

$$E = \left[(X/Na)_{atm}/(X/Na)_{sea\ water}\right] - 1$$

where $(X/Na)_{atm}$ is the weight ratio of any element X to sodium in an atmospheric sample (particulates, rain, etc) and $(X/Na)_{sea\ water}$ is the same elemental weight ratio in sea water (Duce _et al_., 1972).

Chloride loss has been detected from both the large and giant sea salt particles (Junge, 1956; Robbins et al., 1959; Martens et al., 1973). The possibility that chlorine gas is liberated from the airborne particles by reaction with ozone in the air has been rejected, because the reaction would be too slow to be of any importance in atmospheric processes. Oxidation of SO_2, dissolved in sea salt droplets, followed by liberation of HCl, or the reaction of HNO_3 vapour with solid or liquid particles of NaCl have been postulated (Martens et al., 1973) as probable causes. Thus, marine aerosols appear to act both as a source of gaseous Cl and Br, and as a sink for iodine.

Although most airborne sea salt measurements show, that the Cl/Na ratio for the giant sea salt particles is close (within the limits of measurement accuracy) to that of bulk sea water (Junge, 1972), anomalously high Cl/Na ratios have also been observed (Hidy et al., 1974). These are believed to be linked with chemical reactions transforming gaseous chlorine compounds into condensed particulate material.

Considerable variations in the relative composition exist between sea salt particles produced over the open ocean and those in coastal surf zones (Duce and Woodcock, 1971). Several studies have revealed, that a substantial proportion of particles above ocean surfaces (particularly those less than 1 μm in radius) differ in composition from sea water in other respects (Dinger et al., 1970; Twomey, 1971). Ocean surfaces are known to be covered with a thin layer of organic matter, some of which is likely to become admixed to the sea salt particles during their formation (Pueschel and Van Valin, 1974).

In those parts of oceanic areas, even in remote regions, where minute traces of SO_2 and NO_2 are present in the air, airborne particles can be expected to be enriched in sulphate and nitrate. Excess sulphate, primarily as $(NH_4)_2SO_4$, is now a wide-spread phenomenon in both marine particulate matter and precipitation (Junge, 1972). Lodge *et al.*, (1960) have reported the surprising result, that at a ship-based ocean weather station in the Pacific (approx. midway between San Francisco and Honolulu) sulphate concentrations were generally higher than those of chloride.

It appears then, that, in the absence of measurements for a given geographical marine area, it might be unwise to assume the composition of airborne marine particulate matter to be the same as that of bulk sea water.

4 THE BEHAVIOUR OF AIRBORNE SEA SALT

4.1 The influence of wind speed

It can be reasoned that, because airborne sea salt particles are produced at the sea surface, there should be a close correlation between whitecap coverage and sea salt particle concentrations over the ocean surface. This should be so, regardless of what the exact production mechanism(s) involved may be, since the droplets are produced directly or indirectly by the disruption of the sea surface.

The studies of Munk (1974), Mandelbaum (1956), and Woodcock *et al.*, (1953) on whitecap formation and surface coverage indicated that the onset of whitecaps occurs after a "threshold wind speed" (ca. 7-8 m/sec) has been reached.

Toba (1972; 1973) in his extensive investigations on wind waves, observed that the breaking of wind waves began at a wind force of about 3 on the Beaufort scale (ca. 5 m/sec). He concluded that the breaking of wind waves is related to the process of momentum transfer from wind to wind waves, and to the turbulence in the sea water. Toba proposed, that the overall degree of wave breaking was best correlated with the dimensionless parameter $u_* L/\nu$, where u_* is the friction velocity of air, L the significant wave length, and ν the kinematic viscosity of air.

In some of the earliest investigations of airborne sea salt concentrations (e.g., those cited by Neumann, 1949), insufficient number of measurements and/or lack of simultaneous wind data did not permit to establish the existence of sea salt concentration-wind speed relationships. Among the early researchers, Jacobs (1937) appears to have been the first to attempt a correlation of airborne sea salt mass concentrations* (measured as chloride at a shoreline location in southern California) and wind speed. A definite relationship was found, but he cautioned that influence could have obscured the extent of any overall variations.

The first extensive measurements to explore variations in sea salt concentrations with altitude above sea level and with speed were made by Woodcock (1953) in the trade-wind region of Hawaii. Woodcock found a consistent pattern, in both subcloud and cloud layers of air, of a general increase in sea salt concentrations, as the wind speed increased, and provided a graph, showing "sea salt mass concentration _vs._ wind speed". Junge (1963) supplemented Woodcock's data points with sea salt concentration values from other studies (usually made at sea level), which also fitted into the same concentration range. Chesselet _et al._, (1972 a) presented in a similar graph results from Cl and Na measurements in North Atlantic and the Mediteranian Sea, which again covered essentially the same range. Considering the differences in measurement techniques and the variety of conditions and geographical area where the observations were made, the agreement is quite good.

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* Several "conventions" are found in the literature for describing airborne sea salt mass concentrations:

(a) sea salt (presumably all the soluble salts in the airborne particles;
(b) NaCl, about 20% less than sea salt;
(c) Cl, and (d) Na.

The analytical method used for sample evaluation frequently determines which convention is most convenient. Sea salt concentrations can be approximated (bearing in mind the uncertainties of ionic fractionation) by multiplying the values of (c) or (d) by 1.82 or 3.25, respectively.

There is little doubt that, in general, both mass and number concentrations of the giant sea salt particles are highly correlated with wind speed over oceanic areas. Mass concentrations reflect overwhelmingly the contribution of the giant particles, but measurements by Meszaros and Vissy (1974) have shown that the linear correlation coefficient of even the number concentration of giant particles with wind speed was 0.70, although only 0.56 for the large (bubble film) particles.

Fig. 4 shows the Woodcock-Junge-Chesselet _et al._, (W-J-C) range, as well as some other results, which fall considerably outside it. These exceptions are usually those associated with measurements in or near the surf zone (Jacobs, 1937; Holzworth, 1959; Whelan, 1975), very close to the surface of the sea (Monahan, 1968), or aboard ships in heavy seas (Tsunogai _et al._, 1972). In the latter instance, inadvertent inclusion of sea water drops from direct spray could be suspected. As the range implies, considerable scatter in the individual measurements always exists. For some of the more extensive measurements, best-fit correlation equations have been proposed. The regression equations, summarizing the results of Woodcock (1953), Tsunogai _et al._, (1972), and Lovett[*] are given below, and are also shown graphically in Fig. 4.

Woodcock:	$\log S = 0.07U+0.41$	(5)
Tsunogai _et al._:	$\log S = 0.27U-0.48$	(6)
Lovett:	$\log S = 0.07U+0.63$	(7)

where: S = sea salt mass concentration, μg/m^3
U = wind speed, m/sec

[*] Unpublished data, provided by Kaufman, NAVSEC, Philadelphia, Pa.; personal communication, May, 1976. Lt. Cdr. R.F. Lovett's sea salt measurement results from three weather stations in the North Atlantic (west of the British Isles and south of Iceland) have been graphically summarized by Shaw and Jefferis (1976).

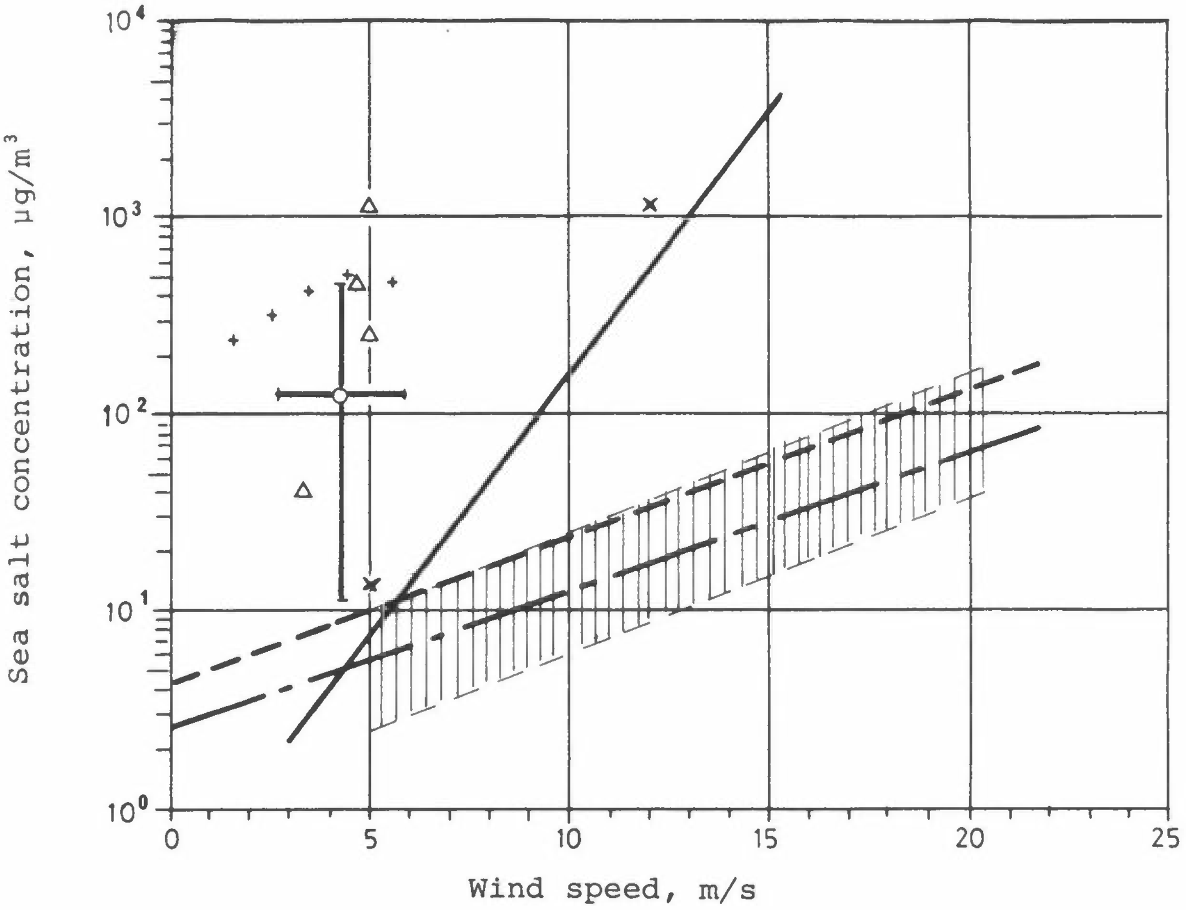

Fig. 4. Dependence of airborne sea salt mass concentrations over-the-ocean and in the surf zone on wind speed.

▥ : "Woodcock-Junge-Chesselet et al., range"
— - — : Woodcock (1953), Eqn. (5)
——— : Tsunogai et at., (1972), Eqn. (6)
— — — — : Lovett*, Eqn. (7)

+ : Jacobs (1937)
× : Riedel (1938)
△ : Whelan (1975)
○ : Holzworth (1959) - average of Farallon Island data; the vertical and horizontal bars indicate the range of concentrations and wind speeds encountered, respectively.

* cf. footnote, p. 25.

As can be seen from Fig. 4, the most dramatic rise in airborne sea salt mass concentrations with wind speed is predicted by Tsunogai et al., (1972), while Lovett's values fall within the W-J-C range.

It is also of interest to note, that Equations (5), (6), and (7) imply a"background" concentration under calm conditions (zero wind speed) of approximately 2.6, 0.33, and 4.3 µg/m³, respectively. Chesselet et al., (1972 b) measured airborne sea salt in western Mediteranean Sea from a floating buoy laboratory during low wind speed (0 to 2 knots) conditions, at 0.5 and 8 m above the sea surface. The sea salt concentrations ranged from about 0.6 to 17 µg/m³*, depending on the wind direction. Although wind speed is the primary factor affecting airborne sea salt generation, several additional factors have also important consequences. These include other meteorological, as well as geographical and oceanographical influences (Roll, 1965). The state of the sea surface is determined, in addition to wind speed, by the fetch and duration of the wind. It is likely, that there is a lag time between the build-up and decay of wind speed and airborne sea salt concentrations. Relatively low sea salt concentrations can be found with high winds, if the wind has just begun, and relatively high concentrations persist with low wind, if the wind has just died down (Rossknecht et al., 1973). Furthermore, increases in oceanic production of airborne sea salt with increasing wind speed might be hidden by increased vertical mixing, which accompanies strong winds (Blanchar, 1963). Chaen (1973) observed, that erratic and extreme values in sea salt concentrations are likely to be measured during periods when there is no wind, or only a week breeze, because of the presence of airborne sea salt particles, produced before the onset of the calm. He reasoned, that it was more proper to consider the relationship between the number concentration of sea salt particles and the overall strengh of the turbulence of the wave field. The latter is related to both wind speed and wind waves, and to air entrainment in wind

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* estimated from µg Na/m³.

waves and the bursting of bubbles. Chaen (1973) proposed a series of empirical equations for estimating airborne sea salt number concentrations, as functions of the dimensionless parameter u_*L/ν[+], in the particle size range from 10^{-12} to 10^{-7} grams nuclear mass of sea salt. The values of the parameter can be estimated from observed wind speed at a reference height, the wave period, and air temperature.

In surf zones, the subaqueous bathymetry controls the breaking wave conditions and influences the production of airborne sea salt. The extent to which sea salt concentrations over water near coasts are affected by the admixture of the surf zone component is thus a function of wind trajectories. Over land, the generally complex topographical features of coastal areas may completely mask any systematic relationship between sea salt concentrations and wind speed.

[+] cf. p. 23 for definition of symbols.

4.2 Vertical distribution over the sea

Individual measurements of vertical profiles of airborne sea salt distributions have shown considerable scatter, but a general decrease in concentration with height (Junge, 1963). Measurements from aircraft over the oceans in subtropical and higher latitudes have encountered very little sea salt at altitudes of about 2 km, and there appears to be a complete lack of sea salt particles above the 3 km level (Junge, 1972).

Toba (1965 a) synthesized observational data of Woodcock (1953, 1957), Lodge (1955), and Durbin and White (1961) and concluded that the vertical distribution of number concentrations of giant sea salt particles is approximately characterized by an exponential decrease (i.e., the sea salt is removed from the air in proportion to its concentration in the atmosphere). He proposed a model for simulating vertical profiles of sea salt over the sea, and Fig. 5 shows an example of Tobas's model distributions compared to those observed by Lodge (1955). The theoretical profile can be seen to deviate from the purely exponential distribution, and approximates a state where the particle number concentration is almost constant at the higher levels. The model calculation requires information on the production rate of sea salt particles at the water surface, the ratio of the areas of upward and downward moving air currents, the velocities of the air in these regions, and the sedimentation velocities and eddy diffusivities of the sea salt particles.

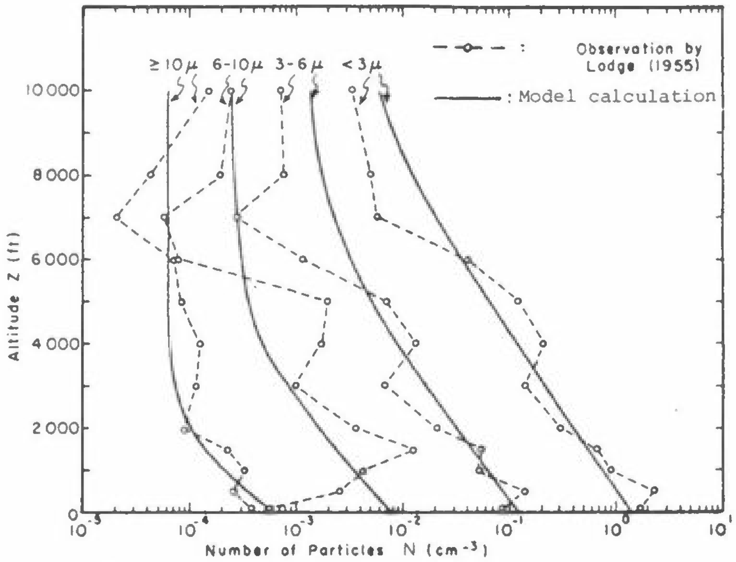

Fig. 5: An example of vertical distribution of giant sea salt particles over the sea calculated Toba's model (Toba, 1965a) and observed by Lodge (1955) (after Toba, 1965a).

Fig. 5 also shows apparent concentration maxima at varying heights above the sea surface, found by Lodge (1955), especially with lower wind speeds. If true (Lodge was unable to verify their existence by repeat measurements), such a elevated maximum would imply a net sink for sea salt at the sea surface. Lodge (1955), and Eriksson (1959) have attributed this anomaly to variations in wind speed and turbulence from time to time and from place to place above the sea, and the associated transport of sea salt from areas at at distance with higher sea salt production.

Sea salt measurements in the sea-air transitional layer, i.e., the lowest 10 og 15 metres above the sea surface, have for the most part been made at only one height, although the more recent investigations, however, by Chesselet et al. (1972 b), and those summarized by Chaen (1973) provide data on sea salt concentrations at several levels above the water surface. The only theoretical model by Toba (1965 b) is based on the assumption of equilibrium distribution of sea salt in the transitional layer, that is, a balance between the particles falling due to gravitational force, and upward eddy diffusion. In the model, the vertical distribution of wind speed, vapour pressure, and temperature are obeying a logarithmic law. Steady state holds with no horizontal gradient, but the fall velocity of sea salt particles, as well as the eddy diffusivity change with height above sea surface. Although fall velocity varies in a cmoplicated manner from droplet to droplet, for simplicity, it is assumed that each droplet of sea water attains its equilibrium salinity for the vapour pressure and temperature of the surrounding air, which are given as functions of height. Consequently, the terminal fall velocity of the droplets is a function of height only, if the mass of sea salt in the droplet is given. Toba then solved the equation of diffusion of the number concentration of sea salt particles for the boundary conditions: $N=N_o$ at $z=0$, and $N=0$ at $z=\infty$, where N is the number concentration of sea salt particles, and z the height above sea surface.

The solution has the form:

$$\log\ (N/N_o) = -M^{2/3}\ U_{10}^{-1}\ X(R.H._{10}, \gamma_{10}, z) \qquad (8)$$

In Eqn. (8), the logarithm of the ratio N to N_o is proportional to the two-thirds power of sea salt mass, M, contained in the particle, and inversely proportional to the wind speed at the 10 m level, U_{10}, with a factor, X, which is a function of the relative humidity at the

10 m level, $R.H._{10}$; the friction coefficient, γ_{10}; and the height, z. Because the value of the friction coefficient does not vary greatly with wind speed, the value of X can be regarded as a function of R.H. only, and the value of N/N_o as a function of M, U, and R.H.

The vertical distribution of the number concentration for each class interval of salt mass plots as a straight line on a log-log graph, according to Eqn. (8). According to this theory, the higher the relative humidity and the lower the wind speed, the more particle concentration decreases with height. Conversely, the lower the relative humidity and the stronger the wind speed, the more uniform is the particle number concentration in the 10 m layer above the sea surface.

Subsequent vertical distribution measurement of sea salt by Toba and his associates, to verify the proposed model, have been analyzed by Chaen (1973). These observations were performed aboard two oceanographic research vessels on various cruises in the East China Sea and the Indian Ocean, and at an oceanographic research tower station in Tanabe Bay. The measurements were carried out at 3 or 4 levels above the sea surface aboard the ships, when sailing, drifting, or at anchor. Wind speeds encountered during the observations ranged from calm to about 17 m/sec. Chaen found, that the mean vertical distribution of measured number concentrations of sea salt particles in the lowest atmospheric layer above the sea surface was very close to a straight line on a log-log plot of N vs. z, as predicted by Toba's model, although individual distributions were variable. The ovserved vertical gradients of the distribution of salt particles, on the average, were also close to the gradient calculated from Toba!s theory for the condition of $R.H._{10}=95\%$, even though the actual relative humidity of the surrounding air at the time of observations was lower than 95%. Chaen suggested that the reason for this was that the concentration of the sea salt solution in each of the droplets was still low and not in equilibrium with the relative humidity surrounding the droplet, as assumed by Toba's model. Many of the sea salt particles, after being produced at the sea surface, just had not been long enough in the air to attain vapour pressure equilibrium. Accordingly, he proposed a modification in the equilibrium theory, by introducing an "effective relative humidity" of 95% at the 10 m level.

Whelan (1975) compiled a vertical sea salt mass concentration profile, shown in Fig. 6, for the first kilometre above surf zones from his own surf zone measurements, and from data of Woodcock (1953), Duce and Woodcock (1971), Chesselet et al. (1972 b), and Hoffman and Duce (1972). An exponential decrease is apparent up to a height of about 5 m, but there are only small changes in sea salt particle mass concentrations from about 20 m to 1 km. The high airborne sea salt concentrations in the lower layer above the surf zone reflect the admixture of freshly generated and relatively massive spray particles from the surf zone to the more "aged" ones transported from the open sea.

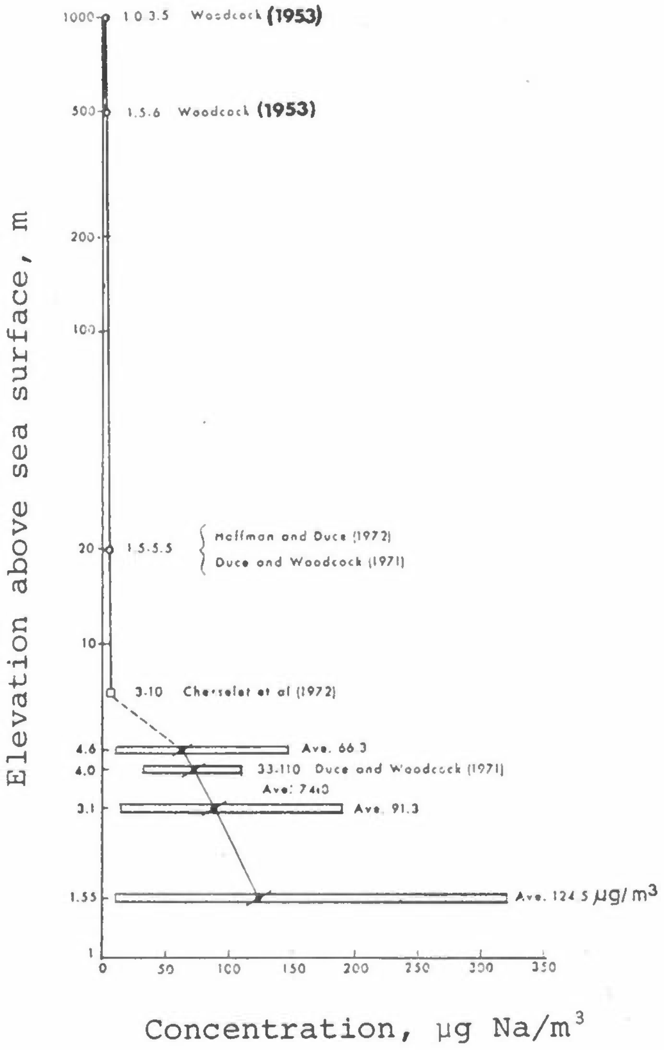

Fig. 6. Vertical sea salt particle sodium (Na) concentration distribution above sea level from 1.55 m to 1000 m. (after Whelan, 1975).

4.3 Inland penetration of airborne sea salt

Observations of sea salt concentrations and the effects they can produce have shown, that airborne sea salt can penetrate far into the interiors of continents. As the oceanic aerosol is transported inland from the coast, the approximately esponential distribution reaching the coast changes into a more complex feature with distance inland. The coastline represents a transition from a source region at sea to a sink region over the land. It also constitutes a discontinuity in terms of surface roughness, heat and moisture content of the air, and changes in wind profiles, convection and turbulence.

Byers et al. (1957), on flights over mid-western U.S.A. (a considerable distance from the nearest salt water body), discovered a striking characteristic of the vertical distribution of giant sea salt particles inland. The particle concentration had a maximum at some elevation above ground (between a few hundred metres to two kilometres) and decreased sharply toward the ground. The measurements showed ratios of maximum to ground level concentrations ranging from 10 to 100. Sedimentation measurements by Toba and Tanaka (1965) had suggested, that the apparent ground sink of giant sea salt particles is not only sedimentation, but that impaction on trees, and other vegetation and obstacles on the ground may constitute a much more important removal mechanism. Subsequently Toba (1965 a), and Toba and Tanaka (1968) developed a theoretical model for the vertical distribution of giant particles inland, taking the above features into consideration. Figure 7 shows a comparison of the observations of Byers et al. (1957) and Toba's (1965 a) model predictions. The model has been presented in exact solution form by Tanaka (1966), and predicts a concentration ratio(maximum to ground level) of about 40, which is in good agreement with the averages from observations of Reitan and Braham (1954), Byers et al. (1957), Podzimek and Cernoch (1961), and Meszaros (1964). Tanaka's model predicts an increase in number concentration of the particles with increasing height, with the level of maximum concentration rising, as the air moves inland. To apply the model, information on particle sedimentation velocities and eddy diffusivities, average wind speed (normal to the shoreline), and an estimated particle impaction efficiency parameter are required. The latter factor cannot be determined for any particular impaction surface, such as a forest or a building, but its average value can be estimated from sea salt concentration observations (Semonin, 1972).

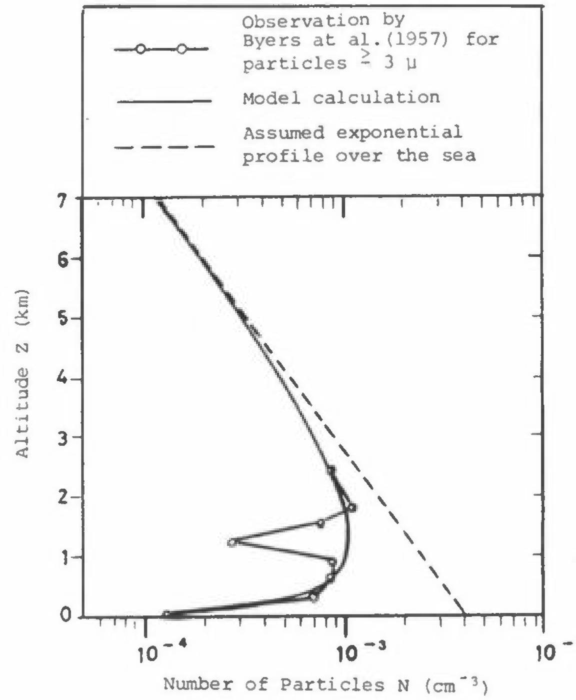

Fig. 7: An example of vertical distribution of giant sea salt particles far inland calculated by Toba's model (Toba, 1965 a) and observed by Byers *et al.* (1957). (after Toba, 1965 a).

Recent aircraft measurements of vertical sea salt profiles by Blifford and Gillette (1972), and King and Maher (1976) have shown generally good agreement with the model predictions.

Hsu and Whelan (1976) developed a model for the vertical distribution of sea salt mass for coastal transition zones, which has shown good agreement with concentrations measured at various heights in the surf zone by Duce and Woodcock (1971), and Hoffman and Duce (1972). The model takes into consideration the influence of irregular terrain features, such as a nearshore cliff or sand dunes, which can act as a source or a sink of salt particles, depending upon the aerodynamic characteristics of wind over the area.

Although measurements of sea salt concentrations at varying distances inland from the shore can be readily made at ground level, the deduction of systematic relationships from such measurements is complicated by the effects of changing topography of the ground, and variations in vegetation cover and air humidity conditions. Kline (1972) studied the trends in sea salt particle depletion with distance inland by comparing his measurements on the island of Hawaii with those of Lodge (1955) from Puerto Rico. He found, that although the concentration values were different, the patterns of change at both locations were compatible, and amounted to about 3 orders of magnitude concentration decrease in 100 km.
The models of Tanaka (1966), and Toba and Tanaka (1968) can also be used for the ground level case, provided reliable impaction-sedimentation ratio estimates for the particles are available. Semonin (1972), and Rossknecht _et al._ (1973) found, that their measurement data could be fitted with these models, but that the fit, was better for the larger particles. Rossknecht _et al._ (1973) also noted, that the models were sensitive to relative humidity (particularly for humidites above about 85%) because of the growth of sea salt particles with increasing relative humidity. They found a sharp decrease in the mass of the giant sea salt particles in the first 4 km inland from the shore, which levelled out farther inland and decrease more gradually. Typically, the total mass at 4 km inland was 15% of that at the coast, while at 30 km the concentration had diminished to about 8%. Semonin's (1972) measurement data indicated a decrease to about 1% of the near the coast in a distance of 68 km; however, the difference in elevation for the two points was considerable.

4.4 Residence time of sea salt over the oceans

Residence time of airborne substances is important for determining budgets of their various components, for estimating removal rates to sinks, and for determining possible transport distances.

The risidence time of substances in a reservoir, such as the atmosphere, is related to the total amount of the substance in the reservoir, the rate of its production of release, and the rate of its removal of decomposition. The residence time then can be taken as the ratio of the total amount to the production rate, or to the removal rate. In order to maintain balance between the production of sea salt at the sea surface and the concentrations of airborne sea salt which exist at any given place and time, there must be significant removal or scavenging processes for the particles. Because airborne sea salt in largely in the giant particle range, sedimentation, rain-out, and wash-out can be regarded as the important removal processes. As previously noted, however, there is a scaricity of reliable data on sea salt production rates. There are similar uncertainties about the relative removal efficiencies and magnitudes of the wet and dry scavenging mechanisms. Consequently, the result of any estimate of the residence time of airborne sea salt will depend on the assumption adopted for its determination.

Table 4 gives a summary of estimates by various authors of mean residence time of airborne sea salt over the oceans.

Table 4: Residence time of sea salt particles over the oceans (after Junge, 1972)

	Residence time, days			
	$M=10^{-12}g$	$M=10^{-11}g$	$M=10^{-10}g$	$M=10^{-9}g$
Particles at R.H. = 80% (Toba, 1965a)	86	17	2.9	0.32
Particles at R.H. = 91.4% (Toba, 1965a)	59	11	2.1	0.23
Estimated from sedimentation (Eriksson, 1957)	82	16	2.6	0.4
Estimated from production (Eriksson, 1957)	3.5	1.0	0.6	0.5
Considering vertical gradients (Junge, 1972)	1.9	1.6	1.0	0.26

M = mass of salt particles, in grams.

Although he considered sedimentation to become important only for particles larger than about $10^{-9}g$, Toba's (1965 a) estimates were nevertheless based on sedimentation rates for all particle sizes. Eriksson (1957) had followed a similar procedure, but regarded residence time of more than 20 days unrealistic for sea salt particles, and made alternative estimates based on particle production. Junge's (1972) estimates took into consideration the vertical gradients of airborne sea salt in the air, but the resulting residence times were considered by him too low. He suggested an average residence time of about 3 days (except for particles larger than $10^{-9}g$). In this estimate, rain-out and wash-out were taken as the controlling, but equally efficient factors in particle removal. Sedimentation was considered unimportant for particles smaller than 5-10 μm radius.

5 AIRBORNE SEA SALT MEASUREMENTS

5.1 General

By the year 1976, the amount of scientific literature on the various aspects of airborne sea salt has grown to such proportions, that no attempt at a complete accounting will be made here. Rather it is intended that the selected references will reflect the historical development and advances, as well as the current state-of-the-art in airborne sea salt measurement techiques and instrumentation. Furthermore, it is hoped that the cited works, in addition to their own merit, will serve as representative examples of a particular aspect of the subject, with the understanding that these are only some of many others.

5.2 Historical

Accounts of the earliest efforts in airborne sea salt measurements are sketchy, particularly in regard to detailed descriptions of sampling techniques and supportive meteorological data. Before World War II, most of the activity in sea salt investigations appeared to have centred in continental Europe.

Neumann (1940), in reporting on his own North Sea measurement results, summarized the earliest studies on airborne sea salt by Duphil, Gautier, and Knoch (cited by Haeberlin, 1929). Their results are shown in Table 5 (as given by Rau, 1956a), which also includes the data of Riedel (1938), and Neumann (1940). The value of 15000 µg $NaCl/m^3$ found by Duphil is very high, and although the wind speed on the occasion is not known, it must represent rather special local conditions at the measurement site. The mass concentration values of Gautier, Riedel, and Neuman are all in the same range and agree with those of Duce and Woodcock (1971), and Whelan (1975), as being representative of airborne sea salt mass concentrations in surf zones. The results of Riedel also demonstrate the wind

Table 5: Summary of early airborne sea salt measurements in Europe (after Rau, 1956 a).

Author	Place of Observations	Method of measurement	Particle number concentration, N/cm^3	Mean radius of particles µm	NaCl concentration µg/m^3 (wind speed, m/s)
Duphil[+]	West coast of France; 20 m from shore	++	++	++	15000
Gautier[+]	Lighthouse in the Atlantic off the French coast	++	++	++	220
Knoch[+]	Zoppot bathing beach on the Baltic Sea	++	++	++	15
Riedel (1938)	Norderney Island, in the North Sea off the German coast; ca 50 m from shore	Konimeter	++	++	140 (at 5 m/s) 1120 (at 12 m/s)
Neumann (1940)	Hörnum on the Island of Sylt, of the western coast of Denmark; at various distances from shore	Konimeter	8.2 at 2 m from water	1.5	275
			4.0 at 60 m from and 10 m above water	1.3	75
			2.15 at 1800 m from and 25 m above water	1.1	30
			0.42 at 9000 m from the water approx. at sea level behind sand dunes	1	4.4

[+] cited by Neumann (1940)

[++] not given

speed effect, while those of Neuman clearly show the decay in sea salt particle concentration with increasing distance from the shoreline.

In the post-Word War II period, investigations on airborne sea salt have expanded and accelerated, in keeping with the generally favourable "climate" for research activities in continental Europe, Great Britain, Japan, and the U.S.A. Paricularly in the U.S.A., such studies were stimulated by the readily available financial support. Further incentives were supplied by the pioneering work of Woodcock and his associates (Woodcock, 1950 a,b,c, 1952, 1953, 1957; Woodcock and Mordy, 1955; Woodcock and Wyman, 1947; Woddcock and Gifford, 1949; Woodcock *et al.*, 1953), and by "working visits" of such eminent atmospheric chemists, as C.E. Junge (Junge, 1954, 1957 a,b; Junge and Gustafson, 1956; Junge and Werby,1958).

The development of sophisticated instrumental analysis techniques in recent years has undoubtedly been the greatest advance in measurement technology, and has made rapid and convenient multi-component analyses possible. Their very low detection limits have permitted flexibility in sampler use and reduced sampling time requirements, thus facilitating greater time resolution in measurements. Among these techniques, atomic absorption (AA) spectroscopy (for Na, K, Ca, Mg); neutron activation (NA) analysis (for Cl, Na, K, I, Br); and proton-induced x-ray emission (PIXE) analysis (for Cl,Ca,K,S,Br,Pb) have been most useful in airborne sea salt investigations. These methods have been of particular importance in the assessment of the existence and mechanisms of ionic fractionation.

Because of the great spatial and temporal variability of sea salt concentrations, it would serve little purpose to detail in this survey all of the virtually hundreds of individual investigations.
It can be said, however, that at the present time investigations of airborne sea salt (and various other components of the

marine aerosol) have spanned all the major bodies of water of the world. The greater accumulation of information, however, has been for the North and South Atlantic, the Caribbean area, and parts of North and South Pacific, particularly in the Hawaii and Japan-Australia areas. The graphical summary in Fig. 4 may be regarded as representing the overall status of our current knowledge on airborne sea salt concentrations in the atmosphere.

5.3 Airborne sea salt and the gas turbine engine

Designers of airborne sea salt separation devices for the protection of gas turbines in marine environments are interested in the mass concentration and size distribution of sea salt particles reaching the turbine inlets under most weather conditions, conceivably encountered in their operation. The objectives of measurement programs to provide such information, as well as the results, could differ from those of geochemical or oceanographic studies, and a brief review of this specialized area of airborne sea salt measurement may be in order.
The various applications and the problems experienced with the use of gas turbine in the marine atmosphere have been reported by Brockett et al. (1967), Bowers (1967), Woodward (1968), Kaufman (1969), Hart and Cutler (1973), and Shaw and Jefferis (1976), among others.

In some of the earliest (in the late 1940's) marine vessel applications, the gas turbine engines used were those designed for aircraft propulsion. Very rapid drop in performance in relatively short time was experienced, which was traced to the deposition of ingested sea salt in the first stages of the compressor of the turbine. The deposits distorted the compressor rotor and stator blade profiles and reduced its compression efficiency. This problem, however, has been virtually eliminated by the introduction of several turns in the air intake ducts and a plenum chamber, as well as routine washing or cleaning of the compressor blades (Kaufman, 1969). On the other hand, the so-called hot-zone corrosion or "suphidation", which affects fuel nozzles, combustors, and the turbine first stage nozzles and blades in the high-temperature section of the engines, has been an ongoing problem (Dalton, 1965; Rogus and Weinert, 1972; Hart and Cutler, 1973; Mitchell and Kievits, 1974; Labadie and Boutzale, 1976). Hot-zone corrosion is an accelerated, often catastrophic surface attack of superalloy gas turbine components in a temperature range of about 760 to 980°C. This mechanism of corrosion has many complexities, and has been studied for over 20 years without complete agreement as to its causes among the experts. Some investigatiors belive, that NaCl (which is in the vapour state at the above temperatures) plays a dominant role in hot-zone corrosion, whereas others postulate that condensed alkaline salts, notably Na_2SO_4, are the most important factor. In either case, hot-zone corrosion is the result of a breakdown in the normally protective oxide coating, and the penetration of sulphur and oxygen into the base metal. The concentration of contaminants required to produce hot-zone corrosion is as small, as a few parts per billion (by weight), and the attack rate normally increases with metal surface temperature. The major source of NaCl and Na_2SO_4 is sea salt, which is ingested into the gas turbine with the inlet air, or fuel contaminants. It was once thought, that Na_2SO_4 was formed in the hot section from Na and SO_2 (or SO_3) during fuel combustion. This theory, however, is currently losing favour, in view of the insufficient high temperature residence time for the reaction to take place (Labadie and Boutzale, 1976).

5.4 Gas turbine-related airborne sea salt measurements

Modification of fuels and the introduction of more resistant alloys and coatings for turbine hot-zone components have provided a partial solution to hot-zone corrosion problems. A concurrent reduction of ingested sea salt, however, is also considered essential (Rogus and Weinert, 1972), which requires an assessment of airborne sea salt concentrations and size distributions in gas turbine inlet air. Towards this end, the U.S.A. Navy and the British Royal Navy have conducted separate, as well as collaborative investigations aboard ships (Kaufman and Pollini, 1967; Morgan _et al_., 1970; Shaw and Jefferis, 1976).

Fig. 8 summarizes the results to date from the various measurement programs by the British NMW-NGTE* and the U.S.A. NAVSEC** (Rogus and Weinert, 1972; Shaw and Jefferies, 1976).

It has been conventional to present gas turbine-related sea salt measurement results in terms of concentrations i ppm[+] (by weight) and wind speed in knots.
Because the size-mass concentration distributions of sea salt particles is an important parameter in airborne particle separation equipment design, all the measurements also have included particle size determination, in addition to total mass concentration of airborne sea salt.

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[*] Naval Marine Wing, National Gas Turbine Establisment.

[**] Naval Ship Engineering Center.

[+] ppm is a weight ratio and means: parts of "sea salt" per million parts or air.
The British NMW-NGTE uses ppm of NaCl, while the U.S.A. NAVSEC prefers ppm of sea salt (cf. footnote, p. 24).

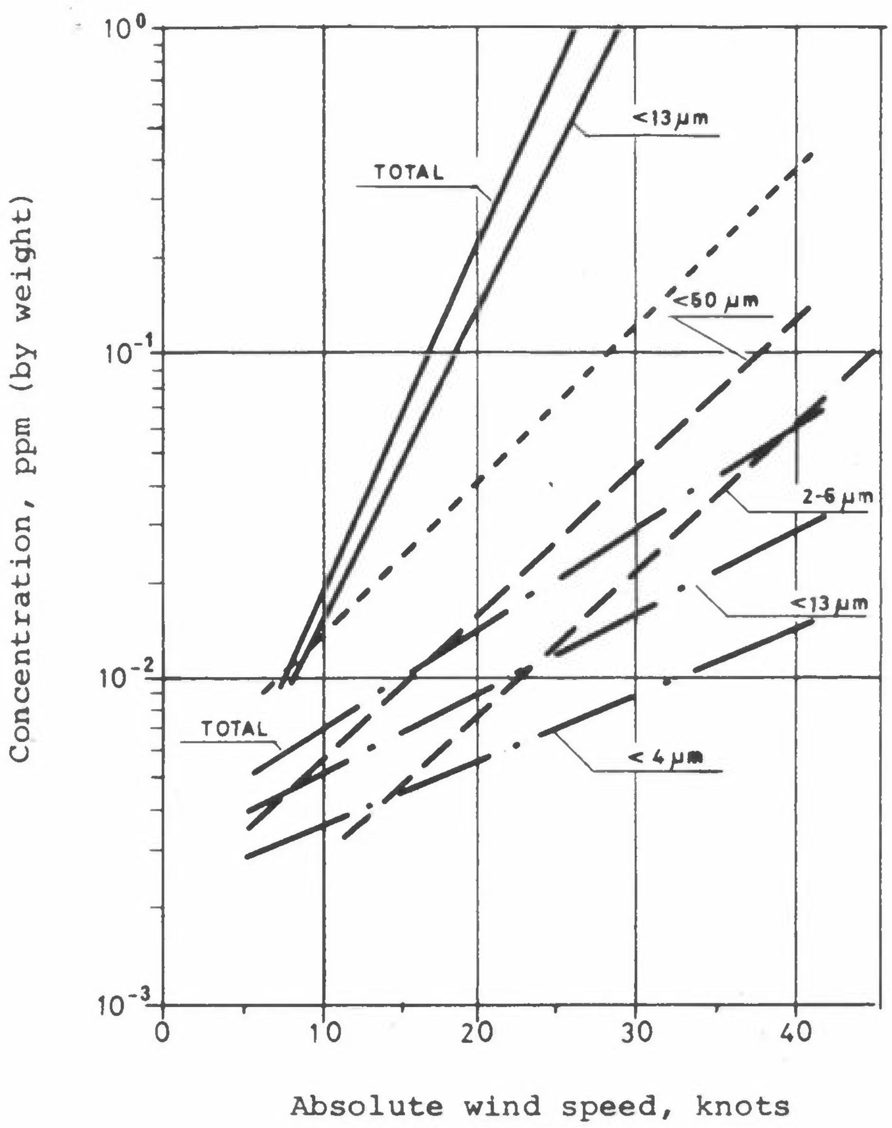

Fig. 8: Dependence of airborne sea salt concentrations, measured aboard ships, on particle size and wind speed.

———— : British NMW-NGTE "standard" data (ppm NaCl)

— — — : Lovett's recommended data (ppm NaCl) (cited by Shaw and Jefferis, 1976).

——·—— : U.S. NAVSEC data (ppm sea salt) (Rogus and Weinert, 1972)

- - - - - - : Average value data from "Asiafreighter", March 1974*

* NAVSEC Report No. 678 - data supplied by Mr.E.R. Kaufman, personal correspondence, March 1976.

Fig. 8 demonstrates the dependence of sea salt concentrations on wind speed for selected particle size ranges (in terms of equivalent aerodynamic diameters*). It has been customary in this regard to refer to the particle fraction equal or less than 4 μm in diameter as "aerosol" to distinguis it from the more massive particles. The aerosol particle fractions of all data sets fall within the W-J-C range, as does Lovett's concentrations af all particles smaller than 50 μm in diameter. The similar slopes of Lovett's curves for both the aerosol fraction and "total" sea salt suggest, that the rates of production for both the aerosols and the more massive particles are the same, and that at high wind speeds the contribution of the smaller particles might be greater than indicated in previous measurements (Kaufman and Pollini 1967). It is apparent, that the two navies differ in their estimates of the total sea salt concentrations (i.e., concentration of particles of all sizes sampled) even though similar sampling methods were reportedly used. The British NMW-NGTE curve for total sea salt gives approximately the same concentration values, as Eqn. (6) of Tsunogai et al. (1972), for corresponding wind speed conditions.

Although severe storms are spectacular events, 5-year observations in the North Atlantic have shown, that in the ocean area bounded by 35°W latitude, 70°W longitude, and 66°N latitude, 2°E longitude wind speeds of more than 30 knots occurred 6% or less of the time, while those of more than 40 knots occurred only less than 1% of the time. The southern latitudes (40°-60°) always have high probability of greater

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* equivalent aerodynamic diameter" is defined as the diameter of a unit-density (1 g/cm^3) sphere, which has the same terminal velocity in a gravitational or inertial force field as the particle in question, without regard to its actual size, density, and shape.

** approximate concersions can be made by:

$\mu g/m^3 = 7.8 \times 10^{-4} \times$ ppm (by weight)

ppm (by weight) $= 1.28 \times 10^{3} \times \mu g/m^3$

than 20 knot winds all year around. Other areas of high wind probability are the North Atlantic and the North Pacific in the winter months, when the winds for the most part are prevailing westerlies (Kaufman and Pollini, 1967). According to such climatological information, the U.S.A. Navy feels that their ships on their predictable routes would be experiencing "heavy weather" (and about 0.06 ppm (by weight) sea salt concentrations) for only about 2% of their sailing time (Rogus and Weinert, 1972). On these transient high concentration occasions, the high wind speeds shift sea salt production towards the more massive particle sizes (Yoshimoto et al., 1976), which are also the easiest to control even with such simple devices, as the afore-mentioned turns in gas turbine air inlet ducts.

In contrast, certain types of high-speed naval vessels, as well as surface-effect ships (e.g., hovercraft and hydrofoils) generate continuously their own dense clouds of sea salt particles, which then combine with wind-produced particles and result in concentrations as high, as 50 to several thousand ppm (by weight) of sea salt (Kaufman, 1975). Fig. 9 shows a summary of measurements aboard ships by Yoshimoto et al. (1976), which reflect contribution of ship-generated sea salt particles to the overall concentrations encountered at various places aboard ships. It gives airborne sea salt concentrations for a wide range of wind and ship speeds, as a function of height above sea surface. Concentrations predicted from the afore-mentioned theories of Toba (1865b), and Chaen (1973) are also included for comparison. It appears, that the model estimates of concentrations reasonably approximate the measurement on drifing ships. When ships are sailing, sea salt particles are generated from the waves that the ship makes, and by the collisions with the bow or hull of the ship of wind-raised billows. The latter spray droplets, although not numerous, dominate sea salt concentrations because of their sheer massiveness aboard ships underway at high speeds. The majority

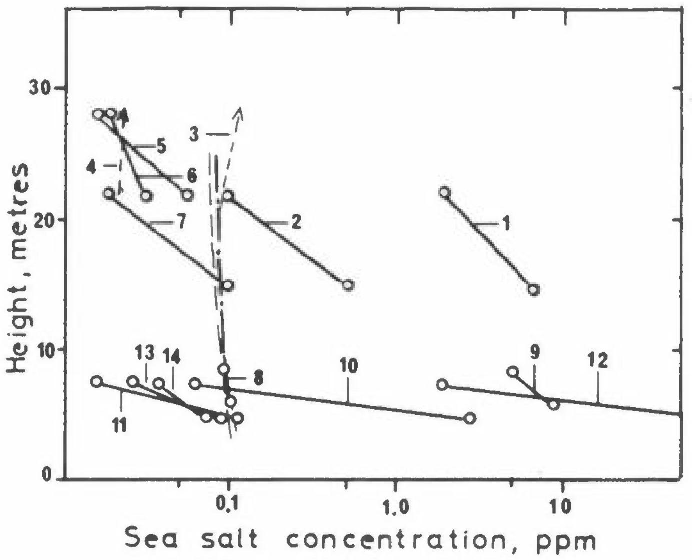

Fig. 9. Influence of wind speed, ship speed, and height above sea surface on airborne sea salt concentrations aboard ships; (after Yoshimoto <u>et al</u>., 1976).

Theoretical:

—·—·— : Toba (1965b) R.H. = 50%

— — — — : Chaen (1973) w_F = 5

Experimental:

———— : indicate range of concentrations.

	U	u_s	w_F
1	16	15	7
2	12	15	6
3	9	0-5	5
4	8.5	0	5
5	6	8-12	4
6	3.5	5-14	3
7	2.5	0-15	2
8	6	18-20	4
9	11	20-23	6
10	10.5	18-24	5
11	4	15-25	3
12	8.5	27-30	5
13	8.5	12	5
14	5	16	3

U = wind speed, m/sec
u_s = ship speed, knots
w_F = wind force (Beaufort scale)

of the sea salt concentrations, measured by Yoshimoto _et al_. (1976) aboard ships sailing at speeds up to 15 m)sec, lie above the British NMW-NGTE "total" particle line in Fig. 8.

In addition to the more common ship-board applications, the gas turbine has also met increasing favour among shore-based electricity generating plant planners (Hart and Cutler, 1973). Here much less corrosion damage has resulted, because of the lower levels of sea salt and fuel contamination encountered. Nevertheless, at some near-shore installations in Great Britain severe corrosion incidents have occured. Airborne sea salt measurements at these sites (Davies and Polfreman, 1973), however, found sea salt concentrations in the air insufficient to be the source of the problem, and fuel-borne sodium was the suspected culprit.

5.5 Airborne sea salt and the North Sea oil platform environment

The only measurements of airborne chloride and sodium on oil or gas platforms anywhere appear to be those reported by Peirson _et al_. (1974). Routine suspended particulate sampling, as part of a larger measurement program, was done on a natural gas platform in the North Sea (at 25 m above water level), located about 55 km from the English Norfolk coast and Petten in northern Netherlands. The result of the measurements, however, are reported as annual averages, which provide no information of the effects of wind speed or the range of airborne sea salt concentrations encountered at the measurement site. Furthermore, the authors suspected, that because of the type of air inlet used, the samplers did not collect a representative portion of the larger airborne sea salt particles. The annual (June, 1972 to May, 1973) average concentration of NaCl* at the platform site was 11.5 $\mu g/m^3$.

* the sum of the reported Na and Cl concentrations, and converted from ng/kg of air.

Similar long-average measurement results for two sampling stations on Shetland Islands have also been reported (Peirson et al., 1974), and may have some relevance. The NaCl concentrations were for Lerwick: 6.4 μg/m^3 (Jan.-Dec., 1972) and 5.4 μg/m^3 (June, 1972 to May, 1973), and for Collafirth: 5.6 μg/m^3 (June, 1972 to May, 1973).

Near-shore measurements of airborne sea salt concentrations, under various weather conditions and at various distances from the German and Danish coast of the North Sea have also been made. The concentrations have ranged from about 20 μg/m^3 (Schmidt, 1972) to 275 μg/m^3 (Neumann, 1940). A surprisingly high 2-year average concentration of 61 μg/m^3 was obtained by Bossolasco and Cicconi (1961). It can be assumed, that all of these results would be affected by contributions from their respective surf zones.

5.6 Methods and instrumentation

In principle, any measurement method suitable for the sampling and evaluation of airborne particles in the large and giant particle ranges might be applied to sea salt concentration and size distributions. The fact, that sea salt particles may be liquid some of the time imposes certain limitations or advantages on the selection of methods and instrimentation. On the other hand, the very definite chemical nature of sea salt (ca. 80% NaCl) enables and simplifies the application of certain procedures which would not be possible to use with particles of more mixed chemical composition.
Because the methods of sampling and evaluation of sea salt particles, and their variants, have been developed and described by many authors, only the chief principles applied will be outlined here.

There are two main approaches to airborne particle measurement: a) find the number and dimensions of the particles and infer mass; b) find the mass of particles (preferably in several size classes) and infer numbers, if needed.

In the (a)-approach: (i) particles are deposited in some manner (e.g., by impaction, thermal and electrostatic precipitation) on an appropriate substrate for subsequent microscopic evaluation (optical, electron, scanning). Special chemical treatment is often used for the substrate to produce a spot reaction with the deposited particle. This identifies the salt particles and often extends the lower size limit of detection; (ii) in situ devices, requiring no particle separation from the air sample, sense some property of single particles (e.g., light scattering, flame scintillation) which is related to the size of the particle; (iii) particles are photographed (e.g., by back-lighting photography, holography) within a defined volume, while suspended in air. Methods (i), (ii), and (iii) provide information on salt particle number concentrations and size distributions. In some cases, methods (i) and (ii) also enable chemical identification of individual particles.

In the (b)-approach: (iv) all particles in the air sample are collected by some separation device. Filtration is by far the most "popular" technique, but wet or dry impaction and electrostatic precipitation have also been used; (v) the particles are separated in relatively narrow size fractions, principally by aerodynamic classification with cascade impactors or centrifuges. Method (iv) will determine the total mass concentration of all sea salt particles (subject ot collection efficiency characteristics of the separation technique), while (v) gives particle mass concentration distribution over the various size classes. Any applicable chemical analysis procedure can be used for chemical component identification in the total mass sample of method (iv), or in the various particle fractions of method (v). A relatively recent review of sampling and evaluation techniques for the large and giant sea salt particles has been given by Mason (1971).
A general account of particulate matter measurement principles and the current commercial availability of instruments for particulate matter mass, size distribution, etc. determinations is contained in a Lawrence Berkeley Laboratory (1975) publication.

5.7 Examples of applications

The examples, illustrating the use of the various measurement methods, are given in this Section in approximately the same order, as the methods were enumerated in Section 5.6. In many studies, however, a combination of several methods and techniques has been necessary to achieve the objectives of the measurement program, and some overlap can be expected.

The sizes of the large and the giant sea salt particles allow the use of optical microscopy for their evaluation, although with some difficulty at the lower end of the large particle range.

1) In the "isopiestic" (equal pressure) method, the particles are collected on hydrophobic surfaces and then examined under the microscope in a known temperature and vapour pressure "chamber". By the measurement of the equilibrium size of the hemispherical droplets, it is possible to determine the weight of chloride in the individual particles (Houghton and Radford, 1938; Woodcock and Gifford, 1949). The accuracy of the isopiestic method has been experimentally investigated by Crozier (1954), and Toba (1966 b) and found adequate, provided the temperature in the humidity chamber can be accurately controlled. The sea salt particles are most commonly collected by impaction on thin glass strips or silver rods, coated with a hydrophobic film (Woodcock, 1952; Moore and Mason, 1954). The collection surfaces may be mounted on a windvane to orient the impaction surface into the airstream. The impaction efficiency is a function of the particle size, density, airstream velocity, width of the collector, and air viscosity (i.e., the Stokes' number). The counts of the collected particles must be corrected (Woodcock and Gifford, 1949), because of the rapidly decreasing collection efficiency with decreasing size of particles. With the air speeds and collector sizes utilized in most practical situations, particle smaller than about 1 μm in diameter (about 10^{-13} g mass) cannot be

collected. The method, however, has the advantage of simplified evaluation, because particles of wide size range are collected in about equal numbers on the impaction surfaces, due to the opposite trends in particle concentrations in the air and impaction efficiency (Junge, 1963).

The isopiestic method has been used for both ground- or sea-level and aircraft measurements of airborne sea salt. Various other types of collectors, such as spiders' threads (Dessens, 1949), sedimentation plates (Woodcock, 1952), and rotating and cascade impactors (Junge and Jaenicke, 1971), have also been employed in connection with the method.

2) The micro-chemical, or "spot test" method was first developed by Liesegang in 1896, and several variants of the method have been successfully used in air chemistry ever since. Basically, the method uses a strip of substrate material, which is coated with a gelatine film, treated with special chemical reagent forming a gel. Particles, collected on the gel, diffuse into the gel and produce a certain physico-chemical reaction, resulting in a circular spot of precipitate. The spot consists of a ring, or several concentric rings (Liesegang) around the original particle. The rings are seen under the microscope, or in photomicrograph enlargement, as halos, or often as coloured rings. The size of the reaction ring is larger, than the particle itself, thus enabling the detection and identification of sub-microscopic particles. The magnifications, i.e., the ratio of the size of the ring to that of the original particle, depends both on the chemical properties of the particle and the gel, and must be determined experimentally. The limit for chloride particles is generally 10^{-13} to 10^{-12} grams chloride mass, but some modifications of the method are reportedly capable of improved detection limits, down to 10^{-16} g (Lodge et al., 1954; Metnieks, 1958). Recently (Preining et al., 1976) spot size vs. aerodynamic diamter* (in the 0.5 to 2 μm diameter range) relationships

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* cf. p. 47 for definition.

have been established in the laboratory for chloride particles, produced from NaCl and sea salt solutions.
The reagent gels, as well as the methods for collecting airborne sea salt particles on the substrates have varied. Silver nitrate (Seely, 1952; Lodge, 1954), mercurous fluosilicate (Lodge et al., 1954; Lodge, 1955; Frank et al., 1972), and silver dichromate (Farlow's reagent) (Toba and Tanaka, 1967; Hama and Takagi, 1970; Chaen, 1973) gels have been used. The salt particles have been impacted on gel-coated glass slides or photo film, either from the free airstream, or in single- or multi-stage impactors (Crozier et al., 1952; Boyce, 1951; Reitan and Braham, 1954; Durbin, 1959; Podzimek and Cernoch, 1961; Singleton and Durbin, 1962; Metnieks, 1958; Toba and Tanaka, 1967; Chaen, 1973). Hama and Takagi (1970) impacted the particles on Farlow reagent-treated photo film in a sequential automatic sampler, while Rau (1956b), and Schmidt (1972) deposited them on glass slides by means of specially designed electrostatic precipitators.
The short-coming of the spot test method is, that all halides will give precipitates with these reagents. However, in view of the geographical locations of sea salt sampling sites and particularly under onshore winds, it is highly unlikely that there can be any significant sources of such particles other than the sea.

Glass slides, coated with magnesium oxide, have also been used for salt particle size measurement (Moore, 1952; Chaen, 1973; Yoshimoto et al., 1976). The diameters of the impacted particles are deduced from the sizes of the craters produced in the oxide film by the impacting particles.
Mass of salt in each droplet may also be calculated on the assumption, that at the impact each drop consisted of sea salt saturated with NaCl.

Electron microscopy has been used for some airborne particle investigations (Junge, 1954; Heard and Wiffen, 1969; Frank et al, 1972; Meszaros and Vissy, 1974; Hobbs et al. 1976). The chemical nature of the particles (dry crystals, in the case of sea salt) can be determined by morphological indentification.

For in situ sea salt particle counting and sizing (i.e., while they are suspended in the air) both the optical (light scattering) particle counters (Junge et al., 1969; Junge and Jaenicke, 1971; Kojuma et al., 1974; Hobbs et al., 1976) and flame photometric particle counters (Vonnegut and Neuberger, 1953; Woodcock and Spencer, 1957; Mason, 1971; Bodhaine and Pueschel, 1972; Doman, 1975) have been used. These devices are of particular value where unstable particulate systems have to be examined and traced, because they make measurements in nearly real time. The information obtained is also in a very usable form for particle number and mass distribution assessment. In the optical counters, the particles are passed in single-file through a small sensing volume, where they are illuminated by an intense light beam. The light scattered by the particles at some angle (90^o and near-forward scattering angles are most frequently used) is detected by a photomultiplier. Each light pulse represents a single particle and its amplitude the size of the particle. The proportionality between pulse heights and particle sizes must be determined by calibration with particles of known sizes and optical properties. The photomultiplier pulses are electronically sorted into various pulse heights, representing particle size ranges. The range of detection is usually from 0.3 μg to about 15 μm in diameter.

The optical counters have certain non-ideal characteristics, that must be considered in their use (Whitby and Vomela, 1967; Lieu et al., 1974). These include particle refractive index and shape effects on the accuracy of size measurements, and channel cross-sensitivities, causing smoothing of the distributions.

The flame photometric counter passes the particles in single-file through a hydrogen flame. The light flashes emitted by each of the scintillated particles are passed through an appropriate narrow-band filter and detected by a photomultiplier. For example, the characteristic yellow light emitted by sodium-emtaining particles has a strong component at 5890 Å and 5896 Å. The photomultiplier signal processing is similar to that of the optical counters. Again, the pulse heights proportional to the mars of chemical substance in the particles must be calibrated particles of known size and appropriate chemical composition. A more sophisticated version of the instrument, the flame scintillation spectral analyzer (Binek, 1970), can give information not only particle size distributions, but also at least in theory, on the particle chemical composition. In practice however, one of the two has to be known or determined by some independent means. In the case of sea salt measurements, the use of pure NaCl particle for pulse height calibrations should be taken into account in data evaluation (Doman, 1975). The practical lower size limit for the simple type flame photometers is about 1 µm in diameter,while the flame scinitillation analyzer has reportedly a range from 0.05 to 10 µm diameter (Doman, 1975). When using these two types of instrument in airborne sea salt studies, the assumption again is made, that all the particle measured are sea salt.

The use of direct in situ photography of airborne sea salt has been infrequent. Monahan (1968) used a raft-mounted photographic system to measure large airborne droplets (of radius greater than 45 µm) close to the sea surface. The backlighting or "shadowgraph" system he used, produced images of the droplets that appeared on the film negative as bright disks with a black dot at the centre against a dark background. The sizes of the images must be then transformed by appropriate mathematical treatment to actual drop sizes. In some instances, Monahan was forced to combine several observations to obtain a single particle size distribution, because of the sparsity of droplet images on each photograph.

Because sea salt is readily dissolved and aborbed in water, airborne sea salt has been collected by liquid "scrubbing" or impingement for total mass concentration determinations (Jacobs, 1937; Tsunogai et al., 1972; Whelan, 1975).

After an initial attempt to separate sea salt particles by means of filtration through a tube partially filled with glass wool, Jacobs (1939) found, that a "wash tower" arrangement was the most convenient and accurate sampling method. The wash tower was made up from a cylindrical glass vessel with a sintered glass filter fitted over the bottom end of the air inlet tube, immersed in about 500 ml of distilled water. The sintered glass broke up the sample air stream into fine bubbles. For his surf zone sampling location, Jacobs used sampling runs of 6-12 hour duration, and operated the sampling train at a flowrate of around 6 litres/min. to collect adequate samples for chloride analysis. Tsunogai et al., (1972) used a similar sampling train, with a washbottle, having sintered glass bottom, and containing 50 ml of distilled water. Most of the sea salt mass and some gaseous constituents in the air were collected. At sampling rates of 0.5-1.5 litres/min, 20 to 50 hour sampling runs were found necessary to collect sufficient sample over the open ocean.

Whelan (1975) sampled surf zone sea salt with the standard Greenburg-Smith impingers, filled with 250 ml double-distilled water. A sampling rate of 12 litres/min. and run duration of 3 hours were used. Whelan found[*] that the Greenburg-Smith impingers had only about 60% collection efficiency for sea salt when the particle concentrations were less than about 25 μg Na/m^3. This may have been partly due to the low sampling rates employed, since the design flowrate of Greenburg-Smith impingers for best collection efficiency for particulates is 28 litres/min.

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[*] T. Whelan, III, Coastal Studies Institute, Louisiana State University, Baton Rouge, La.; personal communication, May, 1976

The method of choice for total sea salt concentration determinations has undoubtedly been filtration. The filtration method has many advantages to offer the user in terms of wide choice among the types (foam, fibrous, membrane, nuclear-pore) and materials (glass, organic, metallic, synthetic) of filtering media; flexibility in selection of filter sizes, shapes, airflow resistance, surface and blank characteristics; generally high collection efficiency; wide range of possible sampling flow rates; and ease of preparation for chemical analyses. The fibrous cellulose (Whatman-type) filters usually have low blanks in interfering substances, are easily processed and are preferred for chemical analyses. They are, however, moisture-absorbent and thus practically unsuitable for gravimetric sample evaluation. In airborne sea salt studies, they have been used by Peirson _et al_., (1974), Buat-Menard _et al_. (1974), Levaggi _et al_. (1975), Vitols (1976), among others.

Glass fibre filters are virtually unaffected by moisture, but may have high blanks (e.g., for chloride). The filters have very low flow resistance and are particularly suitable for high flowrate sampling, but they cannot be subjected to repeated flexing and they tear easily. Sheet-sized glass fibre filters have been used for many years in the U.S.A. for sampling suspended particulate matter with the so-called High-Volume samplers*. The ready commercial availability of the sampler is reflected in its use in numerous sea salt investigations (Holzworth, 1959; Lodge _et al_., 1960; Barger and Garrett, 1970; Hidy _et al_., 1974; Vitols, 1976.

The use of synthetic fibre polystyrene (Simpson, 1972; Hoffman and Duce, 1972; Hoffman _et al_., 1974), high-purity fibre polyethylene (Delaney _et al_., 1971), and urethane foam (Yashimoto _et al_., 1976) as filtering media has also been reported. Organic membrane filters of the Millipore-type have been

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* Cf. for example, Lawrence Berkeley Laboratory (1975).

extensively used in airborne sea salt studies (Lodge et al., 1954; Lodge, 1955; Haryslak and Pollini, 1967; Tsunogai et al., 1972; Semonin, 1972; Rossknecht et al., 1973; Okita et al; 1974; Lovett, cf footnote p. 25). In addition to beeing available in a wide range of controlled pore size (and thus giving absolute collection of particles larger than a given pore size), membrane filters are essentially surface filters and thus suitable for collecting particles for microscopic examination. Furthermore, they can be readily treated or impregnated with chemicals (e.g., for spot testing), dissolved in acetone, and made transparent by the application of liquids of suitable refractive indes. Ease of processing and low blanks of the membrane materials make them suitable for most inorganic chemical analyses. The flow resistance of membrane filters, however, is considerably higher than that of fibrous filters at comparable flowrates, and, therefore, they are principally used in low flowrate filtration applications. They also acquire electrostatic charge readily, which may cause difficulties in gravimetric evaluation.

Particles, suspended in a moving air stream, tend to follow the flow lines of the air around obstacles in their way to a varying degree, depending largely on the inertia of the particles and the aerodynamic drag experienced by them (i.e., the trajectory taken by a particle is a function of Stokes' number). The practical utilization of the inertial properties of airborne particles, in order to capture them on obstacles, has taken several different paths.
The impaction techniques of particle collection, described in connection with the isopiestic and spot test methods, for the most part relied on the wind speed or the movement of an aircraft to create the relative velocity between the particles and the obstacle, necessary for particle impaction to take place. A rather seldom used technique, which employs the same impaction principle, is known as "flying a kite" (Prospero, 1968; Parkin et al. 1970; Wilkness et al. 1974). The "kite" is a mesh of nylon or terylene material, consisting of 0.33 - 0.5 mm diameter monofilament fibres, woven with 50% open area.

Such kites have been "flown" at fixed shoreline sampling sites, or from the bows of ships. Large volumes of air are sampled in this way by the 1-7 m^2 areas meshes during the customary 24-hour sampling periods. A 50% collection efficiency is usually assumed for particles larger than about 2 µm in diameter. After sampling, the mesh is rinsed in distilled water and the soluble sea salt fraction separated from the insoluble "dust".

The rotating (or rotary) impactor (Noll, 1970; Junge and Jaenicke, 1971; Wilber et al., 1974) generates impaction velocities (Which are relatively independent on existing wind speeds) by twirling impaction surfaces, afixed to the ends of rotating arms, through the air. Because the rotating impactor samples large volumes of air in relatively short periods of time, it is particularly useful for the collection of the massive airborne particles at the upper end of the giant particle range, which normally occur in very low number concentrations in the air.

The functioning of the above impaction devices is characterized by the collection of particles from an unconfined air stream. These devices are sometimes called "external flow" impactors. The "internal flow" impactors impart the necessary impaction velocity to the suspended particles by directing the airstream through a confined passage or "jet" onto the impaction surface. These type of impactors have been used in single- or multi-stage configurations. If the impactor has more than two stages (i.e., collection surfaces) in a series arrangement, it is commonly referred to as "cascade impactor". The collection stages can be used dry, coated with adhesive materials, covered with appropriate substrate (Dams et al., 1972), or submerged in liquids. The aforementioned Greenburgh-Smith impinger is an example of a single-stage impactor, utilizing a liquid to retain impacted particles.

Although single-stage impactors (Crozier et al., 1952; Blifford, 1970; Blifford and Gillette, 1972; Kikuchi and Yaura, 1970; Toba and Tanaka, 1967; Chaen, 1973) and 2-stage impactors (Junge, 1954; Junge and Jaenecke, 1971; Yano et al., 1974) have been used for airborne sea salt investigations, it has been the cascade impactor, which has been most frequently chosen for sea salt size-mass concentration distribution determinations and ionic fractionation studies.

Cascade impactors, originally designed by May (1945), are now commercially available in many types of stage configurations, and for a wide range of sampling flowrates. The theoretical basis of their performance and design is now well established (Marple and Liu, 1974), and their true operating characteristics under non-ideal conditions are better understood (Winkler, 1974; Dzubay et al., 1976; Natusch and Wallace, 1976; Willeke and McFeters, 1975; Marple and Willeke, 1976).

Cascade impactors separate airborne particles in relatively narrow aerodynamic diameter* fractions by impacting particles of progressively smaller sizes on consecutive stages of the impactor. This is achieved by reducing the cross-sectional areas of the impaction jets, and thus increasing the impaction velocities. The impaction stages of cascade impactors can have single or multiple jets, which may be in the shape of round holes or long, narrow slots. The collection surfaces are usually flat plates, but cylindrical, or even conical surfaces have also been used (Marple and Willeke, 1976). Cascade impactors with 4 to 8 stages have been used in airborne sea salt studies.

The interpretation of measurement results from cascade impactor sampling presents some complications, because of the hygroscopic nature of sea salt particles, which have been discussed by Duce et al., (1967), and Haenel and Gravenhorst (1974).

Although the ideal cut-off diameters** of each cascade impactor stages can be predicted from theory (Marple and Liu,

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[*] cf. p. 47 for definition

[**] usually the so-called ECD of "50% cut diameter" is used to characterize the collection performance of impactor stages, i.e., the equivalent aerodynamic diameter of particles, which are collected with 50% efficiency on a given impactor stage.

1974; Marple and Willeke, 1976), calibration with particles of known aerodynamic sizes is usually necessary to assess the actual separation characteristics of the impactors. Cascade impactors (and other internal flow impactors) must be operated at either the calibration flowrate, or stage cut-off size corrections must be made for other conditions. Depending on the particular design of the cascade impactor, operating flowrates range from 1 litre/min. to about 1130 litres/min. (Swartz et al., 1973; Willeke, 1975). Judging from accounts in the literature, the most frequently used cascade impactors in sea salt studies have been those, which are operated at relatively low sampling rates (12-28 litres/min). Use of the modified May-design Casella impactor (4 stages, single rectangular -slot jets) has been reported by Metnieks (1958), Feorgii and Metnieks (1958), Duce et al. (1965), Kaufman and Pollini (1967), Haryslak and Pollini (1967), Larssen (1974), Lovett (cf. footnote, p. 27), and Yoshimoto et al. (1967). Winchester and Duce (1966), Lininger et al. (1966), Andren and Harriss (1971), Martens et al. (1973), and Van Grieken et al. (1974) have been among the users of the Andersen Sampler (6 stages, multi round-hole jets). The utilization of the Batelle-design cascade impactor (6 stages, single round jets) has been increasing in recent years, as described, for example, by Duce et al. (1967), Duce and Woodcock (1971), Woodcock et al. (1971), Wilkness and Bressan (1972), Chesselet et al. (1972a,b), Gladney et al. (1974), and Hardy et al. (1976). A 5-stage modification of the Andersen Sampler (Lee and Goranson, 1972) and the high-volume Sierra cascade impactor (5 stages, multi rectangular-slot jets) were used by Vitols (1976) for sea salt particle size distribution measurements.

Of the various types of aerosol centrifuges (Which apply centrifugal force to the particles to classify them according to aerodynamic sizes), only the Goetz "aerosol spectrometer" has been used by Junge et al. (1969) for determining chloride containing particle size distributions. The practical upper size limit for this classifier is about 3 μm diameter, but its lower limit is about 0.1-0.2 μm in diameter.

Although the integrating nephelometer does not measure directly either the number or mass of airborne particles, the measured light scattering coefficient can be related to mass concentrations (Charlson *et al.*, 1969) The instrument is also useful for semi-quantitative *in situ* assessment of the presence in air of deliquescent and hygroscopic particles, such as sea salt, under changing conditions of relative humidity. Its use has been reported by Pueschel *et al.*, (1969), Pueschel and Mendoca (1972), Pueschel *et al.* (1973, and Hobbs *et al.* (1976).

6 DISCUSSION

6.1 Results of airborne sea salt measurements

The great variability of airborne sea salt mass concentrations in space and time can be perceived from Fig. 4. In addition to the wide fluctuations and uncertainties in surf zone and coastal sampling results, the measurements representing the open ocean conditions, i.e., those within the Woodcock-Junge-Chesselet *et al.* range, show a scatter that spans almost one order of magnitude. It also is by no means certain, that data obtained at the same location, by the same methods and instrumentation, and under reasonably similar weather conditions, but on different occasions will agree (within limits of experimental errors). For example, Lodge (1955), in a repeat attempt to study the details of the earlier discovered sea salt concentration maximum above the sea surface, failed to find one at all. Another case in point is illustrated by the large differences between the U.S. and British navy data on total sea salt concentrations under various wind conditions (Fig. 8). At least part of the data were reportedly obtained during collaborative measurements using similar methods aboard the Danish frigate *Peder Skram* on its voyage from Kopenhagen to the U.S.A.

In regard to North Sea oil or gas platform environment, the data of Peirson *et al.* (1974) on Na and Cl mass concentrations at the gas platform site between England and The Netherlands, appear to be the only pertinent ones available at this time. Unfortunatley, these are annual averages, and although monthly averages might be available from the authors, the smoothing of extreme values would be still too great to estimate sea salt concentrations during periods of peak wind speeds. As noted earlier, the representativity of the samples (at least as far as airborne sea salt is concerned) is also in question; because of possible discrimination of the sampling system against the aerodynamically larger particles.

The similar annual average concentration data from the Lerwick site on Shetland Islands in the North Sea may reflect the mixed influences of surf zone sea salt production, transport distance inland (1/4 km), and height above sea surface (76 m) (Cawse, 1974). No particle size distribution determinations have been made at either of these sites.
Some estimates of airborne sea salt mass concentrations and size distributions could perhaps be made from the results of ship-board observations at various heights above the sea surface by Yoshimoto et al. (1976), as presented in part in Fig. 9. These measurements represent short-period sea salt concentrations, inclusive of the ship-generated component. Since there is wave breaking against the supporting structures of an oil platform, sea salt concentrations measured aboard drifting ships could to some extent approximate the oil platform environment, except for dissimilarities in heights above the sea surface.

It is clear then, that only range estimates (and rather wide ones at that) of airborne sea salt concentrations are possible for a given location and wind speed from the information currently available in the literature. Where reliable short-period data on concentrations and size distributions of sea salt at a specific site are required, the need for measurements with appropiate instrumentation and spanning a variety of weather conditions is indicated.

6.2 Methods and instrumentations

As Kaufman has stated*, there are two ways of asking questions about airborne sea salt concentrations:

What is sea salt concentration in air in all forms?

What is sea salt concentration in air in aerosol form?

The selection of appropriate methods and instrumentation will then depend on which question must be answered. In other words, on whether the objective is to measure the entire spectrum of sea salt particles in the air, or is information on only a selcted range of particle sizes required. Thus, for example, if the massive sea salt particles, down, say, to 5 µm diameter, are indeed effectively removed by inlet ducting of gas turbines, a method which measures efficiently and accurately the aerosol fraction of sea salt will provide all the information a turbine inlet separator designer needs. On the other hand, if a representative sample of all airborne sea salt particles is required, the necessity of isokinetic sampling (Fuchs, 1975) is clearly indicated for the giant particle range. With the sole exception of an aircraft sampling system (Allee, 1974), isokinetic sampling has apparently never been attempted in sea salt studies, although it is doubtful, that any of the methods used for airborne sea salt mass concentration and size distribution measurements have provided "true" information in this regard. The exceptions are, perhaps, the "external flow" impactors, when corrections for incomplete particle collection are appropriately made.

A survey of the sea salt literature leaves one with the impression, that developments in the technology or particulate sampling have not kept pace with the rather spectacular advances in chemical analysis techniques in recent years. And yet, the best analysis cannot improve the quality of the sample!

The main tool for airborne sea salt measurements today, the cascade impactor, is now more than 30 years "old".

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* R.E. Kaufman, NAVSEC, Philadelphia, Pa.; personal communication, May, 1976

Nevertheless, it has been only recently, that non-ideal performance of most cascade impactors under real field conditions has been recognized (Marple and Willeke, 1976). Apparent differences in sampling results, when using samplers of different designs to sample the same atmosphere, have also been noted (Hidy _et al_., 1974; Miller and DeKoning, 1974; Vitols, 1976). Such "sampler effects" might be largely cause by differences in sampler intake efficiencies*, but very little is known about the intake efficiencies of most particulate sampling systems under varying wind and particle size conditions (Steen and Andreasson, 1974; Steen and Johansson, 1975; Ogden and Wood, 1975; Agarwal, 1975).

It is true, that more advanced _in situ_ devices, e.g., the optical and flame photometric counters, are now available. Unfortunately, in their present state of development, they remain essentially laboratory instruments, unsuitable for the rugged and hostile conditions in the field. It is also questionable, whether representative samples are obtained with these devices. To avoid coincidence of particles in their sensing zones, these instruments usually utilize very low sampling rates. This can result in substantial losses of particles in the rather complex plumbing of the instruments, before the particles reach the sensing zone, and in contamination of the instrument (Kaufman, 1969). Because of the low sampling rates, these instruments also have very poor (a few percent for particles of 10 μm diameter and larger) intake efficiencies (Liu _et al_., 1974). Attempts to improve the intake characteristics by the use of specially designed inlet systems make the instruments even more complex and unwieldy.

Thus, the need is urgent for the development of reliable, rugged, and sufficiently portable _in situ_ sea salt measurement devices (Kaufman, 1975).

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* "intake efficiency" is the ratio of the particle concentration measured by the instrument to the true particle concentration in the original air sample prior to entrance into the instrument.

The holographic technique (Thompson et al., 1967) is now reportedly capable of resolving particles down to about 0.3 μm in diameter*. Automated assessment systems are replacing the tedious and lengthy manual methods for analyzing the reconstructed images of the particles (Bexon et al., 1976). This technique would provide a means for direct and in situ evaluation of airborne sea salt particles while they are still suspended in the ambient air.

It is clear, that the size of airborne sea salt particles is a most important parameter, which governs their airborne behavior and thus the transport, removal, and the various effects sea salt produces. The currently used microscopic methods provide direct assessment of sea salt number distributions, but are too tedious and time-consuming, and require certain assumption for mass determination of the particles. For the time being then, the cascade impactor remains the instrument of choice for airborne sea salt mass distribution measurements, provided the sampling system has appropirate intake characteristics. The cascade impactor is particularly suitable for studies of sea salt ingestion in gas turbines, because the aerodynamic size information it provides is of prime importance to designers of airborne sea salt separation equipment. A further advantage in its use is due to the mostly liquid droplet state of sea salt particles, under the relative humidity conditions prevailing over the oceans and in near-shore coastal areas with onshore winds. Cascade impactors have high retention efficiencies for liquid droplets, because particle bounce-off and reintrainment of already collected particles does not occur (Winkler, 1974; Dzubay et al., 1976; Marple and Willeke, 1976).
Stricter attention must be paid to the objectives of sea salt measurement programs when selecting samplers and sampling systems. Where information on airborne sea salt over most of its size spectrum is desired, attempts will

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* e.g., advertisement of Laser Holography, Inc., Santa Barbara, California, U.S.A.

have to be made utilize near-isokinetic (Steen and Andreasson, 1974) or at least directional (Sehmel, 1973) sample intakes, to avoid, at least partially, sampler effects, which can be pronounced for the aerodynamically large sea salt particles.

Sample contamination from a variety of sources, particularly in ship and ocean platform environments, is a constant threat (Moyer _et al_., 1972; Rasmussen and Allwine, 1976). With the low-flowrate cascade impactors, very small amounts of sea salt might be collected on each of the impaction stages and contamination while handling and processing the samples is easily possible. Thus, the devising and meticulous execution of procedures for sample handling and treatment are just as important, as the sampling phase itself.

7 CONCLUSIONS

The information examined in this literature survey of airborne sea salt measurements shows that:

1. Airborne sea salt has many diverse effects, both over the oceans and the continents;
2. Mechanical processes at the air-water-land interface of the world's oceans are the principal sources of airborne sea salt;
3. Major sink mechanisms for airborne sea salt are wet removal by rain-out and wash-out, and dry deposition by sedimention and impaction on surfaces;
4. Concentrations of airborne sea salt over the open oceans are mainly a function of wind speed, but local shoreline influences can be significant;
5. Vertical sea salt distributions over the sea, and the vertical and horisontal distributions inland from the coast are aproximately exponential; virtually no sea salt is normally found over the oceans above 3 km altitude;
6. Theoretical models exist for the prediction of vertical and horizontal sea salt particle concentration distributions;
7. The existence of an apparent sea water ion fractionation in airborne sea salt and its causes remain controversial;
8. Despite the very large number and geographical distribution of sea salt measurements, for a given location and wind climatology only broad range estimates of airborne sea salt concentrations can be made according to currently available information;
9. Airborne sea salt concentration and size distribution data, specific to the North Sea oil and gas platform environment, are almost totally lacking;
10. Measurement methods and instrumentation are available for sea salt particle number and mass concentration, and size distribution assessment;

11. Selection of measurement methods and instrumentation must be dictated by the objectives of the study and information requirements;
12. Most current techniques for sea salt mass assessment do not sample airborne sea salt particles representatively over their entire size spectrum;
13. Cascade impactors are capable of providing sea salt mass concentration information in the aerosol size range of relevance to gas turbine protection equipment designers;
14. Appropriate procedures for avoiding sample contamination must be an essential part of measurement programs;
15. Development of reliable and portable *in situ* airborne sea salt monitors, suitable for the rugged marine environment, is lagging, but is urgently needed.

8 RECOMMENDATION

It is recommended, that three additional sources of information, not available for this study, be examined for relevant information on airborne sea salt:

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(2) Lovett, R.F. The occurence of airborne sea salt and its meteorological dependence. M.Sc. thesis, Heriot-Watt University, Glasgow, Scottland.
(Note: Lt. Cdr. Lovett's thesis has not yet been published.)

(3) Relevant papers, presented at various cooling tower symposia, conferences, and meetings, on drift measurement techniques and instrumentation.

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