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THESIS

A COMPARATIVE STUDY OF THE COASTAL MARINE AEROSOL

by

Alan Anthony Simoncic

December 1977

Thesis Advisor:

Kenneth L. Davidson

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A Comparative Study of the

Coastal Marine Aerosol

by

Alan Anthony Simoncic Captain, United States Air Force B.S., United States Air Force Academy, 1970

Submitted in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE IN METEOROLOGY

from the NAVAL POSTGRADUATE SCHOOL December 1977

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ABSTRACT

Aerosol size distributions near the coast of Panama City, Florida and off the Southern California coast near the Channel Islands are investigated in this study. The relationships of the coastal marine aerosol to wind speed, relative humidity, stability, and sub-synoptic circulation are examined. Relative humidity and stability are shown to have the largest effect on the aerosol distribution during periods of light winds. Coalescence and sedimentation of droplets greater than 1.5 µ radius are most pronounced when the wind speed and sea surface production of salt nuclei are weak. When wind speeds exceed 7 m/sec, a state of equilibrium between sedimentation and production of these larger droplets appears to exist. An apparent zone of transition between the two bubble bursting sea-salt producing mechanisms is observed near .5 µ radius. The highest correlation between wind speed and particle concentration occurs under unstable conditions. Secondary circulations are shown to be important determinants of the coastal marine aerosol in the absence of synoptic scale forcing.

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I. INTRODUCTION

The military is currently very interested in the performance of electro-optical weapons systems in an atmosphere of varying turbidity. For example, a number of electro-optical systems which utilize the visible as well as IR wavelengths are being developed by the Navy for use in surveillance and intelligence gathering operations in the marine boundary layer. These systems are limited by the extinction of the propagated energy due to absorption and scattering by aerosols. The effect of absorption depends on the composition of the particulates and wavelength of the energy and the effect of scattering depends on the concentration and size of the scatterers. For most applications the scattering processes in the atmosphere are caused by particles of size comparable to the wavelength of the radiation.

The size distribution of the marine aerosol is known to depend upon the wind speed, relative humidity, stability, and air mass trajectory. In order to evaluate accurately and predict the atmospheric effects on these electro-optic systems, it is necessary to know the dependence of the aerosol size distribution on the foregoing meteorological parameters.

The nature of the aerosol size distribution in a coastal marine environment is investigated in this study. Data from aerosol observations off the coast of Panama City, Florida and off the Southern California coast near the Channel Islands

were analyzed. These coastal regimes, which represent a mixture of continental and marine aerosols, should contain aerosol distributions somewhat different from the typical marine environment. The relationship of the coastal marine aerosol to wind speed, relative humidity, stability, and sub-synoptic circulation is examined. Furthermore, an attempt is made to evaluate the use of the friction velocity as a valid aerosol distribution predictor.

II. BACKGROUND

A. THE ATMOSPHERIC AEROSOL

With the recent increasing concern over the pollution of our atmospheric environment, the examination of the tropospheric aerosols has also increased. Particulate matter enters the atmosphere through either natural or man-made processes; approximately 10% of the total concentration is believed to originate from combustion and industrial processes while the natural sources, including soil dust, volcanoes, and oceans account for the remaining 90%. The size range of aerosols observed by current methods extends from $10^{-3} \mu$ to $10^{3} \mu$ radius (1 $\mu = 10^{-6}$ m = micron). Depending on their size, amount of soluble matter, and the relative humidity, these particles may act as condensation nuclei and aid in the precipitation process.

Mason (1975) classified condensation nuclei into three groups according to radius: Aitken (< 0.1 μ), Large (0.1 μ - 1 μ), and Giant (> 1 μ) particles. Essentially, Aitken nuclei are produced by man-made sources and larger nuclei by natural processes. Therefore, it is not surprising to see Aitken nuclei dominate the size distribution spectrum over continents. The marine aerosol above .1 μ is composed of sea-salt particles produced by spray and bubble bursting mechanisms on the water surface. These mechanisms are quite complex and their contribution to the size distribution will be discussed in detail later.

The vertical profiles of trace constituents that are produced over the continents have been shown to be rather uniform in space and time above 5 km. This "background" aerosol is affected slightly by anthropogenic activities and is far away from local natural sources. Its number concentration is almost identical with the concentration of Aitken nuclei over ocean areas. Past experiments have shown the concentration of the background aerosol to be about 300 cm⁻³ over remote ocean areas. However, a recent experimental cruise (R/V Meteor) by Junge and Jaenicke, 1971 in the mid Atlantic vielded observed concentrations of 600 cm⁻³. Measurements by Hidy, et al. (1973) on San Nicolas Island, 130 km, westsouthwest of Los Angeles, have shown the background aerosol to be a mixture of material from both marine and continental sources with an average concentration of 2400 cm⁻³. Samples taken over oceans of the South Atlantic (Meszaros and Vissy, 1974) resulted in Aitken particle counts of between 300- 450 cm^{-3} .

B. CHARACTERISTICS OF THE MARINE AEROSOL

An idealized size distribution of the continental and marine aerosol is supplied by Junge (1972) in Figure 1. The significant feature is the shift of the maximum of particles as a function of total particle concentration. Over the ocean the sea-salt aerosol, which is usually confined to the lower 2 km, is superimposed on the background aerosol. Junge reasoned that the concentration of the background



Figure 1. Idealized Size Distributions of Continental and Marine Aerosols

cloud nuclei decreases below the marine inversion due to the effect of washout, or coalescence, due to the larger water droplets in the cumulus clouds.

Another aspect of the aerosol distribution is the slope of the number density versus radius curve. Friedlander (1961) proposed a theory of self-preserving size distributions which helps to explain why all atmospheric size distributions are similar. He proposed that the similarities can be explained by solutions to the kinetic equation which describe the relationship between particle size distribution and time. Experimental results have indicated that the size distribution over a particular range of sizes of continental aerosol has a -4 slope and follows the relation

$$\frac{dN}{dr} = C\phi r^{-4}$$
(1)

where N is the number of particles/cm³, r the particle radius, C a constant, and ϕ the volume of particles per unit volume of aerosol.

Blifford (1970) measured the size and number distributions of atmospheric aerosols at various altitudes over the ocean 250 km west of Santa Barbara, California. Samples were taken by an aircraft equipped with a jet impactor and the data was obtained from direct microscopic counting techniques in the laboratory. The aerosol distribution at approximately 15 meters above the sea surface is presented in Figure 2. The curve has a rather steep negative slope at



Figure 2. Size Distribution at 15 Meters, 250 km West of Santa Barbara

the small particle end which becomes slightly positive at around .4 μ radius. For particles larger than .8 μ , a fairly constant slope of about -2 to -3 is observed.

The results of the R/V Meteor experiment, where several aerosol counters were used, are shown in Figure 3. Above 10 μ the exponent of a power function fit to the data is approximately -6 and between 0.3 μ and 10 μ it is variable but on the average around -3. The maximum of the size distribution occurred at 0.3 μ with a secondary maximum at 0.03 μ .

It is possible that, due to increased human activity in the Northern Hemisphere, Junge and Jaenicke's Atlantic experiment did not explore the undisturbed marine environment. Meszaros and Vissy (1974) describe the results of aerosol samples taken over the oceans of the Southern Hemisphere by means of membrane filters. An example of the number concentration and size distribution over the Atlantic between (a) 0° and 20° South and (b) 40° and 60° South can be found in Figure 4. Chemical analyses were performed and it was observed that the maxima in the concentrations of all particles and of sodium chloride particles occur at approximately .1 µ radius in both cases. Up to .5 µ radius the slope of the distribution is approximately -5. Between 0.5μ and 1.5μ , however, the decrease of the concentration with increasing particle size is very moderate (-1 to -2), while for radii larger than 1.5 μ the slope is close to -3. This has been interpreted to indicate that the form of the



Figure 3.

Results of the R/V Meter Cruise. Measurement Systems Used: ---- Combination of CCN Counter, Optical Counter, and Impactors: --- Double Stage Impactor (Junge and Jaenicke, 1971)



Figure 4. Size Distributions over Remote Ocean Areas (— All Particles; — NaCl Particles)

distribution is produced by the combined effect of particles formed in different ways.

Oceanic measurements have shown that the concentration of sea-salt particles decreases exponentially with height, with little variation of the size distribution. Ericksson (1959) reported that there exists a level a few hundred feet above the surface where the concentration decreases with height in periods of high wind force and increases with height in lower wind forces. He reasoned that there is little or no production of sea-salt in regions of light winds and that coagulation and fallout in the lower levels combined with horizontal transport due to vertical shear produce a maximum concentration at some upper level.

Toba (1965a and b) proposed that the average decrease in concentration with height can be explained by a combination of sedimentation, diffusion, convective processes and the humidity distribution. He suggested that the line between the aerosol vertical distribution and the process of production of sea-salt particles at the sea surface is found within the lowest layer of the atmosphere where the eddy diffusivity and relative humidity sharply change.

The distribution of eddy diffusivity near the sea surface is closely related to the wind speed. The larger the eddy diffusivity near the surface the more sea-salt particles that will be supplied. Toba considered eddy diffusivity in the form

$$D = kU_{*}(Z + Z_{0})$$
 (2)

where k is the von Karman constant, Z the height above the sea surface, Z_0 the roughness length, and U_* the friction velocity which is a function of the momentum transfer over the sea.

The relative humidity in the first few meters over the ocean is known to decrease rapidly with height. The particles produced near the surface in a region of high humidity grow larger and thus have a greater terminal velocity due to gravity than those at the top of this layer.

During light winds the number concentration near the sea surface increases with height. Since it results from a nonsteady state, an inversion of vertical gradient of the particle concentration is most likely to be found in small particles which have a longer residence time. Ericksson (1959) computed the fall velocities for given relative humidities, salinity, and radius. During high wind periods, giant size sea-salt particles are produced at the surface and through the diffusion process are mixed throughout the atmosphere. The largest particles may fall back into the ocean due to excessive terminal velocity or be entrained in the wave crests. Smaller particles are free to rise to cloud height where coalescence with larger cloud drops and washout usually occur.

Measurements of salt nuclei greater than 10⁻¹⁴ gm over the North Atlantic by Moore and Mason (1954) revealed the existence of two distinct types of size distributions (Type I and II). The curves for the observed Type I and Type II

nuclei distributions are reproduced in Figure 5. Type I distributions were observed for wind speeds between 6-15 m/sec and were thought to be residuals of spray droplets produced by breaking waves. The presence of a discontinuity or a sharp change of slope in the Type I distribution was explained in terms of a loss of the larger nuclei by sedimentation. In strong winds, the part of the curve to the right of the discontinuity probably represents a state of equilibrium between production and loss by sedimentation. In light winds and stable conditions the slope should be steeper due to the fact that the loss by sedimentation is greater than production and larger nuclei are not easily transported vertically under stable conditions. The Type II distributions were only observed when the wind speeds were less than 7 m/sec and resembled a high concentration continental aerosol. In winds of up to 15 m/sec the measured concentrations of large sea-salt nuclei rarely exceeded 10 cm⁻³.

The effect of stability on the concentration of atmospheric condensation nuclei was well documented by Moore (1952). He used an Aitken counter to measure the relationship between concentration of nuclei and the intensity of vertical mixing over the North Atlantic. The results indicated a decrease by as much as a factor of 4 in the number of Aitken particles near the surface on days with cumulus clouds as compared to days with stratus clouds. This would indicate that convection plays an important role, at least in the transport of smaller particles.



Chemical analyses by various investigators have indicated that between 0.1 μ and 1.0 μ radius the marine aerosol is composed of a background component of continental origin and a sea-salt component. Sodium chloride was found to predominate above 1 µ radius while particles of continental origin predominate below 0.1μ radius. Results of a cruise off the Grand Banks in the North Atlantic (Ruskin, et al., 1976) indicated that the continental particles are composed of sulfate compounds and a smaller amount of sulfuric acid. Aerosols over remote ocean areas (Meszaros and Vissy, 1974) were shown to be comprised of variable concentrations of ammonium sulfate, sulfuric acid, sodium chloride, and particles similar in structure to ammonium sulfate. The sum of these four types of identified particles accounted for 75-95 percent of all particles greater than .3 µ radius. In other words, practically all the particles in a pure marine atmosphere, undisturbed by continental particle sources, are soluble in water.

C. RELATIVE HUMIDITY EFFECTS

A solid particle which is composed wholly, or in part, of a pure water-soluble substance will undergo a sudden transition to a saturated solution droplet when some critical value of relative humidity, less than 100%, is reached. The relative humidity at which this transition occurs depends on the size and chemical composition of the particle. The smaller the particle, the lower the critical humidity. Below the transition point, solid particles acquire small amounts of water by the process of adsorption. At relative humidities above the transition point, a particle (or, more properly, an aqueous solution droplet) grows by the absorption of water vapor (Fitzgerald, 1975).

A pure water droplet is said to be in equilibrium with its surroundings if it neither evaporates nor grows. This only occurs when the equilibrium vapor pressure over the surface of the droplet is equal to the vapor pressure of the surrounding air. Winkler (1973) describes the equilibrium growth of aerosol particles due to humidity as complex and depending on the relative proportion of soluble and insoluble material in the particles and on the chemical composition of the soluble component. Complex ionic mixtures, similar to those present in atmospheric aerosols, show material influences and lower the water vapor pressure to a much less degree than the same amount of pure salts. In such complex mixtures the various salts become dissolved only gradually with increasing relative humidity until at a sufficiently high humidity all soluble material is in solution.

Measurements have shown that with increasing humidity a sodium chloride crystal undergoes a phase transition to a saturated solution droplet at a relative humidity of approximately 78%. Figure 6 describes how the equilibrium radii of droplets containing specified masses of sodium chloride vary with the relative humidity. The equilibrium radius of the droplet increases with increasing humidity until the air becomes supersaturated by a critical amount, corresponding to



Figure 6.

• Equilibrium Relative Humidity and Corresponding Radii (Mason, 1975)

the maximum of the curve in this figure. If this supersaturation were maintained, theoretically the droplet will grow without bound. With decreasing humidity a sodium chloride solution droplet crystallizes at a humidity between 35-45%. Since the relative humidity at a height of about 15 meters over the ocean surface goes below 40% very infrequently, seasalt droplets will have little opportunity to crystallize (Fitzgerald and Ruskin, 1977).

Since the later discussion refers to the distribution of sea-salt particles by bolt mass weight of salt (grams), radius of dry crystals (μ) or radius at ambient humidity (μ), the scale in figure 7 is furnished as a reference.

D. THE PRODUCTION OF AIRBORNE SEA-SALT

Although the spectrum of the marine aerosol above .1 μ radius is known to consist of sea-salt particles, very little is certain about the concentration and mechanisms of production. Because of the smallness of the particles and limitations of the sampling equipment, earlier experiments did not measure the quantity of sea-salt particles much less than 10^{-12} gm.

Woodcock (1953) determined that the mass distribution of "giant" (> 10^{-12} gm) sea-salt nuclei varies with wind speed. Increases in the amount of air-borne salt near cloud bases were shown to be related to increases in wind speed at the sea surface, with the greatest proportionate increase in particle number occurring at the large end of the weight range. The results of Woodcock's measurements for wind forces of 1,

3, 5, and 7 on the Beaufort scale are shown in Figure 7. The line (a) gives the size distribution of continental aerosol for comparison. The line (b) is an extrapolated size distribution of the marine aerosol. Chemical analysis of Woodcock's bulk aerosol samples between $.1 \mu$ and 1μ indicated a maximum of sea-salt around 0.3μ and a lower limit in the vicinity of $.1 \mu$ radius. These distributions indicate total concentrations of all sea-salt particles of no higher than a few per cubic centimeter (Junge, 1972). According to Mason (1975), over a rough sea the concentration of sea-salt particles greater than 2 μ radius rarely exceeds 1 cm⁻³ and the total concentration of all salt particles rarely exceeds 10 cm⁻³.

Moore (1952) observed a distinct correlation between wind speed and concentration of sea-salt larger than 10^{-11} gm up to wind speeds of 15 m/sec. He also found a linear increase in concentration of particles larger than 10^{-9} gm with increase in wave height. Results of experiments by Monahan (1968) reveal an abrupt increase in concentration of sea water droplets larger than 45 μ radius at a wind speed of approximately 9 m/sec, measured 47 cm above the sea surface.

Moore (1952) also analyzed the visibility observations at two ocean weather ships and determined that the opacity for a given humidity increases with wind speed. He attributed this increase to an observed increase in the concentration of large nuclei. Another result indicated that at lower humidities, the increase in opacity was more pronounced, and Moore believed this was due to the dehydration of larger droplets. These conclusions would indicate that the aerosol



Figure 7. Size Scale and Average Size Distribution of Sea-Salt Nuclei Measured by A.H. Woodcock (Mason, 1975)

distribution is more variable and sensitive to wind speed in drier air, and this feature should be most noticeable in the larger size ranges.

As the wind speed increases over the ocean, gravity waves are generated and begin to break at a critical wind speed generally agreed upon to be near 7 m/sec. Air that is entrained by these breaking "whitecaps" rises to the surface sometime later in the form of bubbles. The principal mechanisms of sea-salt production are thought to be the direct spraying of droplets off the crests of breaking waves and the bursting of bubbles in areas of whitecaps and foam. Droplets produced by direct spraying are generally larger than 45 µ and, due to large fall velocities, are not airborne long enough to evaporate and become light enough to be transported upward (Monahan, 1968). Toba's model (1965b) showed that the net production of sea-salt particles at the sea surface seems to increase with particle mass even beyond 10^{-8} gm (20 μ), but that the transport by eddy diffusion is not sufficient to carry the particles upward against gravity beyond this size. The presence of particles larger than 10⁻⁸ gm in the atmosphere is generally attributed to coalescence of sea-salt droplets within and below clouds.

Some examples of residence times for different sea-salt particle sizes taken from Junge (1972) are found in Table I. It would seem then that particles in the .1 μ - 20 μ range, at least, are produced by the bursting of bubbles.

In efforts to photograph the rupture of the surface bubble film, Kientzler, et al. (1954) found that their camera exposure was too long to capture this rapid phenomenon. However, they were able to see the formation of the "Rayleigh" jet which projects upward, continues to rise as a thin column, and then breaks into droplets of varying sizes. Day (1964) describes this process in the following manner. Each bubble, as it reaches the surface, develops a spherical film-cap which drains, thins, and bursts. Fragments of the film are thrown out and are dragged upward by the air which escapes from the bubble orifice. Water, rushing down the sides of the bubble cavity, emerges from the center as a narrow jet. A schematic of this process is shown in Figure 8. The larger drops (L) are formed by disintegration of the jet (J). Smaller particles (S) are formed by bursting of the bubble film.

Kientzler's experiment was significant in that no droplets of large enough size to be resolved by the film and optical system were observed from .2 - 1.8 mm diameter bubbles until after the jet formation. This was interpreted to indicate that the larger droplets are not produced when the bubble film is broken. On the average, the droplets produced by the jet mechanism were approximately 1/10 of the original bubble size. 1 mm diameter bubbles were observed to produce droplets of approximately 50 μ radius. The smallest observed were of 2 μ radius and deduced to have been formed by a bubble of approximately .04 mm diameter. Therefore, the jet mechanism can be considered a source of salt particles greater than 10^{-12} gm (1 μ).

	Residence Time 7, days					
	M 10" grams	M = 10 ^{-#} grams	M - 10-10 grams	M = 10 ⁻⁺ grams		
Toba's value w: for 80% relative humidity	86 .	. 17	2.9	0.32		
Toba's value w, for 91.4% relative humidity	50 ·	11	9.1	0.23		
Brikmon's estimate from	82	16	2.6	0.4		
Brikmon's estimate from production	3.5	1.0	0.6	0.5		
Our estimate*	1.9	1.6	1.0	0.26		

Table is taken from Tobs (1965c, Table 2). The variable M is the mass of particles.

Table I.

Residence Times of Sea-Salt Particles over the Oceans



Figure 8.

The Formation of Sea-Salt Droplets by the Bursting of Bubbles (Mason, 1975)

Mason (1954) utilized a cloud chamber to study bubble behavior in both distilled and salt water. After expansion, a dense cloud of tiny droplets was observed rising vertically in the space above the salt water, but not above the distilled water. Bubbles of 3 mm diameter produced 100-200 of these condensation nuclei, the majority of which are estimated to have salt contents between 10^{-15} gm and 2 x 10^{-14} gm. This would correspond to droplets of approximately .1 μ to .3 μ radius at 80% relative humidity. Mason also observed a second group of droplets produced by the shattering of the bubble film. These were projected sideways at an angle of ten to 15 degrees above the horizontal and slightly larger, containing between 2 x 10^{-12} - 5 x 10^{-10} gms of salt. However, the numbers of these droplets were always small, on the average, there was only about one droplet in this size range.

The number of droplets which rise vertically from a bursting bubble is strongly related to the state of compression of the film of organic material on the water surface. Paterson and Spillane (1969) have shown that with an increase of film pressure the number of nuclei produced decreases markedly. This would indicate that the production of seasalt droplets originating from the bubble film mechanism would be suppressed in regions of high organic activity on the sea surface. Aerosol samples taken by Woodcock (1972) over Hawaiian and Alaskan seas may help explain where the transition between the jet and film sea-salt production mechanisms occurs. His observations, using an improved slide collection
technique, show an increased average particle production for sea salt particles less than 2 x 10^{-14} gm (.3 μ radius) in Hawaii where marine organic productivity is low. In contrast, the mean distribution curve for particles over the organically rich Gulf of Alaska fails to indicate an increased slope of the concentration curve among particles of the same size range. These curves are shown in Figure 9. The presence of surface active films arising from the biologically productive Alaskan waters is thought to suppress the production of film droplets.

Statistical analysis by Meszaros and Vissy (1974) showed that with increasing particle radius the correlation between wind speed and chloride concentration increased. This meant that smaller chloride particles are formed by the bubble film mechanism than by direct spraying. The distribution curve gives evidence that the transition between these two chloride formation mechanisms lies between $.2 \mu$ and $.4 \mu$. Thus the maximum at $.1 \mu$ gives the maximum of chloride particles formed by the bubble film process.

Moore (1952) found evidence that the particle concentrations below 1 μ are not correlated with wind speed. This would indicate that most of the particles between .1 μ and 1 μ are not produced by bursting bubbles. Other experiments using the effects of relative humidity on particle growth indicate that a considerable proportion of marine particles between .1 μ and 1 μ must differ in composition from seasalt (Junge, 1972). Meszaros and Vissy (1974) found that, in this size range, sodium chloride varied from 4-50% of the



Figure 9. Distribution Curves over Alaskan and Hawaiian Waters (Woodcock, 1972)

concentration for all particles. The observations by Hidy <u>et al</u>. (1974) off the coast of Southern California revealed that 11% of the aerosol sampled contained sea-salt, the remainder being a combination of sulfates, nitrates and soil dust.

E. AEROSOL MODEL

Recently, various aerosol models have been developed in an attempt to accurately describe marine aerosol distributions as a function of one or more parameters. This is essential for the calculation of optical propagation through the atmosphere as aerosols scatter and absorb energy. Since the aerosol distribution is known to be dependent on relative humidity and wind speed, these two variables usually are the key parameters of each model.

One model in particular has been developed by Fitzgerald and Ruskin (1977) on the basis of the North Atlantic observations. They applied the effects of relative humidity on the equilibrium growth of aerosol particles to the sea-salt mass distribution determined by Lovett (1975) in the North Atlantic. Lovett presents empirical log radius mass distributions in the form of the following power law:

$$\frac{dN}{d \log r_d} = C r_d^{-\nu}$$
(3)

where r_d is the dry particle radius and C and v depend on the wind speed V in the following manner:

v = 3.317 - .03 V (4)

$$C = 0.2 - 0.0196 V + 0.0121 V^2$$
 (5)

These expressions are valid only over a wind speed range of $3-17 \text{ m/sec}^{-1}$.

Formulae have been derived (Fitzgerald, 1975) for the equilibrium size of aerosol particles composed of a single pure salt as a function of relative humidity. For a sodium chloride particle the relationship between particle radius and relative humidity may be expressed as

$$r = \alpha r_d^{\beta}$$
 (6)

where α and β are functions of the relative humidity as described by Fitzgerald (1975). Equations (3) and (6) are combined to describe the aerosol size distribution as a function of relative humidity and wind speed, giving

$$\frac{dN}{d \log r} = \frac{c}{\beta} \left(\alpha^{\nu/\beta} \right) \left(r^{-\nu/\beta} \right) . \tag{7}$$

Comparison between the aerosol distributions derived from the above model and those observed in two coastal marine environments is made within this study.

and

III. TURBULENCE THEORY

A. BOUNDARY LAYER CONSIDERATIONS

The importance of turbulent exchange processes in the surface boundary layer has long been recognized. Panofsky (1969) describes atmospheric turbulence as consisting of horizontal and vertical eddies by which the air is mixed. The two mechanisms by which eddies are formed in the atmosphere are heating from below and wind shear. Heating produces convection and the change in wind speed with height produces mechanical turbulence. Because there is no wind at ground level, and there is usually some wind above the ground, mechanical turbulence is common. This type of turbulence increases with increasing wind speed (at a given height) and is greater over rough terrain than over smooth terrain. The terrain roughness is usually characterized by a roughness length, Z, , which is proportional to the size of the eddies that can exist. The relative importance of heat convection and mechanical turbulence is characterized by the Richardson number, R_i . The Richardson number is a measure of the relative rate of conversion of convective to mechanical energy. For example, negative Richardson numbers of large magnitude indicate that convection predominates resulting in strong vertical motion. As the mechanical turbulence increases, the Richardson number approaches zero.

Finally, as the Richardson number becomes positive, the thermal stratification becomes stable and damps the mechanical turbulence. For $R_i > 0.25$, vertical mixing disappears.

The effect of the wind on the underlying surface is termed the shearing or Reynolds stress, τ , and is characterized by a downward momentum transfer. The Reynolds stress may be represented by

$$\tau = -\rho \langle u'w' \rangle \tag{8}$$

where u' = fluctuating horizontal wind velocity

w' = fluctuating vertical wind velocityρ = density of air

It is convenient to express Reynolds stress in terms of the friction velocity U_* so that

$$\tau = \rho U_{*}^{2}$$
(9)

where U_* is constant throughout a region of constant momentum flux. Hence, U_* is a measure of the downward transfer of momentum in the lower 50 meters of the atmosphere. Over the ocean an increase in the near surface winds would lead to a greater momentum and energy transfer for surface wave and sea-salt aerosol production. The relationship between the turbulent transfer of heat and moisture in the marine boundary layer and the generation and transfer of aerosols is not well known and, unfortunately, is not investigated in this study.

B. MOMENTUM TRANSFER, U, , RELATIONS

A thorough discussion of the boundary layer expressions is presented in several references, e.g. Lumley and Panofsky (1964). The similarity approach of Monin and Obukhov (1954) is used to define a representative length scale, L , for the surface layer of the atmosphere,

$$L = \frac{-U_{\star}^{3} T_{o}}{kg \overline{w'T'}}$$
(10)

where

g = gravitational acceleration

T = ambient temperature

k = von Karman constant = 0.35

The selection of the Monin-Obukhov length as a stability scaling parameter is based on the assumption that friction velocity, U_{\star} , and vertical heat flux ($\overline{w^{*}T^{*}}$) are constant in the surface layer. This scaling length, using dimensional analysis, leads to the development of a dimensionless function, $\phi_{\rm m}(Z/L)$, which can be used to represent the mean horizontal wind variation with height, $d\bar{u}/dZ$, in the surface layer. The following expression is the empirical relationship for the wind shear in this development,

$$\frac{d\bar{u}}{dZ} = \frac{U_{*}}{kZ} \phi_{m}(Z/L)$$
(11)

As vertical turbulent heat flux $(\overline{w^{TT}})$ decreases to zero, indicating neutral stability, $\phi_m(Z/L)$ must approach 1 if Equation (11) is to take on its expected form under neutral conditions. Assuming that convective mixing is negligible under neutral conditions it follows that for values of $\phi_{\rm m}$ (Z/L) near 1 or Z << 1 mechanical turbulence is of primary importance. Thus, the absolute magnitude of <u>L</u> becomes an indicator of the vertical extent to which mechanical turbulence controls the turbulent regime.

Observational experiments by Businger <u>et al</u>. (1971) produced a definite relationship between the Richardson number, R_i ,

$$R_{i} = \frac{g(\partial \theta_{v}/\partial Z)}{\overline{\theta}(\partial u/\partial Z)^{2}}$$
(12)

and the Monin-Obukhov length, L , where θ_v is the virtual temperature. The following expressions are approximations for the unstable and stable conditions respectively,

$$Z/L = R_{i}$$
(13)

$$Z/L = \frac{R_i}{1-\alpha R_i}$$
(14)

where α is an empirically derived constant equal to 0.5.

Of interest in this study is the rate of viscous molecular turbulent kinetic energy dissipation, ε . Wyngaard, <u>et al</u>. (1971) considered the dependence of ε on momentum fluxes and height in deriving the following empirical expression

$$\varepsilon = U_{\star}^{3}/kZ \phi_{\varepsilon} (Z/L)$$
 (15)

Since Z/L and R_i are functionally related, equations (11) and (15) can be rewritten as

$$\frac{d\bar{u}}{dZ} = \frac{U_{\star}}{kZ} f_{m} (R_{i})$$
(16)

$$\varepsilon = U_*^{3} / kZ f_{\varepsilon} (R_i)$$
(17)

where f_m and f_{ϵ} are stability corrections equal to 1 under neutral conditions. In near neutral conditions, the turbulent kinetic energy production is assumed to be equal to the rate of molecular dissipation of turbulent kinetic energy and from equations (16) and (17) the following relation is valid

$$\varepsilon = U_{*}^{2}(\partial \bar{u}/\partial Z)$$
(18)

Assuming neutral conditions, the combinations of equations (16) and (18) yields

$$U_{a} = (\varepsilon kZ)^{1/3}$$
(19)

Now the friction velocity U_{\star} can be estimated from either mean wind profiles using the integrated form of equation (16) or from velocity fluctuation data involving turbulent energy dissipation by using equation (19). The latter approach is used in this study.

and

IV. DATA COLLECTION

A. DURATION AND LOCATION

Aerosol and meteorological data for this study were made available through Calspan Corporation, Buffalo, New York, from two separate experiments. During a ten day period in February 1977, Calspan Corporation provided limited meteorological and cloud physics support during a study of marine boundary layer phenomena conducted on the Gulf of Mexico (Mack and Katz, 1977). The experiment was performed on the Naval Coastal Systems Laboratory's (NCSL) offshore platform "Stage I" located approximately 20 km SW of Panama City, Florida as depicted in Figure 10.

A second experiment which provided data for this study was conducted along the coastal waters of Southern California (Figure 11) during a 12 day period in July 1977 aboard the Naval Postgraduate School (NPS) R/V Acania. Under contract from NPS, Calspan Corporation provided limited meteorological and aerosol physics support during a study of air quality parameters and marine boundary layer characteristics (Mack, 1977). This region contains primary shipping lanes and a number of drilling platforms all of which contribute to atmospheric contamination.

The following discussion will be limited to equipment used to measure the meteorological parameters actually analyzed in this study. A listing of the Panama City and





Southern California data may be found in Tables V and VI, respectively, at the end of the text.

B. PANAMA CITY INSTRUMENTATION

"Stage I" provided a stable platform for measuring the meteorological parameters necessary to describe and study the aerosol distribution and behavior in the marine boundary layer. The instrumentation installed by Calspan included a Sling Psychrometer, Bechman-Whitley wind system, Gardner small particle detector, and Royco Model 225 Particle Counter. The wind speed and direction was monitored continuously at the 20 meter level while wet and dry bulb temperatures were obtained hourly at the 17 meter level. A Foxboro temperature system (4 sensors) provided continuous temperature measurements at 4 levels; sea surface, 4.5, 9.0 and 24.5 meters. This data was recorded in an hourly log. Ten minute averaged aerosol size spectra were obtained continuously with the Royce counter at the 17 meter level, and a printout of aerosol concentration in 5 size intervals was provided every ten minutes. The Gardner Counter measured the concentration of particles greater than .0025 μ diameter on an hourly basis.

The majority of the time the Royco instrument operated in "threshold" mode where number concentration (per 2.8 liters) of particles greater than the following size ranges were measured: 0.5 μ m, 0.7 μ m, 1.4 μ m, 3.0 μ m, and 5.0 μ m diameter. For a shorter period of time the instrument was operated in the "window" mode producing number concentrations between the

above size ranges. The particle counter and sensor are shown in Figure 12. The environmental air was drawn continuously through a sampling line of 3 meter length and 5 cm inside diameter. The flow rate through the counter's sensing volume was set at 2.8 liters per minute.

The Royco Model 225 sampler utilizes a near forward scattering optical system (Figure 13) which is ideal for monitoring large volumes of ambient gases where suspended particles can vary widely in composition, size, and optical properties. The aerosol is drawn through the sensor into a beam of focused light. As each particle passes through the illuminated volume, it scatters a pulse of light which is then detected by a photomultiplier tube. The photomultiplier output is then processed elctronically to produce a pulse height spectrum from which the particle size spectrum is deduced. The height of each pulse is proportional to the square of the diameter of the particle.

Whitby and Liu (1973) note that the important characteristics of an optical counter are the sampling flow rate and the size of the optical viewing volume. The sampling flow rate determines the minimum counting period needed to obtain a statistically accurate count, and the size of the optical viewing volume determines the maximum aerosol concentration the instrument can accept without loss of particle count due to "coincidence", i.e., the loss of particle count due to the presence of more than one particle in the optical viewing volume at the same time. The viewing volume of the Royco 225 is 4.0 mm³ and the collection aperture half angle is 25 degrees.





This model is also equipped with a sheath air inlet which diverts part of the aerosol stream through an external filter before reentry to the viewing volume. This sheath improves the performance of the instrument by preventing the recirculation of particles in the optical chamber and by confining the aerosol stream to a narrower region. Thus, the broadening of the pulse spectrum due to variation in illuminating intensity is reduced.

C. SOUTHERN CALIFORNIA INSTRUMENTATION

The location of the sensors aboard the R/V Acania are shown in Figure 14. Again, a Royco Model 225 Optical Particle Counter was used to measure the aerosol concentration of the coastal marine boundary layer. This instrument was operated continuously in the threshold mode where number concentration (per .28 liters) of aerosols greater than the following size ranges were measured: $0.3 \ \mu\text{m}$, $0.6 \ \mu\text{m}$, $1.2 \ \mu\text{m}$, $3.0 \ \mu\text{m}$, and $5.0 \ \mu\text{m}$ diameter. The mainframe and sensor were located near the bridge of the Acania with the origin of the sampling line positioned forward of the pilot house roof at a height of 7 meters above the sea surface. The sampling line was 6 meters long with an inside diameter of 5 cm. The air was sampled through the viewing volume at a rate of .28 liters per minute. A Gardner small particle detector was again used to measure the Aitken nuclei concentration.

A sling psychrometer was used to measure the wet/dry bulb temperatures and relative humidity determined from psychometric tables for a height of 5 meters. The mean wind





measurements were obtained at four levels using cup anemometer wind profile register systems supplied by the NPS. Calspan recorded the wind, humidity, and aerosol measurements in an hourly log.

Velocity fluctuation measurements were obtained with Thermo-Systems Model 1210 hot wire anemometer probes mounted with hot film sensors (platinum coated, 60 mil quartz fibers) installed by the NPS. The anemometer was a Thermo-Systems Model 1054B. The sensors were small enough to resolve the viscous dissipation scale without making corrections for wire length. Wind fluctuation data were recorded on a 14 channel tape recorder. The placement of these sensors required exceptionally long cable runs. Therefore, adjustments were made in the bridges for resistance and capacitance of the wirelength to insure a correct response.

The mean and fluctuation wind data were logged by the NPS developed MIDAS (Microprogrammable Integrated Data Acquisition System). This system is fully automated to sample the tailored list of sensors every 30 seconds and 20 minute averaged output values were printed.

V. ANALYSES PROCEDURES

A. VELOCITY FLUCTUATION ANALYSIS

The dissipation of turbulent kinetic energy, ε , can be related to the mean wind velocity at any given level, \bar{u} , and the RMS value of the velocity fluctuation, $\bar{u'}^2$, in a frequency band specified by a lower frequency limit, f_{ℓ} , and an upper frequency limit, f_{u} (Fairall, <u>et al.</u>, 1977). The relationship is

$$\varepsilon = \frac{(4/3)^{3/2} (u_{RMS}^*)^3}{(\bar{u}/2\pi)[f_{\ell}^{-2/3} - f_{u}^{-2/3}]^{3/2}}$$
(20)

In this procedure recordings were made of both the cup anemometer wind speed and the corresponding hot wire voltage output. The sensor wind speed is given by

$$v = V_0^2 + B(\bar{u})^{\frac{1}{2}}$$
 (21)

where v is the hot wire voltage output, and V_0^2 and B are constants obtained by laboratory calibration using a TSI Model 1125 Calibrator. Differentiation of equation (21) produces the following relationship between the velocity fluctuation and the voltage fluctuation:

$$u'_{\rm RMS} = \frac{4v(\bar{u})^{\frac{1}{2}}}{B} v'_{\rm RMS}$$
 (22)

Substitution into equation (20) yields

$$= \frac{(4/3)^{3/2} [4v(\bar{u})^{\frac{1}{2}})/B]^{3} (v_{RMS}^{*})^{3}}{(\bar{u}/2\pi) [f_{g}^{-2/3} - f_{u}^{-2/3}]^{3/2}}$$
(23)

Values of $f_{l} = 5$ Hz and $f_{u} = 200$ Hz were selected for the cruise and since amplifiers with known gains, G, were required, further reduction leads to

$$\varepsilon = (3.53 \times 10^3) [V_0^2 + B(\bar{u})^{\frac{1}{2}}]^{3/2} (\bar{u})^{\frac{1}{2}} [v_{RMS}^*/BG]^3$$
 (24)

The friction velocity, U_{\star} , was then calculated from equation (19) for each of three levels and averaged to produce over 400 values from 19-27 July. Voltage fluctuation data from level 3 proved to be erroneous and were not included in the calculations. Obviously erroneous values of U_{\star} owing to erratic behavior were also neglected. U_{\star} values were then averaged about the aerosol observation times to correspond to a given aerosol distribution.

B. AEROSOL ANALYSIS

Analyses were performed on 215 aerosol samples during the SC cruise which were confined to the time period of the valid velocity fluctuation measurements. The observations included date and time, humidity, relative wind speed and direction, ship's speed and heading, Aitken concentration, and aerosol concentration as determined by the Royco 225 optical counter (Table VI). Wind and ship's speeds were recorded in knots. The analyzed aerosol observations for the PC experiment were limited to 137 cases during the period 18-23 February. Cold frontal passage at approximately 00002, 24 February and subsequent advection of continental dust through 25 February were reasons for neglecting the aerosol samples for these days. Aerosol counts prior to 18 February were determined with the Royco instrument in the window mode and were not included in this study. Observations were generally made hourly and recorded in a log. They included date and time, humidity, wind speed and direction (knots), Aitken concentration, and data from the optical particle counter (Table V).

Computer programs were developed to plot the aerosol size distribution as a function of radius (R) in microns versus $dN/d \log R (cm^{-3})$ where N is the number of particles greater than a given radius as measured by the Royco instrument. The program also included provisions to plot size distributions predicted by Fitzgerald's model. For this the observed relative humidity and wind speeds were used with equation (7). Initially the average aerosol distributions for both the SC and PC experiments were computed and compared to the respective predicted model distributions.

Subsequently, the variations in the average aerosol distributions with respect to four different categories of wind speed, relative humidity, and friction velocity were plotted for the SC data. The categories chosen for each of the above respective parameters are as follows: 0-2, 2-5, 5-8, 8-12 m/sec; 90-99, 80-90, 70-80, and 60-70 percent; and 0-.15, .15-.25, .25-.35, .35-.70 m/sec. Friction velocity data was not available from the PC experiment; therefore, variations in the aerosol distributions were plotted with respect to categories of wind speed and humidity only. Because of essentially different meteorological conditions, the categories were chosen as follows: 0-3, 3-7, 7-10, and 10-15 m/sec; and 85-99, 70-85, 55-70 and 40-55 percent.

Visual inspection of these plots may indicate satisfactory relationships between the aerosol concentration and the above parameters. However, a statistical means of viewing these relationships was also deemed necessary. Wind speed, humidity, and U_{\star} values were cross correlated with number concentration of particles in graduated size ranges. This procedure was accomplished by a Biomed Regression/Correlation computer program which produced corresponding correlation coefficients.

The nature of the diurnal variation of the aerosol concentration during the SC and PC experiments was investigated in this study. A computer program averaged the aerosol concentrations, wind speeds, humidities, and friction velocities about each hour and plots showing variations with time are produced. The aerosol plots depict the number of particles per cm³ within specified size ranges versus time. The SC data produced curves representing the number of particles between the following size ranges: .15-.30 μ , .30-.60 μ , .60-1.5 μ , and 1.5-2.5 μ radius. Diurnal variation of concentration for the PC data utilized the following slightly different size ranges: .25-.35 μ , .35-.70 μ , .70-1.5 μ , and 1.5-2.5 μ radius.

Finally, diurnal variations of the aerosol size distribution for the SC and PC experiments were calculated using techniques similar to those described above. Average size distributions for the following two time periods were plotted: 0000-1200 hrs and 1200-2400 hrs.

C. ERROR ANALYSIS

The optical particle counter has an advantage over the membrane filter or impactor sampling techniques. For example, the latter require the samples to be taken to a lab for microscopic inspections and the aerosols may possibly be disturbed or altered due to contamination. Although the optical counter provides continuous "in situ" aerosol measurements, there are ample causes for counting errors. Because light scattering is a function of size, shape, and refractive index of the particles, careful calibration is necessary.

The Royco 225 model counter used in these experiments was calibrated using monodisperse latex spheres of known refractive index (1.6). Laboratory experiments by Lieberman and Allen (1969) showed a good correlation between the theoretical response curve for a near forward optical system and measurements using latex sphere and glass beads of refractive index 1.6 (Figure 15). Of most significance is the "fold" in the curve or zone of multi-valued response in the region of 1 μ diameter. Figure 16 is provided to illustrate how the response curve varies with particles of different refractive index. It is evident that when measuring particles of refractive index 1.6, a zone of ambiguity exists between



Figure 15.

Theoretical Response Curve and Experimental Results for a Forward Scattering System



Figure 16.



approximately .3 μ and .6 μ radius and may vary with the aerosol refractive index. Lieberman and Allen (1969) state that the instrument will still produce valid data if the zone is encompassed within a size range or channel. Since the SC counter measured between .3 μ and .6 μ radius and the PC counter between .35 μ and .70 μ radius, it is assumed that this multi-valued zone is compensated for.

Counting errors can also arise from flow rate considerations. If the particle sizes are large and the number of particles small, enough particles must be counted to obtain good statistical resolution. When a small random number of particles is counted, the statistical error in counting is equal to the ratio of 1 over the square root of the number of particles counted (Zinky, 1962). The counter should be operated over a longer time period (10 minutes) to sample a larger volume or an increase in the flow rate will reduce the error. It then seems quite possible that the flow rate of the counter used in the SC cruise (.28 liter/min) provided too small of a sampling volume to obtain an accurate count of the larger particles.

Zinky (1962) also states that a vertically aligned inlet tube is recommended to prevent any deposition in the line due to settling. It has already been mentioned that the sampling lines used in each experiment were considerably long and aligned horizontally. Many of the larger particles may not have remained airborne long enough to reach the illuminated volume.

Errors in the calculation of the friction velocity may have come from various sources. Since calculation of U₄ from Equation (19) is only valid for near-neutral conditions, any substantial departure of the Richardson number from zero would result in inaccurate values. The measurement of the dissipation of turbulent kinetic energy was large dependent on the accuracy of the voltage output. The signal response is sensitive to electromagnetic energy, and any local radio or radar transmission may introduce noise to the system. Additionally, under the light wind conditions which prevailed on the SC experiment, the lateral motion of the anemometers due to ship pitch and roll may have resulted in erroneously high readings.

VI. RESULTS

The data from the Southern California (SC) cruise proved to represent an atmosphere somewhat different from a typical marine environment. The Aitken particle population averaged almost 8500 cm⁻³ which is about 4 times higher than that observed by Hidy, <u>et al</u>. 130 km west southwest of Los Angeles. This high concentration is suspected to be due to a combination of pollution from merchant ships' exhaust, combustion from the drilling platforms, and offshore flow from the nearby populated coastal cities.

The average wind speed and relative humidity were 3 m/sec and 86 percent, respectively. This data was used to compute the prediction from Fitzgerald's model (Eq. 7) which is compared to the average SC distribution in Figure 17. The vertical bars represent one standard deviation either side of the mean. There is generally good agreement between the two below .4 μ radius, with a larger experimentally observed concentration above this range. Although sea-salt production should have been minimal during this time period because of low wind speeds, the characteristic hump at around 1 μ , to a certain extent, reflects the contribution by sea salt nuclei. A similarity exists here with Moore and Mason's (1954) observation of a discontinuity where the slope changes and becomes rather steep in the region of the larger size range. The larger concentration in this range may be solely



Figure 17.

Average Aerosol Size Distribution for the SC Experiment and Distribution Predicted by Fitzgerald's Model due to the influence of atmospheric contaminants such as combustion by-products, soil dust, or smoke. Considering previous experiments, this range does indeed contain a mixture of both continental and marine aerosols possibly resulting in the increase over Fitzgerald's model.

As previously mentioned, the low flow rate of the optical counter may account for the low concentrations at 2 μ . However, since the wind speed reached 8 m sec⁻¹ only 6 times, this may have been a truly representative concentration of droplets as agreement is also shown with Fitzgerald's curve.

Figure 18 presents the synoptic situation for three days at the beginning, middle, and end of the experiment. A persistent thermal low is located in the desert area of Southern California and the isobaric pattern off the coast reflects a rather weak gradient. Therefore, smaller scale circulations should prevail in this area of little or no synoptic forcing.

Plots showing the variations of the average distributions with respect to wind speed, relative humidity, and friction velocity (U_{\star}) , are shown separately in Figures 19, 20, 21. The number of observations in each category is placed in parentheses. These figures indicate that the size distribution has a better relationship with the relative humidity than to the wind speed and U_{\star} . Correlation coefficients between these parameters and the number concentration of particles in a given size interval are produced in Table II. Since diurnal variations tend to reflect a negative relationship between relative humidity and wind



19 JUL

23 JUL



26 JUL

Figure 18. Synoptic Situation during the SC Experiment

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Figure 19. Varia

Variation of SC Size Distribution with Wind Speed



Figure 20. Variation of SC Size Distribution with Relative Humidity





SC

INTERVAL	RH	WIND SPEED	n*
.15 – .30 µ	.375	127	162
.3060 µ	.383	056	107
.60- 1.5 µ	.376	017	077
1.5 - 2.5 μ	037	022	.132

TOTAL

Correlation Coefficients for the SC Experiment Table II.
speed, the results in this table are not surprising. The negative correlation of relative humidity with the concentrations in the large size range indicates that sedimentation of large droplets, which grow with increasing humidity, is most important when the wind speed and sea surface production of salt nuclei are weak. Although these larger droplets also exhibit a small positive correlation with U_{\star} while the wind speed correlation remains negative, this result does not appear to be significant.

An attempt was made to examine the influence of stability on the size distribution. The summer months are characterized by the occurrence of stratus and fog off-shore below the marine inversion. Two days are compared with the assumption that they represent the unstable and stable atmospheres. According to the daily observation log, stratus clouds in the morning becoming stratocumulus by afternoon were observed on 19 July. 26 July was characterized by clear skies. The average distribution for both days is presented in Figure 22. The correlation coefficients between concentration and wind speed and U, show a trend toward positive values from the stable to the unstable day with U* eventually becoming positively correlated in the unstable day (Table III). The increase in the size distribution on 26 July in the size range greater than $.3 \mu$ seems to be due to increase in the average wind speed and occasional gustiness as whitecaps were reported during the afternoon. The stable stratification assumed in this case allows generated sea-salt nuclei to accumulate and the concentration to increase at the 7 meter





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	-			-

INTERVAL	RH	WIND SPEED	U*
.1530 µ	.814	205	,129
.3060 µ	.837	184	.165
.60- 1.5 µ	.792	110	.147
1.5 - 2.5 µ	104	049	.075

19 JUL UNSTABLE

-	-
3	C

INTERVAL	RH	WIND SPEED	U+
.1530 µ	.610	395	372
.3060 μ	.754	499	535
.60- 1.5 µ	.835	615	634
1.5 - 2.5 µ	.355	413	273

26 JUL STABLE

Table III.Correlation Coefficients for 19 July and
26 July

level. The lower average wind speed associated with the unstable period does not allow for much sea-salt production. An unstable atmosphere can lead to convective processes which may vertically transport aerosols and create higher concentrations at an upper level as proposed by Ericksson (1959) and Toba (1965a & b). Hence, a decrease in the size distribution on 19 July is observed. This evidence gives credence to the possibility that friction velocity is a better indication of aerosol size distribution than wind speed. On both days the correlation of the concentration with humidity is lowest in the largest size interval. This relationship is most pronounced on the unstable days and may be explained by sedimentation due to mixing and resulting increased coalescence.

The averaged diurnal variations of wind speed, relative humidity, friction velocity, and aerosol concentration are shown in Figures 23, 24, and 25. Again the negative relationship of aerosols to wind speed and U_{\pm} in the size range of generally less than 1 μ is indicated. A satisfactory relationship with relative humidity is not evident and this is probably due to transport by a secondary circulation. A land-sea breeze type of effect could account for the observed decrease in concentration of the particles "smaller than 1.5 μ . As the heating over the land generates an on-shore flow along the coast, the wind increases and persists through the afternoon. The average wind direction derived from the observations of five random days during the experiment is shown in Figure 26. A westerly wind is



Figure 23.

23. Average SC Diurnal Variations of Wind Speed and Relative Humidity



Figure 24. Average SC Diurnal Variation of Friction Velocity







Figure 26. Average SC Wind Direction

seen to dominate during the peak wind periods. The decrease in the aerosol population may be explained by a horizontal divergence effect in the marine boundary layer as largest accelerations are found near the coast. The average size distributions displayed in Figure 27 reflect the decrease in the aerosol population due to this sub-synoptic circulation. Although the relative humidity increases slightly in the early evening hours, the smaller nuclei show a stronger relationship with the wind speed. This again implies that a large part of the coastal marine aerosol is of continental origin. The outflow of circulation aloft is probably responsible for the introduction of continental particulates to the marine environment. The minor peaks in the small particle concentration and also the somewhat greater increase in the large particles during the afternoon should be attributed to sea-salt production.

The Panama City (PC) observations more closely resembled a marine environment. The Aitken particle count was lower and averaged 2600 cm⁻³ while the distribution curve showed a marked change from the Southern California data. Winter time synoptic scale features predominate in this region of the Gulf Coast. Cold frontal passages and an accompanying influx of continental air into the Gulf of Mexico are frequent occurrences. Subsequent movement of the high pressure ridge into Florida and off its eastern seaboard provides the circulation which reestablishes moist southerly flow and return of the marine aerosol. Figure 28 provides the synoptic analyses for the period of the experiment. Stable



Figure 27. Average SC Diurnal Variation of the Aerosol Size Distribution

••••



18 FEB

21 FEB



23 FEB

Figure 28. Synoptic Situation during the PC Experiment

conditions prevailed in the Panama City area at the beginning of the period; but, after frontal passage early on 20 February, south to southeasterly flow developed and persisted for the remainder of the experiment. The influx of this warm, moist air contributed a destabilizing effect in the lower levels of the marine boundary layer.

The average wind speed and relative humidity for PC were 8.4 m/sec and 71 percent, respectively. Fitzgerald's curve for these average conditions and the average aerosol distribution for the entire period are shown in Figure 29. Good agreement exists only for particle size range greater than .9 μ radius. The observed concentrations are approximately an order of magnitude lower than Fitzgerald's prediction for aerosols smaller than .5 μ radius. A significant aspect of the distribution is the positive slope observed between approximately .5 μ and 1 μ radius which appears to be the result of sea-salt production. Actually, good agreement is shown with Blifford's (1970) observation off the Pacific coast with respect to both slope and number concentration.

Plots showing the effect on the average distributions due to wind speed and relative humidity separately are shown in Figures 30 and 31. Relatively good correlations seem to exist between these parameters and the aerosol distributions. Correlation coefficients are presented in Table IV. The synoptic scale effects predominate over diurnal variations and wind speed and relative humidity are both positively correlated to the concentration. The highest correlation of



Figure 29. Average Aerosol Size Distribution for the PC Experiment and Distribution Predicted by Fitzgerald's Model



Figure 30. Variation of PC Size Distribution with Wind Speed



Figure 31. Variation of PC Size Distribution with Relative Humidity

INTERVAL	RH	WIND SPEED
.2535 <i>µ</i>	.620	.559
.3570 μ	.726	.554
.70-1.5μ	.654	.511
1.5 - 2.5 μ	.729	.442

Correlation Coefficients for the PC Experiment Table IV.

TOTAL

PC

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concentration to humidity at Panama City is witnessed in the 1.5 μ to 2.5 μ interval. This result may indicate that equilibrium tends to exist between production and sedimentation in this interval as hypothesized by Moore and Mason (1954). Disagreement exists in that the steeper negative slope is found during periods of strongest wind. The plot showing the effect of relative humidity on the size distributions results in small variations in the .25 μ - .35 μ interval. This probably indicates that the majority of these particles represent a mixture of continental and marine nuclei.

The stability influence was investigated by comparing observations on 18 February and 21 February. Temperature measurements at various levels on the platform made it possible to examine the lapse rates and determine the stability. The average distributions for these days grouped according to wind speed, and respective correlation coefficients are shown in Figures 32 and 33. The low humidities on 21 February resulted from the earlier intrusion of continental air, but southeast to southwest flow persisted most of the day. Although this trajectory helped to advect in warmer air, production of sea-salt dropped off as the wind decreased considerably below 7 m/sec. A much larger decrease is observed in the distribution curve on 21 February as compared to 18 February when the wind speed decreased below 7 m/sec. This agrees well with Moore's (1952) finding that the change in opacity is well marked during periods of low humidity. Also the decrease in the slope of the curve between $.5 \mu$ and



	1000	
	-	
• •		
100	-	

INTERVAL	RH	WIND SPEED
.2535µ	.169	.065
.3570µ	.104	.334
.70- 1.5 µ	.279	.605
1.5 - 2.5 #	.385	.442

18 FEB STABLE

Figure 32. Correlation Coefficients and Variation of the Size Distribution with Wind Speed, 18 February



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INTERVAL	RH	WIND SPEED
.2535µ	.746	.615
.3570µ	.692	.651
.70-1.5µ	.558	.737
1.5 - 2.5 #	.253	.571

21 FEB UNSTABLE

Figure 33. Correlation Coefficients and Variation of the Size Distribution with Wind Speed, 21 February

Tamera 1.2

l μ appears to be a function of decreased relative humidity. This again reinforces the premise that sea-salt nuclei predominate in the size range above approximately .7 μ .

The correlation between concentration and wind speed for all size ranges is greatest on the unstable day. As with the SC data, this is consistent with momentum and diffusion theory. Again the small correlation with relative humidity exhibited by the larger nuclei is probable caused by growth and sedimentation in the absence of significant sea-salt production. When generation was occurring on 18 February, the large particles exhibited the largest correlations with humidity and wind speed. This stable stratification evidently was produced by a previous frontal passage and northerly flow of cold air and accompanying continental particulates. Therefore, a large portion of the aerosol at the beginning of the experiment may have been composed of non-hygroscopic material.

Figures 34 and 35 display the average diurnal changes in wind speed, relative humidity, and aerosol concentration. Again positive correlations are noted as relative humidity and wind speed, although containing quite a bit of scatter, tend to vary accordingly. Of most significance would be the obviously high concentration of droplets in the $.7 \mu - 1.5 \mu$ range. Noting that the average wind seldom went below 7 m/ sec, this would indicate that sea-salt nuclei production is greatest in this size range. A diurnal representation of the average aerosol distribution is presented in Figure 36.











Any transport of aerosols due to a land-sea breeze effect should be ruled out as a satisfactory relationship does not seem to exist.

VII. CONCLUSIONS

The coastal marine aerosol is shown to be a highly variable function of the interaction between synoptic and mesoscale processes. Important meteorological parameters such as wind speed, relative humidity, and stability are dependent upon secondary circulations between land and sea in the absence of large scale forcing.

The minimum concentration in the size distribution curves at .4 μ - .5 μ radius may indicate that this size is indeed the transition zone between the two bubble bursting sea-salt producing mechanisms. Since the slope on either side of this zone is steeper during the Panama City experiment, wind speeds of greater than 7 m/sec result in the generation of sea-salt particles larger than .25 μ radius. Sedimentation of particles larger than 1.5 μ appears to be most significant during periods of low wind speed. During strong winds a state of equilibrium between sedimentation and production exists for these larger particles.

Relative humidity variations have the largest effect on the aerosol size distribution in the absence of sea-salt production. The concentration of the coastal marine aerosol is most sensitive to wind speed effects at low relative humidity. Friction velocity seems to be a better indication of the aerosol size distribution than wind speed under unstable atmospheric conditions. Also during light wind



periods, instability appears to result in a decrease in concentration at the observation height. Enhanced diffusion during periods of sea-salt production causes vertical transport of sea-salt from the sea surface and an increase in concentration.

Any effect of surface organic film possibly suppressing the production of small sea-salt particles could not be examined because of the absence of significant generation off the Southern California coast.

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