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Investigations on Turbulent Fluctuations of Particle Concentrations and Relative Humidity in the Marine Atmospheric Surface Layer

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Results are presented from measurements on the variation of sea spray concentrations and relative humidity (RH) in the lower 15 m of the marine atmospheric surface layer. Particle total area concentrations were determined from measurements of near-forward scattering intensities with an optical device. Both time dependent variations and mean values were recorded as a function of height from 0.5 to 15 m. The profiles of particle total area concentrations and relative humidity show trends similar to previously published particle concentration profiles measured with a rotorod impaction sampler. Hence our previous results are confirmed by using a completely different technique, based on different physical principles. The time dependent measurements show that fluctuations in the particle total area concentrations are correlated with the motions of the waves under the sampler. Spectral analyses reveal a height dependent frequency, which may be due to the wind profile, to the wave-induced turbulence structure, and to changes in the particle size distribution. Simultaneous time-resolved measurements of RH do not reveal a distinct wave influence. This analysis indicates that water vapor is transported by turbulent diffusion on a time scale much faster than 1 s. The time scale for the response of the largest sea spray particles to the the surface displacements is estimated to be longer than 5 s. Theoretical implications are indicated. The results have implications for humidity transport and the constant flux assumption in the surface layer and may be applied to study interaction between wind and waves in the open ocean.

1. INTRODUCTION

Aerosols are important to a large number of processes in the marine boundary layer (MBL). Particles that are freshly produced at the surface affect the moisture and heat budgets by evaporation. Hence they influence both the micrometeorological climate and larger-scale meteorological processes and climatology. Suspended aerosols, by scattering and absorption of radiation, contribute to the attenuation of electromagnetic radiation in the atmospheric transmission windows. This limits the performance (range) of both active and passive electro-optical systems. The visibility decreases owing to the reduction of the contrast ratio by scattered light, which not only limits the performance of electro-optical systems but also has impacts on our experience of everyday life. Aerosols are also of importance for air and water pollution and cloud physics as well as electro-optical surveillance of the marine boundary layer and the sea surface from aircraft and satellites.

In view of the above, a model for the vertical structure of the marine aerosol is required to explain a wide range of processes in the MBL. Adequate descriptions of the particle size distributions under the ambient atmospheric conditions are not available, although good progress has been made recently for a model at ship deck level (10 m) [*Hughes*, 1987]. This model could be used as input to describe the vertical structure. On the other hand, such a vertical structure model requires a mathematical framework describing the micrometeorological processes that determine the (vertical) dispersal of particles in the MBL.

Recently a model was proposed by *de Leeuw* [1989], based on a mixed layer model by *Fairall et al.* [1982], with the assumption of equilibrium. This model predicts the correct trends in 65% of the profiles of 1.06-μm backscatter coefficients measured in the mixed layer (up to 1 km) during our 1983 experiments in the North Atlantic [*de Leeuw*, 1986*a*]. A comparison with profiles of particle concentrations, measured during the same and other campaigns with a rotating impaction sampler [*de Leeuw*, 1986*b*, *c*, 1987], shows that the predicted concentrations near the air-sea interface are too high for particles larger than about 10 μm in diameter. These particles have the largest impacts on the heat and moisture budgets. Hence for application of the model to quantify the humidity exchange over the sea, the surface flux profile relationship has to be reconsidered.

The mixed layer model applies best to the smallest particles observed in the above experiments (10 μm in diameter). For various applications (e.g., optical and near-infrared properties), particles much smaller than this are of major importance. In addition, under breaking-wave conditions (see *Monahan and O'Muircheartaigh* [1986] for a comprehensive discussion of this phenomenon) the observed profiles show minima and maxima. These observations were confirmed by particle measurements during the Humidity Exchange Over the Sea (HEXOS) pilot experiment (HEXPILOT) at the North Sea in 1984 [*de Leeuw*, 1987]. The observed phenomena were assumed to be caused by particles that are temporarily trapped in eddies that come into existence by the separation of flow at the stagnation points on breaking waves [*de Leeuw*, 1986*b*, 1987]. In the following this is referred to as the wave rotor mechanism.

Processes competing with the wave rotor mechanism that are to be considered in the models include turbulence, eddy diffusion and direct production of particles by wave disruption. These processes will have different effects on particles of different sizes. Theoretical considerations of the various processes may be used to decide on the relative importance of each of them. The complexity of the problem [cf. *Davidson and Fairall*, 1986; *Fairall and Davidson*, 1986] and the lack of data to test the assumptions involved indicate the need for more

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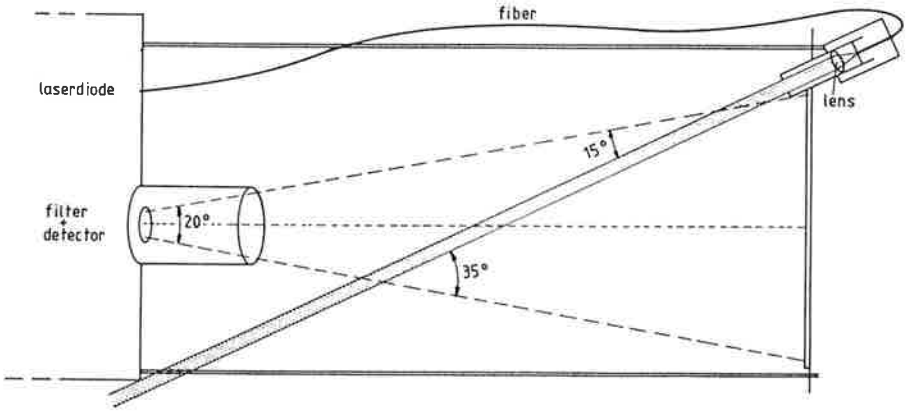

Fig. 1. Schematic representation of the scatterometer.

experiments. Laboratory experiments under controlled conditions [*Mestayer et al.*, 1988] are expected to contribute substantially to the modeling of particle behavior after ejection at the sea surface. However, more field experiments are required to validate in oceanic conditions the conclusions derived from the laboratory experiments.

Not only the height dependence above the air-sea interface is of importance, but also time dependent behavior and in particular the cross correlation between the particle concentration variations and the wave phase. The previous measurements of size distributions of particles larger than 10 μm in diameter [*de Leeuw*, 1986*b*, *c*, 1987] were averages over 4 min. Time dependent measurements, on the other hand, would yield valuable information on the phase relationship between the particle density fluctuations and the surface displacements. This could lead to better insight into the various mechanisms determining particle dispersal near the air-sea interface.

Furthermore, the production of the particles is expected to be inhomogeneously distributed over the sea surface (see *Wu et al.* [1984], who observed discrete patches of particles in the 40- to 400-μm-diameter range, with a frequency similar to the oceanic wave frequency). The highest production rates are expected in the whitecap regions, but in very high winds, direct production by wave disruption is favored. The relative contributions from both these production mechanisms could be revealed from time dependent measurements.

In this paper we report measurements of the height and time dependent behavior of sea spray aerosols and simultaneous measurements of relative humidity. Variations in the aerosol total area concentrations were derived from measurements of forward scattering intensities (G. de Leeuw et al., An angular scattering device for the determination of turbulent fluctuations of aerosol concentrations, submitted to *Review of Scientific Instruments*, 1988, hereinafter referred to as de Leeuw et al., 1988). The results from these optical measurements confirm our previous observations with an impaction technique that were described in the preceding paragraphs. Hence two independent methods, based on different physical principles, yield similar results.

The optical device is fast enough for time dependent measurements of the interactions between the waves and the aerosols. Time series of aerosol concentrations as well as of relative humidity were measured at heights between 0.5 and 3 m above the instantaneous sea surface by using a wave follower, and at fixed heights between 1 and 13 m above the mean sea surface. The measurements were made during the HEXOS Main Experiment (HEXMAX) [*Katsaros et al.*, 1987] in October and November 1986 at Meetpost Noorwijk, a platform in the North Sea 9 km from the Dutch coast. The equipment was mounted on a horizontal mast extending in a northerly direction from the NW corner of the platform. In this way the measurements were made at 13 m from the platform structure. Flow perturbation is expected to be less than 2–3% at this distance, as deduced from a wind tunnel study by *Wills* [1984].

2. Description of the Optical Scatterometer

Forward scattering by particles was measured with an instrument developed by the Physics and Electronics Laboratory, Netherlands Organization for Applied Scientific Research (TNO). This optical scatterometer has been discussed in detail by de Leeuw et al. (1988). The scheme of the instrument is shown in Figure 1. Pulsed near-infrared radiation (wavelength of 0.904 μm) is emitted by a diode laser. An optical fiber is used to guide the laser beam to the emitter side of the scatterometer. The divergent beam at the end of the fiber is collimated to form a parallel beam that is directed into the sample volume, under an angle of 25° with the optical axis of the receiver. The receiver consists of a photodiode and a lens. With a receiver field of view of 20°, radiation is detected that is scattered by the particles in the sample volume, at angles between 15° and 35° from the forward direction of the laser beam. The detected pulses are amplified and further processed with a synchronized peak detector and a computer-controlled analog to digital converter.

The emitter laser diode and the receiver photodiode are mounted in a single watertight box which also contains the electronics. The receiver and emitter sides are connected with two rigid rods, which allows a free airflow through the sample volume. The receiver looks into a beam dump to shield it from direct solar irradiation or reflections from the water surface which may cause spurious background signals. Both units are mounted in protective cylinders to shield them from pollution by turbulent deposition of sea spray aerosols.

The advantage of the detection of scattered light, as compared with the extinction measurements used for instance by *Monahan* [1968] and by *Wu et al.* [1984] is the sensitivity to smaller particles. The optical scatterometer used in the present experiments is not capable, however, of detecting individual particles. This is due to the relatively high concentrations of the particles contributing to the scattered signal. Therefore

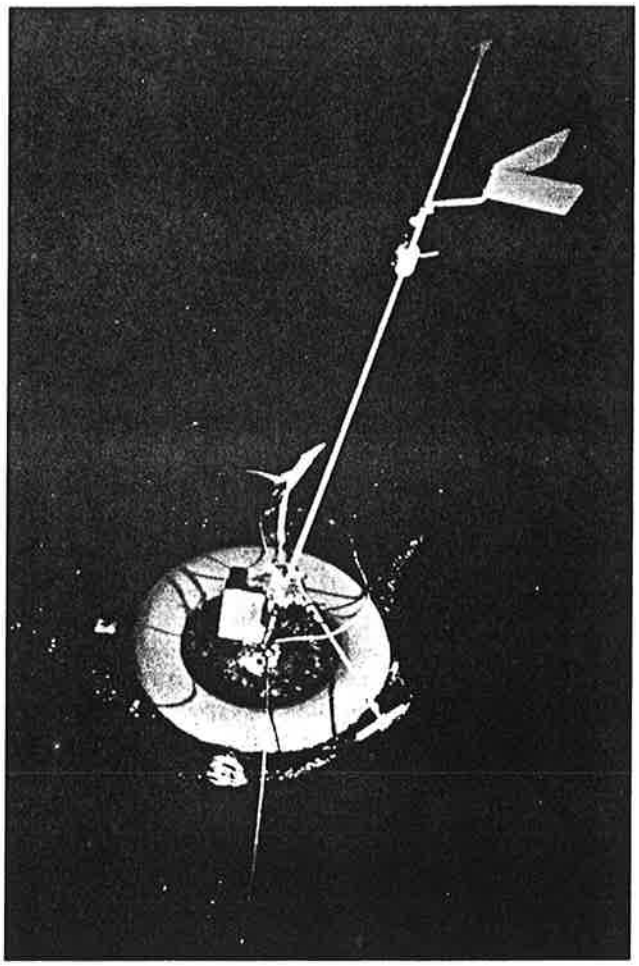

Fig. 2. Wave follower with scatterometer and humidity sensor. In this photo the humicap was not shielded from pollution by deposition of sea spray aerosol.

size distributions could not be obtained from our experiments. As was shown by de Leeuw et al. (1988), the scatterometer signals are proportional to the aerosol total area concentrations. In the following these are briefly referred to as aerosol concentrations.

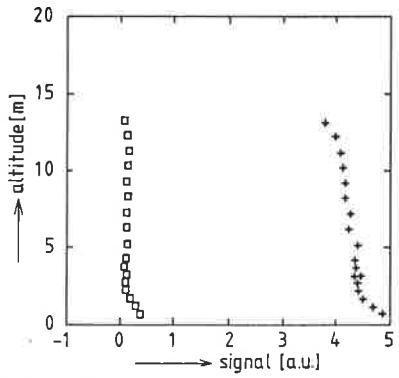

Fig. 3. Profiles of mean particle concentrations (squares) and relative humidity (pluses) measured in a wind speed of 7.7 m/s, on November 15, 1986, between 1635 and 1652 UT. The particle data were not calibrated; the humidity data were calibrated afterward and are plotted on a scale from 0% (0 arbitrary units, or a.u.) to 100% (5 a.u.). Note the strong gradients near the surface in both the particle concentrations and the relative humidity.

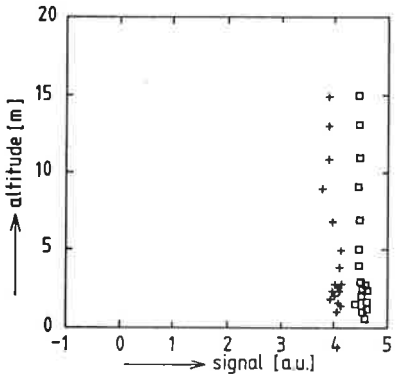

Fig. 4. As Figure 3, in a wind speed of 14.2 m/s, on October 24, 1986, between 1250 and 1400 UT. The gradients that were observed near the surface in moderate winds (compare Figure 3) have disappeared.

3. HUMIDITY MEASUREMENTS

Simultaneously with the measurements of forward particle scattering, relative humidity was recorded with a Väisälä Humicap capacitance hygrometer, mounted in a 30-cm-long protective tube with forced aspiration to shield the sensitive element from pollution by sea spray droplets. The response time of this instrument is about 1 s.

4. EXPERIMENTS

The scatterometer weighs only 2.4 kg, exclusive of the cables for power supply and data transmission, and the humicap weighs 1.4 kg. This allows for mounting of the instruments on the simple wave follower that is also used for measurements of particle size distributions near the air-sea interface [*de Leeuw*, 1986*b*, *c*, 1987] (see Figure 2). In this way, measurements could be made at heights of 0.5 to 3 m from the instantaneous sea surface. The vertical resolution was 0.25 m. In addition, data were recorded at fixed heights above the mean sea surface for profiling of the surface layer up to about 15 m with a resolution of 1 m.

The analog signals from the scatterometer and the humicap were recorded with a computer-controlled analog to digital converter. Either time series were recorded or the data were directly averaged for a quick look at the surface layer profile. As will be shown, however, the particle concentrations vary over a wave period, and rather long averaging time had to be chosen to obtain reliable means. The recording speed of the time series was about 200 Hz in this configuration.

5. RESULTS

For the experiments presented in this paper the scatterometer had not been absolutely calibrated. Because of temperature drift the response may have varied from day to day. Therefore the instrument was allowed to stabilize under ambient conditions before each measurement. Measurements at the same height but separated in time by several hours gave similar results when the conditions had not changed.

In some cases the response changed during the measurements at the lowest heights owing to pollution of the windows by spray deposited at the receiver lens (the protective cylinder was kept short to retain the full field of view). After each series of measurements the scatterometer was checked carefully for such effects. The results presented in this paper were not influenced by spray at the windows.

The consistency of the results is also indicated by the profiles shown in Figures 3–5. With the scatterometer and humi-

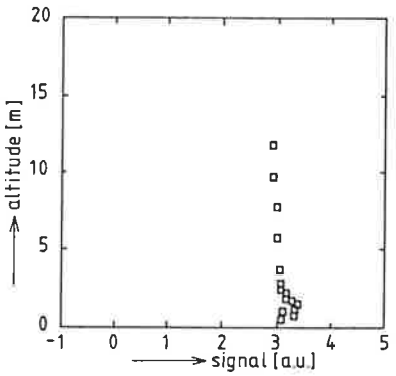

Fig. 5. Profiles of mean particle concentrations measured in a wind speed of 12 m/s on October 22, 1986, between 1615 and 1705 UT. The concentration maximum between 1 and 2 m is in good agreement with previous observations of particle size distributions [*de Leeuw*, 1986*b*, *c*, 1987].

cap moving downward, a sample was taken every 2 m down to 3–4 m above the mean sea surface. Then samples were collected with the instruments mounted on the wave follower. The profile was completed with another set of measurements taken with the instruments going up, at such heights that the spaces between the previous altitudes were filled up. Thus profiles were obtained with data at intervals of 1 m and better vertical resolution near the air-sea interface. In most cases the profiles obtained while going upward and downward were consistent.

6. Profiles of Mean Particle Concentrations

Examples of profiles of mean particle concentrations are shown in Figures 3, 4 and 5. Since the scatterometer was not absolutely calibrated, the mean particle concentrations are presented as the amplitude (in arbitrary units) of the signal received by the scatterometer due to scattering of 0.904-μm radiation by the particles, at angles between 15° and 35° (see section 2).

The profiles in Figure 3 were measured on November 15, 1986, in a wind speed of 7.7 m/s. The data at 0.8 m were recorded with the sampler at fixed height, just above the wave tops. The data at 0.5 m were recorded with the scatterometer mounted on the wave follower. The data at 13.3 m and down to 1.3 m (at 2-m intervals) and the data at 2.3 m and up to 12.3 m (also at 2-m intervals) fit perfectly into one profile. Above 2.3 m the concentrations are almost constant, while close to the sea surface a strong gradient is observed. At 0.5 and 0.8 m the concentrations are similar.

The particle concentration profiles vary with ambient conditions, and an obvious increase toward the surface, as displayed in Figure 3, was observed in only a few cases. Examples of other types of profiles are shown in Figures 4 and 5. The profile shown in Figure 4 was recorded on October 24 in a wind speed of 14.2 m/s. Although some scatter is observed in the data below 3 m, these do not indicate an obvious gradient near the sea surface. The data in Figure 5, on the other hand, show an increase of particle concentrations below 3 m, with a maximum occurring at about 1.5 m, followed by a decrease in the concentrations down to about 0.5 m. These data were recorded in a wind speed of 12 m/s.

The three examples in Figures 3, 4, and 5 confirm previous measurements of particle size distribution profiles for particles larger than 10 μm. The latter were measured with a rotating impaction sampler [*de Leeuw*, 1986*b*, *c*, 1987]. A model was proposed to explain the variations of the profiles with wind speed. The theoretical aspects will be further pursued in the discussion.

7. Profiles of Mean Relative Humidity

Profiles of mean relative humidity (RH), measured simultaneously with the forward scattering profiles, are also plotted in Figures 3 and 4 (in Figure 5 the RH data are missing owing to hygrometer malfunction). The variation of RH with height was observed to be similar to the particle concentration profiles in 80% of the 45 cases for which validated profiles were available for both particle concentrations and relative humidity. This indicates a strong coupling between the relative humidity and the particle concentration. In particular, near the sea surface, freshly produced particles with the composition of sea water evaporate until equilibrium is reached with the ambient relative humidity. Obviously, this enhances the local water vapor concentration. On the other hand, the humidity profile is also affected by the evaporation at the sea surface and the (upward) transport of water vapor.

Some of the mechanisms that are responsible for the transport of particles also influence the transport of water vapor. Since gravitational forces on molecular species can be neglected, vapor is more likely than the particles to be removed from the surface layer by turbulence. The observed similarities between the profiles of particle concentrations and the profiles of RH is therefore indicative of the release of water vapor from the freshly produced particles. Theses considerations lead to the conclusion that the droplet fluxes (i.e., the fluxes of liquid water) near the air-sea interface must decrease with height owing to the release of water vapor that is removed by turbulent diffusion.

8. Time-Resolved Measurements of Particle Concentrations

Time series of forward scattering intensity and RH, recorded at fixed heights of 2, 4, 8, and 12 m above the mean sea surface, are shown in Figure 6*a*. In Figure 6*b*, similar data are presented that were recorded from the wave follower at heights of 1, 2, and 3 m above the instantaneous sea surface. Fluctuations in the particle concentrations are clearly observed throughout these figures. The amplitudes of the concentration fluctuations decrease with height above the mean sea surface (Figure 6*a*). Visual observations, supported by video recordings, indicate that the fluctuations are correlated with the motions of the underlying water surface.

The wave structure in the North Sea is complicated, and the wavelengths are variable. The intervals between the passage of crests under the sampler were observed to vary from less than 2 s to more than 5 s. As a result, some of the time series show irregular intensity variations, although the variations in other samples show a rather periodical behavior. This is reflected in the Fourier spectra determined from the scatterometer signals. As an example we show in Figure 7 the Fourier spectrum calculated from the time series recorded at 2 m above the mean sea surface (compare Figure 6*a*). Features in the Fourier spectra include the following:

1. Most spectra have a dominant range of frequencies between about 0.3 and 0.6 Hz with a maximum relative energy density near 0.5. Secondary maxima are observed both at lower frequencies (0.2 Hz) and at higher frequencies (0.8 Hz). This indicates that the time scale for particles to respond to

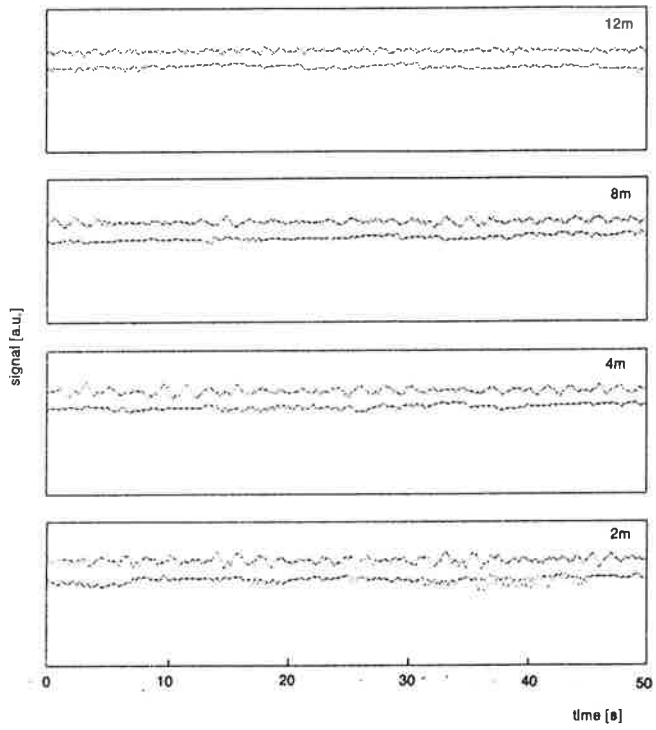

Fig. 6*a*

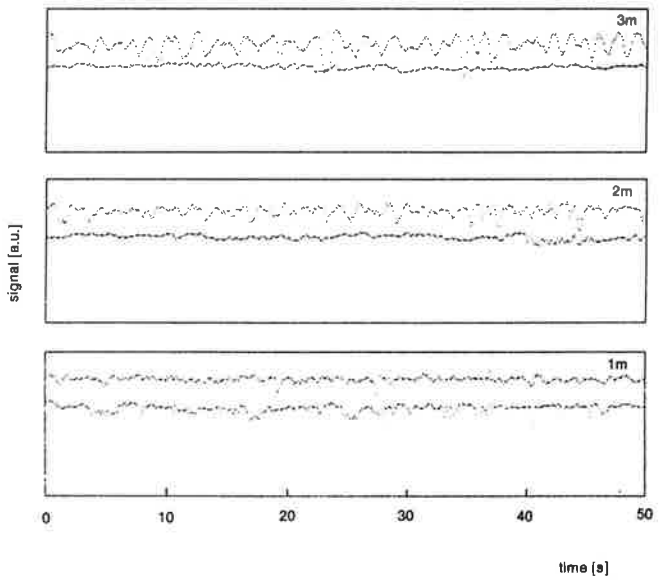

Fig. 6*b*

Fig. 6. Time series of forward scattering intensities (top curves) and relative humidity (bottom curves), both in arbitrary units. The data were recorded on October 24 in a wind speed of 12.1 m/s. Reference RH was 66.4%. (*a*) Measurements at fixed heights of 2, 4, 8, and 12 m above the mean sea surface. (*b*) Wave-following measurements at 1, 2, and 3 m above the instantaneous sea surface. Note that the data at different heights are not correlated, since the measurements were made consecutively.

the surface displacements is more than 5 s. On the other hand, these time scales will be strongly dependent on particle size, with faster time scales for the smaller particles.

2. The dominant frequency increases with increasing height above the mean water surface. The maximum increase we observed is 20–40%. Explanations for this increase are presented in the discussion.

3. In some of the Fourier spectra derived from wave-following measurements at specific heights (approximately equal to the wave height), a dominant frequency is missing.

4. In some Fourier series the ratio of the energy densities at the dominant frequency and at the next dominant higher frequency changes as the height above the instantaneous sea surface increases (wave-following measurements). As an example, for the data in Figure 6, the ratio of the energy densities of the peaks at approximately 0.8 and 0.5 Hz first increases from 0.5 at 0.5 m to 2 at 1.5 m, goes through a minimum at 1.75 m, and reaches a second maximum of 2 at 2.25 m.

These features are further discussed later on in this paper.

9. Time-Resolved Measurements of Relative Humidity

Turbulent fluctuations in the relative humidity are not obvious in our data set. In contrast to the expected behavior, the spectral analyses of the time series do not display any frequency dependence. Probably, this is due to the slow response of the humicap. On the other hand, this instrument should have been fast enough to indicate slow fluctuations due to the waves. The absence of any wave influence in the humidity spectra indicates that water vapor is apparently equilibrated by turbulent diffusion at a time scale much faster than the one that applies to particle transport. The same conclusion was reached from comparison of the profiles of mean particle concentrations and mean humidities. Apparently, the surface displacements are slow enough to allow for "instantaneous" adjustment of the humidity profile. This leads to the conclusion that the time scale for humidity transport must be much faster than 1 s (i.e., the response time of the Humicap).

10. Discussion

The results presented in the previous sections are a selection from a comprehensive data set on the behavior of particles in the marine atmospheric boundary layer that was collected by the Physics and Electronics Laboratory TNO during the HEXMAX experiment. Mixed layer studies with lidar [cf. *de Leeuw*, 1988], measurements of aerosol particle size distributions with optical particle counters from the deck of the platform, and profiles of particle size distributions in the surface layer [cf. *de Leeuw*, 1986*b*, *c*, 1987] complement this data set. In addition, extensive meteorological and aerosol data were collected by other HEXMAX participants at Meetpost Noordwijk, aboard a ship and aircraft, and on shore (see *Katsaros et al.* [1987] for a description of the experiment). A complete analysis of all our data has not been made yet. Nevertheless, the results presented in this paper show some interesting trends which are discussed below.

The mean profiles of particle concentrations presented in Figures 3–5 were deduced from the intensities of 0.904-μm electromagnetic radiation scattered by particles in the forward direction (15°–35°). The scatterometer was developed as a small and lightweight instrument that could be used for measurements on our wave follower. The objectives of these measurements were (1) to confirm previous observations of particle concentration profiles with a Rotorod rotating impaction sampler [*de Leeuw*, 1986*b*, *c*, 1987] and (2) to find indications on the particle transport mechanisms near the air-sea interface (i.e., wave rotor and surface undulation effects versus higher-frequency turbulence and buoyancy).

The data in Figures 3–5 confirm our previous observations concerning the shapes of profiles of concentrations of particles

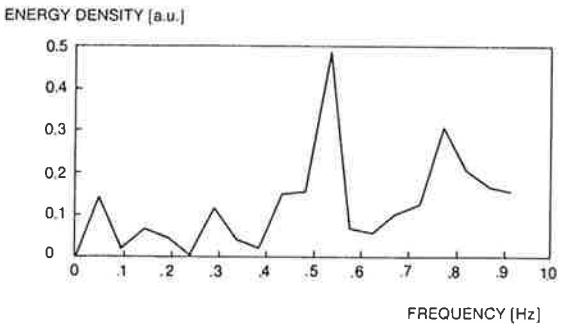

Fig. 7. Fourier spectrum of the scatterometer signals measured at fixed height of 2 m above the mean sea surface (compare Figure 6*a*).

larger than 10 μm in diameter [*de Leeuw*, 1986*b*, *c*, 1987]. The two experiments are based on different physical principles. Thus we have independent sampling methods which give consistent results.

We have thus established that the observed variations in the particle concentration profiles are not due to an artifact. On the other hand, much scatter occurs in the whole data set, and not all profiles show trends that are as obvious as those presented here and in previous publications. In many cases the profiles are mixtures of either those in Figures 3 and 5 or those in Figures 4 and 5. Altogether, the three types of profiles presented in these figures have about the same frequency of occurrence. This will also depend on atmospheric conditions, however. During the HEXMAX experiment, high-wind conditions prevailed.

Now it has to be decided which physical processes are responsible for the observed variations of particle concentrations with height and which processes are responsible for the frequently encountered deviations from the observed trends. Competitive mechanisms to be considered are turbulence and wave-induced perturbations (including eddies formed by separation of flow on breaking waves).

Assuming that all memory concerning the initial ejection velocity has been lost relatively fast, the particles are subjected to the forces exerted on the surrounding air parcel. Therefore the changes in the particle velocities are caused by the fluctuating flow velocity field. The horizontal flow velocity field $u(x, z)$ can be described by

$$u(x, z) = U(z) + U_w(x, z) + u'(z) \tag{1}$$

where $U(z)$ is the mean wind field along the horizontal coordinate x that depends on the altitude z, $U_w(x, z)$ is the deviation of the velocity field due to the waves (i.e., due to flow separation including surface undulation effects), and $u'(z)$ represents the turbulent deviation from the mean velocity field. $U_w(x, z)$ varies along a wave. Similarly, the vertical flow velocity field is given by

$$w(x, z) = W(z) + W_w(x, z) + w'(z) \tag{2}$$

For the description of particle motions, a slip velocity (u_p, w_p) should be included (compare (3) and (4)).

In view of the wave rotor term (U_w, W_w), horizontal advection effects cannot be neglected near the surface. For simplicity we assume a uniform wave pattern (which usually does not apply in the North Sea) and neglect lateral flow perturbations. Hence for the description of particle concentration fluctuations in the marine atmospheric surface layer the continuity equations should be used in two-dimensional form.

Alternatively, a one-dimensional approximation could be used which takes the horizontal advection effects into account. *Ling and Kao* [1976], for example, proposed a one-dimensional K model, with K representing an effective average over a wave period. However, such an approach is not expected to yield useful information on the competition between the wave rotor mechanism and higher-frequency turbulence in the dispersal of particles near the air-sea interface. *Ling and Kao* [1976] present results from calculations on droplet, humidity, and temperature profiles, evaluated for different stabilities and reference relative humidities, all at a wind speed of 15 m/s. The predicted droplet profiles decrease in all cases, in contrast with our observations. In particular, strong gradients were predicted between the troughs and the crests of the waves. This might be caused by the use of a height dependent normalized source strength, which is strongest in the trough. Also the size of the particles considered by Ling and Kao ($D = 190$ μm) may be of influence. In the field, such large particles are rarely observed [*de Leeuw*, 1986*c*]. In fact, they have been observed thus far only from rafts at altitudes below 1.5 m [*Monahan*, 1968; *Wu et al.*, 1984]. Therefore the phenomena we observed with the optical scatterometer will be caused mainly by somewhat smaller particles (e.g., in the 10- to 100-μm-diameter range) which are more abundant. On the other hand, the wave tank experiments by *Koga and Toba* [1981] show a tendency for large particles ($D > 450$ μm) to be vertically uniformly distributed in the wave troughs.

As an alternative to avoid the complexity of solving the two-dimensional continuity equations, a Lagrangian approach could be used, i.e., the numerical solution of the equations of motion for a statistically relevant number of particles. Following *Wu* [1979], the x and z components describing particle motion can be written as

$$du_p/dt = -3/4\ C_d(\rho_a/\rho_w)(u - u_p)\ |u - u_p|/D \tag{3}$$

$$dw_p/dt = -3/4\ C_d(\rho_a/\rho_w)(w - w_p)\ |w - w_p|/D \tag{4}$$

where the velocity field (u, w) is given by (1) and (2) and includes the wave effects. C_d is the drag coefficient, which changes with Reynolds number and thus with the particle diameter D, and ρ_a and ρ_w are the densities of air and water. The terms $u - u_p$ and $w - w_p$ account for the finite response times of the particles to changes in the velocity of the surrounding air parcels (i.e., a slip velocity term).

In addition, the response to changes in RH have to be taken into account, since these affect the particle diameter D (see *Mestayer and Lefauconnier* [1988] for a discussion):

$$dm/dt = -2\pi D f D_v(\rho_a/\rho_w)(q_{sd} - q) \tag{5}$$

D_v is the diffusivity of water vapor, f is a ventilation factor, q_{sd} is the saturation specific humidity at the droplet surface, and q is the specific humidity of air close to the droplet.

Mestayer et al. [1988] used the Lagrangian approach to calculate the turbulent transport of particles from a point source. The results compare favorably with wave tank simulations of this model. Wave influences were not considered, however, either in the calculations or in the experiment.

Inclusion of the wave-induced terms U_w and W_w into the wind profiles would reveal the importance of the wave rotor mechanism on particle transport near the air-sea interface. In addition, the combination of such a theoretical approach with field experiments could yield valuable information on the flow fields in the surface layer. In particular, the occurrence of flow

separation in the wave troughs, hard to measure under oceanic conditions, might be established.

In addition to the theoretical approach, time dependent measurements as presented in this paper may prove to be a valuable contribution to the solution of the above problems. The analysis of the experimental data yields some interesting results. In the following these are discussed and compared with some other relevant experiments.

Our data are in good agreement with the results from measurements on wave-induced turbulence by *Lai and Shemdin* [1971]. As in the present experiments, Lai and Shemdin observed a wave-induced peak in the frequency spectrum, which decreases with height above the mean water level.

The dominant frequencies in the particle concentration spectra occur at 0.3–0.6 Hz. Wave spectra in the North Sea usually have a maximum energy density near 0.1–0.2 Hz, whereas the frequencies of the swells are even lower. The occurrence of a dominant peak in the particle concentration spectra at higher frequencies than the wave spectra indicates that the wave-induced fluctuations in the particle concentrations are more sensitive to the wave frequency than to the energy in the waves. Apparently, the longer waves are slow enough to allow for adjustment of the particles to the wave-induced surface displacements. The occurrence of a secondary peak at 0.2 Hz in the particle spectra therefore indicates a longest time scale for the response of particles to surface displacements of more than 5 s. This time scale applies to the largest particles. Obviously, shorter time scales will apply to smaller particles. The shift of the dominant frequency in the particle concentration spectra with height above the sea surface might therefore reflect a change in the particle size distribution. Owing to gravitational settling, the larger particles are removed somewhat faster than the smaller ones, resulting in larger gradients for the larger particles [cf. *de Leeuw*, 1986*b*, 1987]. Hence the particle size distribution becomes more skewed with increasing height above the sea surface. Consequently, the influence of the smaller sea spray particles on the spectra increases. Since the response time scales will be shorter for these particles than for the larger ones, this results in a shift of the particle concentration spectra toward higher frequencies.

This effect of the change in particle size distribution will be enforced by the damping of the effect of the longer (read slower) waves at higher altitudes. The slower waves allow more time for the concentrations to adjust. Therefore at larger heights the effects of the slower waves will be damped out, thus giving rise to a shift in the frequency spectrum because shorter waves gain relatively more importance.

Comparison of particle spectra with simultaneously measured particle size distributions, both at various heights, could yield more experimental information on the size dependent time scales.

In addition, the observed height dependence of the frequency of the particle concentration fluctuations might also be caused by some other mechanisms, and the various mechanisms could enforce each other. The increase of wind speed with height above the sea surface, for example, results in an increase of the particle advection velocity. In that case, a sampler that is fixed at a horizontal position "sees" the aerosol patches at higher frequencies as altitude increases. This explanation is supported by the observations by *Lai and Shemdin* [1971] concerning the phase shift between the wave-induced velocity and the surface displacement. In low wind the horizontal velocity perturbation was observed to lag behind the surface displacement, while in high wind it leads the wave. This applies both to measurements at fixed height and to wave-following measurements. The phase shifts are height dependent.

Another alternative could be that the eddies that come into existence in the wave troughs break up into smaller turbulence cells as they move higher up. This too would result in the observed increase of frequencies with height above the waves.

The preliminary analysis of the frequency variations in the wave troughs shows that three regions may be discerned, with different height dependencies of the frequency spectra. Measurements near the significant wave height yield a rather smooth spectrum; i.e., dominant frequency ranges are not observed. Apparently, many frequency components contribute to the particle concentration fluctuations. We suppose that the mean heights of these measurements correspond with the mean critical (or matched) level, where the wind speed equals the wave speed [cf. *Kraus* [1972]. In this concept a matched layer exists around the critical level, where the streamlines are closed. The extent of the matched layer depends on the ratio of the wind speed to the wave speed [*Lai and Shemdin*, 1971]. However, in a nonuniform wave field the height of the matched layer will vary from wave to wave. In that case, a sampler mounted on a wave follower at a height near the critical level passes many streamlines. Consequently, it sees a variety of frequencies during the sampling period, which results in a smooth frequency spectrum, as observed during some of our experiments. When the sampler is mounted below or above the mean critical level, the measured spectrum may be dominated by a small range of frequencies, which will depend on the streamline pattern. It is well conceivable that close to the air-sea interface a single frequency which is approximately equal to to the wave frequency will dominate, while further away from the interface higher frequencies become more dominant. This has also been observed in our measurements, as described in the experimental sections. A theoretical approach to verify these mechanisms was outlined earlier in this section.

As discussed above, knowledge of the phase relation between particle concentrations and surface displacements may be useful to explain our observations. However, also the production mechanisms will influence the phase shifts, in contrast to the turbulence measurements [e.g., *Lai and Shemdin* 1971] which are not affected by the particle concentrations.

The distribution of particle concentrations along a wave has been studied by *Koga and Toba* [1981] for submillimeter particles. In these wave tank studies the highest concentrations were observed in the lee of the wave. In the oceanic measurements by *Wu et al.* [1984], patches of particles were observed with a frequency similar to the wave frequency, for particles in the 40- to 400-μm range. However, the wind conditions during both experiments were quite different. The wave tank measurements by Koga and Toba were made in a reference wind speed of 16 m/s but at a fetch of only 16 m, whereas Wu made his oceanic measurements at a very long fetch but in wind speeds of only 6–8 m/s. The wave tank data were explained to be caused by direct wave disruption, a mechanism that was unlikely to occur during the experiments by Wu et al. The particles observed by Wu et al. must have been large jet drops which probably had just been ejected from the surface. During our previous measurements of particle concentration profiles in the North Atlantic, very few particles larger than 64

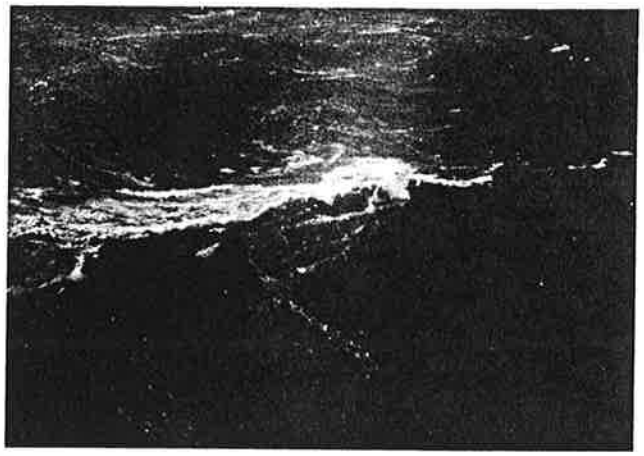

Fig. 8. Breaking wave photographed from Meetpost Noordwijk. The whitecap is at the windward side of the breaking wave.

μm were detected in winds of 6–8 m/s, however, and the concentrations of these particles were less than $10^{-6}/\mu m/cm^3$ [cf. *de Leeuw*, 1986*b*].

In view of the prevailing strong winds during the HEXMAX experiment, the particles responsible for the fluctuating scattering intensities may have been both directly produced particles and large jet drops. Directly produced particles are likely to be observed at the lee of the crest. However, as is shown in Figure 8, the whitecap of a trailing wave can be at the windward side of the crest, where it would give rise to high particle production rates. Because of the interaction with the wind fields, the directly produced particles and the jet drops will be mixed, but close above the air-sea interface the two production mechanisms are expected to yield different phase relations between the particle concentrations and the wave motions.

An analysis of the scatterometer measurements in relation to our simultaneous video recordings is expected to reveal more information on this subject. New experiments are required to obtain better insight, however, and simultaneous recordings at the same place should be made of surface displacements and fluctuations in the particle concentrations. Also, simultaneous measurements should be made of turbulent fields of humidity and flow velocities, together with determinations of particle size distributions, preferably at various heights.

The results presented in this paper have various implications. The enhanced residence times of large sea spray particles, caused either by the proposed action of the wave rotor mechanism or by turbulence, will affect the moisture and temperature fields near the surface. More time is available for evaporation as the particles are suspended longer. Further, the constant flux assumption that is frequently used in mixed layer models does not apply under these conditions. As discussed above, the time scale for the transport of water vapor by (molecular) diffusion (<1 s) is much faster than the time scale for particle transport, i.e., the transport of liquid water (more than 5 s for the largest droplets). Hence the loss of liquid water by evaporation of the particles cannot be balanced by condensation on the droplets that fall back into the sea. As a result, there is a positive flux of liquid water, which varies with height above the sea surface. Also the total moisture flux (i.e., liquid water and vapor) will not be constant.

11. SUMMARY AND CONCLUSIONS

A fast scatterometer has been constructed for time dependent measurements of fluctuating aerosol concentrations near the air-sea interface. The instrument was used both for profiling of the surface layer (0–15 m) and for recording of time dependent fluctuations. Relative humidity was recorded simultaneously. The similarities between the particle concentration profiles and the humidity profiles indicate that the humidity gradients near the air-sea interface are strongly influenced by the evaporation of freshly produced sea spray particles. This leads to the conclusion that liquid water is transported from sea to air, while the vertical flux of liquid water (as droplets) near the air-sea interface will decrease with height. This is in contrast with the constant flux assumption that is frequently used in mixed layer models.

Time dependent measurements revealed the influence of the waves on the particle concentrations, which may extend to above the highest altitude where we measured (15 m). The particle concentration fluctuations are correlated with the wave motions. Spectral analyses show that the fluctuations at the dominant frequencies are related to the wind waves, rather than the swells which usually contain more energy. As a result, the particle fluctuation spectra are shifted to higher frequencies with respect to the common wave spectra.

The frequencies are height dependent, which may have various causes that could add to the observed features. The shift of the particle size distribution to smaller sizes with increasing height above the water surface would result in a shift of the frequency spectrum toward higher frequencies because the response time of the smaller particles to the surface displacements is shorter. Hence higher-frequency waves become more dominant at higher altitudes because the effect of the longer waves is damped. In addition, there is the effect of the wind profile and the effect of breaking up of the eddies into smaller ones. From the observed spectra the response time scale for the largest sea spray particles to the surface displacements is estimated to be longer than 5 s.

The humidity spectra are not correlated with the surface displacements. This indicates that the time scales for the transport of water vapor are much faster than those for particle transport. The spectra indicate that the response requires less than 1 s.

The results are of importance for modeling of production and dispersal of aerosol particles near the air-sea interface. A theoretical approach has been outlined which may be used to obtain inferences from the experimental data on the contributions of various transport mechanisms, such as turbulence and the previously proposed wave-rotor model. The application of this approach may also yield information on flow separation in oceanic conditions, which is important for wave generation models.

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