11. SURFACE-LAYER PROFILING OF AEROSOL CONCENTRATIONS, PARTICLE SIZE DISTRIBUTIONS AND RELATIVE HUMIDITY.

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Introduction

Particle-assisted transport of liquid water may contribute substantially to the humidity exchange over the sea. Obviously a significant increase in the humidity content of the surface layer (defined here as the lower 10-20 m above the air-sea interface) is due to the evaporation of freshly produced sea spray droplets, but also liquid-vapour conversion at higher altitudes may affect the humidity profile. Little information is available, however, both on the production of particles at the sea surface and on the subsequent dispersal. The production estimates by Monahan et al. [1986] rely on an empirical model based on whitecap observations combined with wavetank measurements of the production rate per whitecap area. Fairall et al. [1983], on the other hand, derived their estimates from application of a mixed-layer model to particle size distributions measured at about 10 m above sea level. The vertical variation of particle concentrations predicted by this model compares favourably with experimental data obtained with lidar in the mixed-layer [De Leeuw, 1988a]. In the surface layer good agreement is often obtained for particles smaller than about 15-20 μm, but only in low-wind conditions when whitecapping is negligible. In moderate to high winds the model fails. For the large particles that are most important for humidity transport the model also fails in low winds [De Leeuw, 1988b]. Obviously the surface flux relationship that is used in the model is not correct. The surface production rate is overestimated. In high winds the particle concentrations do not increase exponentially toward the sea surface. Previous measurements [De Leeuw 1986, 1987] revealed minima and maxima in the concentration profiles which become more obvious as wind speed increases. These results were explained in terms of a wave-rotor model, cf. Figure 1. In the proposed model an eddy airflow is assumed to come into existence in the wave troughs under breaking wave conditions. The particles are assumed to be temporarily trapped in the eddies. Hence the action of the wave rotor results in longer suspension times, which could explain the shape of the profiles. Consequently, in that case also more time would be available for evaporation, which affects the humidity content of the surface layer.

In addition to the proposed wave-rotor mechanism, higher-frequency turbulence acts on the particles and keeps them in suspension, which has a similar effect on the humidity. There is no reason, however, why extended suspension caused by turbulence would result in the observed profiles. On the other hand, the interaction with the waves (i.e. deposition of particles in the waves) should also be considered, as well as the two different production mechanisms, viz. bubble mediated production of jet drops and direct production by wave disruption. The two production processes find place at different heights with respect to the water surface. Jet drops are generally produced in the wave troughs,

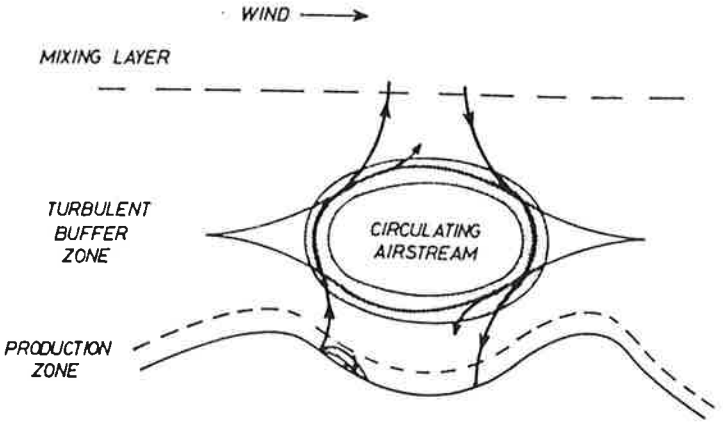

Figure 1.
Schematic representation of the wave-rotor model.

while direct production occurs at the crests. The latter becomes more important as wind speed increases and may thus contribute to the maximum in the aerosol profile that develops in higher winds.

Measurements of particle size distribution profiles alone do not yield conclusive information on the relative importance of the above processes of production and dispersal. Therefore, during the HEXMAX experiment also the time-dependent variations in the particle concentrations were measured. This was achieved with an optical device (further referred to as an optical scatterometer) that measures particle total area concentrations from the intensity of laser light that is scattered by the particles in the sensor volume between angles of 15° and 35° from the forward direction (cf. De Leeuw et al. [1988] for a description). The sample frequency is 200 Hz in the present configuration. It was mounted on our wave-rider buoy, together with a capacitance hygrometer (max. response rate 1 Hz), to determine particle concentration fluctuations and relative humidity at heights between 0.5 m and 3 m above the instantaneous sea surface. The profiles were completed with measurements at fixed height above the mean sea surface up to 15 m. Using a horizontal mast, samples were taken at 13 m from the platform structure. In addition to the measurements with the scatterometer and the humicap, particle size distribution profiles were determined with the Rotorod rotating impaction sampler. The efforts were particularly focused on the lower 5 m.

Objectives.

The objectives of our measurements during the HEXMAX experiment were.

1. to confirm previous observations on the variation of particle concentration profiles in the surface layer [De Leeuw, 1986, 1987].
2. to test the hypotheses involved in the wave-rotor model that was proposed to explain the above observations.
3. to obtain experimental information on the various production and dispersal processes near the air-sea interface and the interaction with the humidity field.

Particle size distributions.

In the storm on October 20 some samples were taken from the deck of the platform (unfortunately profiles could not be measured in these high

winds). The particle volume distribution measured at 13.25 GMT, in a windspeed of 25 m/s, is presented in Figure 2. The wave field was not fully developed during the storm. Nevertheless the particle size distribution has an extreme shape with a strong volume mode near 100 μm. In the evening the wind went down to 20 m/s. The shape of the particle size distributions hardly changed, however, for particles up to 75 μm. The volume concentrations of larger particles decreased slightly, resulting in a shift of the maximum volume concentration to 75 μm. This particle size distribution persisted all evening.

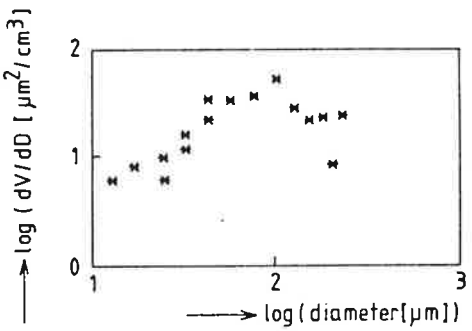

Figure 2.
Particle volume distribution measured with the Rotorod impaction sampler in the storm of October 20.
The instantaneous wind speed was 25 m/s.
Note the mode at 100 μm.

Profiles.
Profiles were recorded, both of mean particle size distributions, particle total area concentrations and relative humidity and of time-dependent fluctuations of the latter two quantities. The particle measurements were made with two devices that are based on different physical principles. The results are in good agreement (Figure 3). In some cases, however, the data obtained with both sampling methods did not reveal the same features, cf. Figure 4. In that particular case this was due to a change in the ambient conditions between the Rotorod and the scatterometer measurements. The particle size distributions in Figure 4 were recorded just before sunset, the scatterometer and humidity measurements were made after sunset. In the mean time humidity went down from 90% to 75%, while the particle concentrations increased due to the decrease in the mixed-layer depth. These features were confirmed by lidar measurements, particle measurements with Knollenbergs and by the increase in the scatterometer signal.

The optical scatterometer and the humicap were applied for simultaneous recording of particle total area concentration profiles and humidity profiles. The similarities between these profiles (cf. Figure 4) indicate that the humidity gradients near the air-sea interface are strongly influenced by the evaporation of freshly produced sea-spray particles.

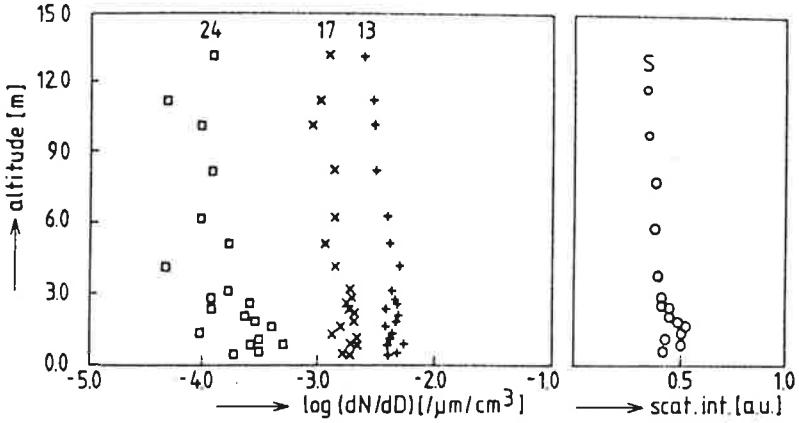

Figure 3. Profiles of particle size distributions (lhs Figure), mean particle total area concentrations measured with the scatterometer (S) measured on October 22, 1986, in a windspeed of 12 m/s. The numbers on top of the profiles in the lhs of the Figure indicate the particle diameters.

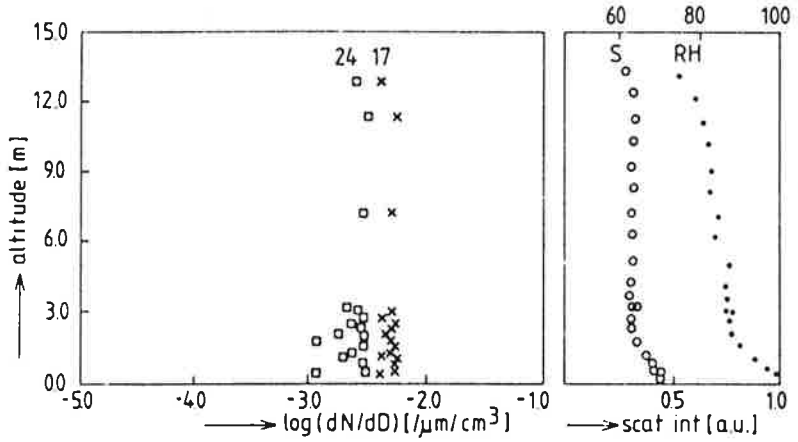

Figure 4. As Figure 3, measured on November 15, 1986, in a windspeed of 7.7 m/s. The curve indicated with RH in the rhs Figure shows the rekative humidity. Note the strong gradients near the surface in both the particle total area concentrations and the relative humidity.

The scatterometer was especially constructed for the HEXMAX experiment, for two purposes. In the first place an instrument was required for quickly profiling of the surface layer in support of the Rotorod measurements. However, due to wave-induced particle-concentration fluctuations (see below) rather long averaging times were required to obtain reliable mean values. The time-consuming data processing of the Rotorod samples is not necessary, however. Therefore the optical scatterometer is a useful instrument that supplements the Rotorod. The advantage of the latter is the size information in the profiles.

Time-dependent measurements.
The second application of the optical scatterometer was the measurement of time-dependent aerosol concentration fluctuations near the air-sea interface. The results reveal the influence of the waves on the particle concentrations, cf. Figure 5. The amplitudes of the concentration

fluctuations decrease with height above the mean surface, but the effects have been observed up to the maximum sampling height of 15 m [De Leeuw, 1988c]. Visual observations, supported by video recordings, indicate that the fluctuations are correlated with the surface displacements caused by wind waves. This is confirmed by the spectral analyses which reveal a maximum energy density in the 0.3-0.6 Hz range, i.e. at higher frequencies than the common wave spectra. This means that the concentration fluctuations are more sensitive to the surface displacements caused by the waves than to the energy in the waves, as expected. The frequencies are height dependent which is probably due to the increase in wind speed with altitude. Alternatively this might be caused by breaking up of the eddies into smaller ones.

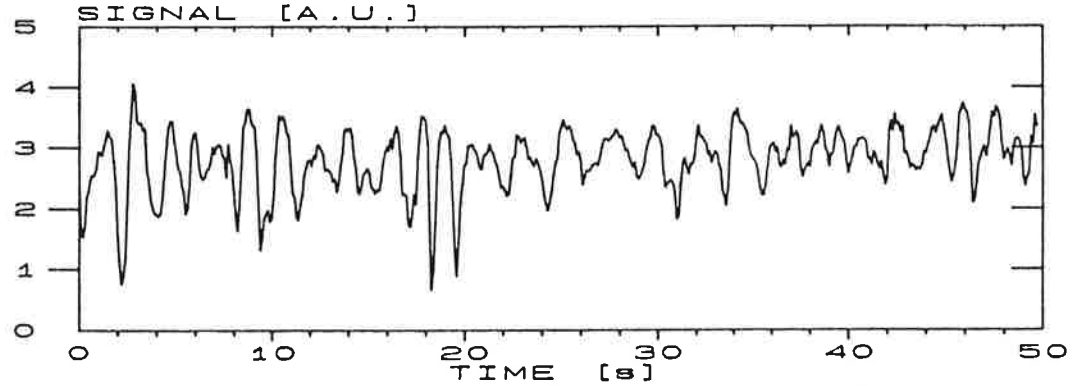

Figure 5. Time-dependent fluctuations of particle total area concentrations, measured at 2.5 m above the instantaneous sea surface with the scatterometer mounted on a waverider buoy. Windspeed was 12.1 m/s. The variations in the scatterometer signal are correlated with the surface displacements.

The time-dependent humidity spectra are not correlated with the surface displacements. Together with the observed wave-induced particle-concentration fluctuations, this indicates that the time scales for the transport of water vapour are much faster than those for particle transport. Due to the fast removal of vapour, the equilibrium between water and vapour is disturbed (cf. Figure 6). Hence the evaporation losses cannot be balanced by condensation. This leads to the conclusion that liquid water is transported form sea to air. As a result, the liquid water flux decreases with height in the surface layer, in contrast with the constant flux assumption that is frequently used in mixed-layer models.

The results are also of importance for modeling of production and dispersal of aerosol particles near the air-sea interface. A theoretical approach has been outlined by De Leeuw [1988c], which may be used to obtain information 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.

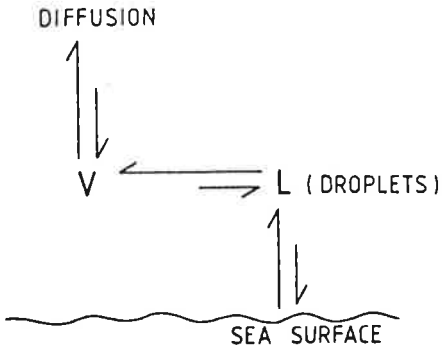

Figure 6.
The dynamic equilibrium between liquid water (L) and vapour (V) shifts due to the fast removal of vapour from the surface layer.

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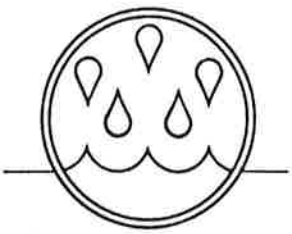
PROCEEDINGS OF THE NATO ADVANCED WORKSHOP

HUMIDITY EXCHANGE OVER THE SEA MAIN EXPERIMENT (HEXMAX) ANALYSIS AND INTERPRETATION

Dellenhove, Epe, The Netherlands

April 25-29, 1988



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HEXOS Contribution No. 16