JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 92, NO. C13, PAGES 14,631–14,635, DECEMBER 15, 1987

Near-Surface Particle Size Distribution Profiles Over the North Sea

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Profiles of particle size distributions near the air-sea interface were measured over the North Sea as part of the Humidity Exchange Over the Sea pilot experiment in 1984. Minima and maxima in the concentration profiles were observed, in agreement with results obtained previously in the North Atlantic. Differences are ascribed to the improved vertical resolution in the present measurements, while the shorter wave period in the North Sea has an influence, too. The results are qualitatively explained in terms of a wave-rotor model.

1. Introduction

Vertical profiles of particle concentrations provide valuable information on particle transport [*Wu*, 1979] and on the contribution of sea spray aerosol to the atmospheric humidity flux [*Smith et al.*, 1983], as well as on surface production rates [*Fairall et al.*, 1983; *Monahan*, 1986] and deposition [*Fairall and Larsen*, 1984]. In spite of these wide fields of applications, minimal information is available on the vertical distribution of particles near the air-sea interface (cf. *Wu* [1979] for a review). Both laboratory studies and oceanic measurements of particle profiles were generally carried out at heights relatively far above the wave tops. These experiments gave rise to the assumption of a logarithmic profile, based on mass concentration measurements made under different conditions [cf. *Blanchard and Woodcock*, 1980]. However, recent laboratory and field experiments, carried out both above the wave tops and in the troughs, revealed the existence of nonlogarithmic concentration profiles near the water surface. These results apply both to particles with sizes of the order of 1 mm (laboratory experiments by *Koga and Toba* [1981]) and to particles in the 10- to 100-μm-diameter range measured over the North Atlantic [*de Leeuw*, 1986*a*]. The oceanic measurements indicate that the minima and maxima in the concentration profiles occur only at wind speeds higher than about 7 m/s. These field experiments were qualitatively explained by a wave-rotor model based on flow separation studies.

To model the particle behavior near the sea surface quantitatively, more experimental data are required under a variety of meteorological and oceanographic conditions. In this paper, results are presented from measurements of vertical profiles of aerosol particle size distributions over the North Sea. The measurements were performed in November 1984 at Meetpost Noordwijk, a platform 9 km from the Dutch coast, as part of the Humidity Exchange Over the Sea (HEXOS) pilot experiment [*Oost et al.*, 1985]. The oceanographic conditions in the North Sea (wave period and height, swell, fetch, sea temperature, etc.), as well as the influences of the European mainland and Great Britain on the atmospheric conditions, might give rise to particle profiles that are different from those in the middle of the North Atlantic.

Particle profile measurements in the heavily polluted North Sea area and models for production and deposition are important to determine the influence of air-sea exchange on the atmospheric cycle and to estimate pollution of the sea due to atmospheric inputs [*Duyzer*, 1985]. Although the major pollutants are in a size range smaller than the particles considered here (>10 μm), the results of the present study lead to the development of a model [*de Leeuw*, 1985*a*] that is expected to apply even better to smaller particles.

Paper number 7C0589.
0148-0227/87/007C-0589$05.00

2. Experiments

Size distributions of particles larger than 10 μm in diameter were measured with a Rotorod inertial impactor. The sampler consists of two polished stainless steel rods, mounted in a retracting collector head on a motor which rotates at a nominal speed of 2400 rpm. The linear velocity of the rods is 10 m/s. Particles impacted on the rods are retained by a sticky coating (silicone). Microscope images of the rods are digitized to determine the particle size distribution by computer. The performance of the Rotorod was discussed in a previous paper [*de Leeuw*, 1986*b*], where also a comparison was made between marine aerosol spectra measured with the Rotorod and data obtained by other sampling methods.

For measurements from just above the sea surface up to 2 m the sampler was attached to a float which followed the waves. The Rotorod was mounted on a 40-cm extension rod to reduce the influence of the float on the measured particle size distributions. Measurements with the float were carried out at heights of 0.12, 0.20, 0.35, 0.50, 0.75, 1.0, 1.5, and 2.0 m above the instantaneous water surface. In addition, measurements at heights from 1 to 21 m were made with the sampler fixed to the platform. Overlapping measurements between 1 and 2 m were made to investigate the difference between the two sampling approaches. The measurements were made at a distance of about 9 m from the legs of the platform, except those at 15 m and higher, which were performed from the decks.

3. Results

Samples were collected at heights varying from 0.12 m in calm sea or from 0.5 m in rough water up to about 21 m. Altogether a total of 121 samples were collected, resulting in eight profiles, measured in wind speeds ranging from 4.1 to 12.4 m/s. Unfortunately, all low-wind profiles were measured in SE winds, in the lee of the platform (samples were taken at the west side). Since these measurements are influenced by the

platform, they are not discussed here. The remaining four profiles were measured in western winds and are representative for undisturbed marine conditions (the influence of the platform is discussed in the next section). Examples of undisturbed profiles, measured in winds of 8.9 and 12.4 m/s, are presented in Figures 1 and 2. Particularly in the high-wind situation (Figure 2), the sample points are rather smoothly distributed to result in concentration profiles with clear maxima and minima.

Particle gradients in the lower 2-m region above the water surface show a definite dependence on both wind speed and particle size. The concentrations of the largest particles (66 μm) decrease with increasing height. In the highest wind of 12.4 m/s the concentrations of smaller particles ($\leq$49 μm) appear to increase, however, from 0.35 m to reach a maximum at 1 m. As wind speed decreases, the sizes of the particles for which positive concentration gradients are observed in the lower 0.5 m become smaller. At 8.9 m/s an increase is observed for particles up to 28 μm, while at 8.1 m/s (not shown) this is only observed for particles of 12 and 15 μm. In the latter case the concentrations of particles of 21 μm are virtually constant up to 0.5 m. It is noted that in the high-wind situation (12.4 m/s), samples were not taken below 0.35 m, which might somewhat distort the comparison with the lower-wind cases. The maximum at 1 m is very pronounced at 12.4 m/s. In lower winds this maximum is less clear. At $u = 8.9$ m/s it has also shifted to a height of 0.75 m. Secondary maxima are observed at heights around 2 m (2–2.3 m at 12.4 m/s and 1.5–2 m at 8.9 m/s).

4. Influence of Platform

A hugh structure, 12 m wide and 7 m high, at about 11 m above the sea surface is likely to distort the flow to an appreciable extent. A wind tunnel study on the flow characteristics of the platform [*Wills*, 1984] led to the conclusion that at 16 m from the rail, accurate measurements can be made (with appropriate corrections). At 10 m, however, flow distortion is so high that only mean velocities can be measured accurately. These results apply to heights between 5 and 17 m.

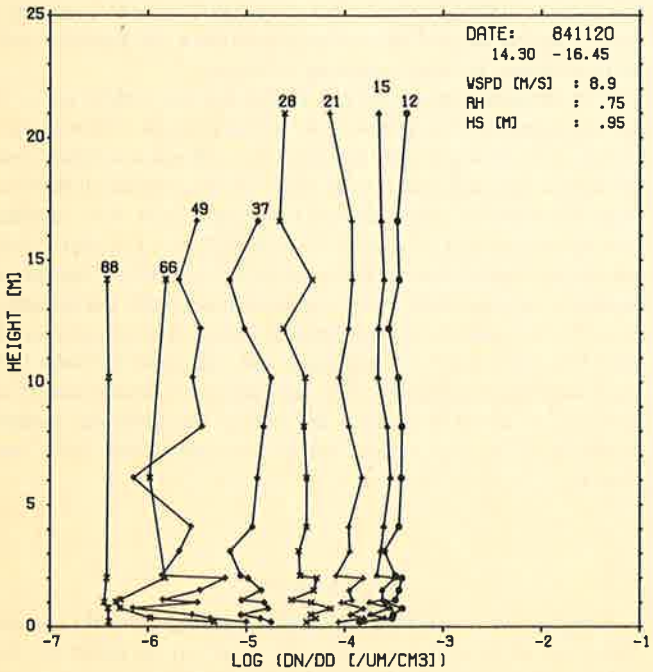

Fig. 1. Vertical variations of particle concentrations, measured on November 20, 1430–1645 UT. Parameter is the particle diameter in micrometers. WSPD is wind speed; RH is relative humidity; HS is significant wave height; DN/DD is number of particles per diameter interval in μm^{-1} cm^{-3}.

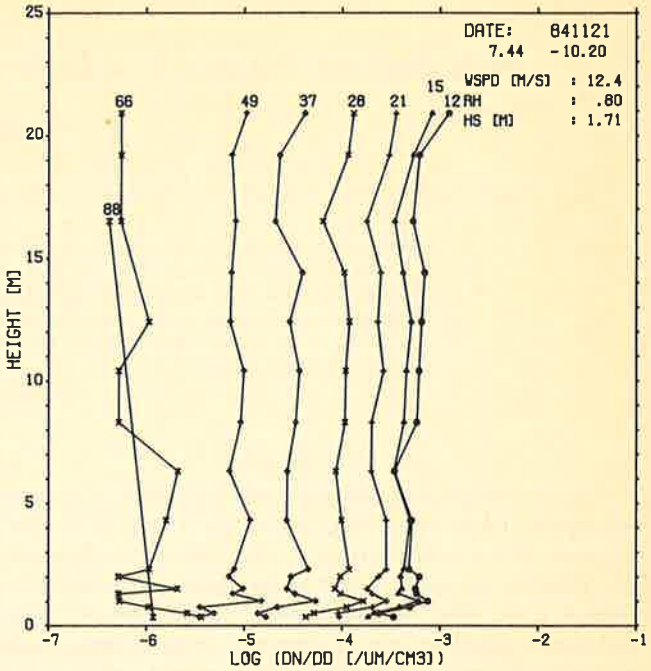

Fig. 2. Vertical variations of particle concentrations, measured on November 21, 0744–1020 UT. Parameter is the particle diameter in micrometers. Abbreviations are defined as in Figure 1.

Our samples were taken at 9 m from the legs of the platform and, at 15 m and higher, from the decks at about 1 m from the rail. Regarding the results from the model study cited above [*Wills*, 1984], particle fluxes at these positions are expected to be influenced by the platform structure. On the other hand, the results in Figures 1 and 2 show that the particle concentrations vary smoothly with height and are not notably affected by the platform.

Near the sea surface, below about 2–3 m height, concentration variations are observed due to wave-induced flow, as explained below. At a distance of 9 m from the windward side of the platform this flow is not likely to be distorted to a large extent, since the platform has a rather open structure between the 10-m-long legs. Vertical flow distortion at 5 m above the surface is 1–2% at 10 m from the platform [*Wills*, 1984].

5. Particle Transport Near the Sea Surface

To explain the results presented in section 3, particle transport and other processes affecting particle size distributions are to be considered. Aerosol transport near the sea surface occurs because of various mechanisms. Particles that are freshly produced at the sea surface have a limited ejection velocity and tend to return to the sea surface because of gravitation. In still air the maximum ejection height is about 18 cm [*Blanchard*, 1963; *Wu*, 1979]. On the other hand, particles are kept in suspension because of turbulence and convective transport. In addition, at high winds flow separation occurs,

causing eddies in the wave troughs, and particles may be temporarily trapped in the circulating airstreams [*de Leeuw*, 1986*a*].

In the atmosphere, particles change in size and weight because of evaporation and condensation of water vapor. This affects particle transport because of the dependence of the various transport properties on particle mass. Also, the particle size distributions change with humidity. Therefore particles traveling through various humidity regimes may be detected in different size bins, which makes the analysis of the vertical variation of particle concentrations rather complex. The effects of the above processes on the variation of particle size distributions with height above the sea surface are discussed below.

Particle Trapping in Eddy Airflow

At a wind speed of about 7 m/s (the onset of whitecapping) and higher, stagnation points come into existence, resulting in flow separation [*Banner and Melville*, 1976]. In a wave-following frame of reference this results in closed streamlines [cf. *Hsu et al.*, 1981]. Particles may be temporarily trapped in such eddies, as described extensively by *de Leeuw* [1986*a*]. Since the present experiments give rise to refinement of the proposed qualitative model, a detailed description of the particle-trapping process is presented below using the (simplified) scheme in Figure 3. Reference is made to a wave-following coordinate system, and velocities are taken positive to the right.

Assuming that the horizontal velocity of a rising air bubble is the same as the wave velocity, the horizontal velocity of a freshly produced particle is zero. Because of the returning airflow in the bottom of the trough the particles gain negative velocity and lag behind the wave. Near the crest, in the lee of the wave, the airflow tends to drag the particles upward. If they get over the wave top, the particles are accelerated by the flow and may gain sufficient velocity to overtake the wave. Near the windward side of the crest the particles are dragged downward by the airstream. Subsequently, they are decelerated by the returning airstream in the wave trough, and the whole process starts all over again.

Because of gravitational settling, relatively high particle concentrations will occur in the periphery of the eddy airstream (compare the calculations by *Maxey and Corrsin* [1986]). As a result, a sampler on a wave follower which integrates the particle concentrations at fixed height above the instantaneous sea surface registers enhanced particle concentrations at heights corresponding to the bottom and the top of the eddy. Consequently, a double maximum is observed in the concentration profile, as in Figures 1 and 2. The lower maximum is more intense, since the particles are forced downward by gravitation. A second explanation for the higher concentrations in the first maximum with respect to the second one may be that the sampler is outside the eddy whenever the float is on top of the waves. Hence at greater heights the mean particle concentrations are lower than the ones registered close to the sea surface. The minimum (which is obviously to occur between the two maxima) will be more pronounced, since a fraction of the freshly produced particles (which in the absence of an eddy might have traveled upward) are now dragged away from the center.

In low winds these effects are revealed only for light particles, since these follow the airstream relatively easily. In higher winds the airflow becomes stronger, and heavier particles may be trapped as well.

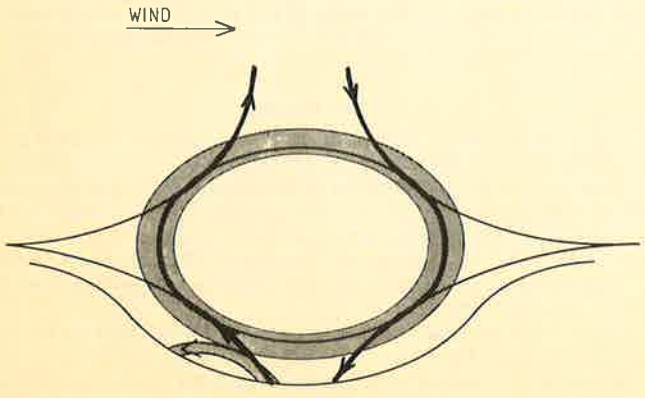

Fig. 3. Schematic representation of the qualitative model of particle transport by eddy airflow, as described in the text (not to scale).

Dispersal of Sea Spray Particles

During the process described above, a fraction of the particles disappear from the eddy, because, owing to inertia, not all particles can follow the air circulation. At the top of the eddy this may result in upward transport into the mixing layer. This process may be assisted by turbulent forces. In the mixing layer, further dispersal may occur because of convection by the wind and by thermal-induced air motions, as well as because of turbulence and gravitational forces [cf. *Fairall et al.*, 1984]).

Deposition of Particles on the Sea Surface

When particles are swept out of the air circulation at the bottom and the sides of the eddy, this will result in deposition on the water surface. However, not only freshly produced particles are injected into the eddy, but also particles are deposited into the circulating airflow from the above mixing layer. These "aged" particles are subjected to the same forces in the eddy as the fresh particles and may be trapped temporarily. For particles of the order of 1 μm, which constitute the major fraction of atmospheric pollutants, this trapping is likely to be more efficient than for the large sea spray particles considered in the present experiments. Hence the trapping mechanism may turn out to be an important process in the removal of atmospheric pollution at the sea surface. Deposition may be enhanced by interactions between the particles trapped in the circulating airflow.

Humidity Effects

In the above discussion, humidity effects were not taken into account. A droplet ejected with the composition of seawater will evaporate until its vapor pressure equals ambient relative humidity. Generally, particles are assumed to attain their equilibrium size instantaneously [*Toba*, 1965; *Goroch et al.*, 1980], although *Chaen* [1973] concluded from his measurements that the particle size can be better described with an enhanced effective relative humidity. *Fairall* [1984] gives a plot of particle humidity response time versus particle radius. Extrapolation of this plot shows that for particles that are of interest for this analysis (10–100 μm in diameter), response times vary from about 0.05 to 5 s. Hence it is reasonable to

assume that the smallest particles are in constant equilibrium with their surroundings, but this certainly does not apply to the larger ones. This is confirmed by wave tank experiments [*Mestayer et al.*, 1985].

With respect to this discussion there is the question of the variation of relative humidity with height. On the sea surface, relative humidity is usually assumed to be 100%, while it decreases strongly in the viscous sublayer. In the adjacent layer, humidity transport is determined by turbulence and diffusion, while vapor pressure gradients depend on thermal stability [cf. *Roll*, 1965]. Under stable stratification, positive gradients have been observed in the bottom layer, with a constant humidity layer above. In the North Atlantic we made some humidity measurements between 0.2 and 2 m with our wave rider buoy. The few results indicated structured humidity profiles similar to the particle concentration profiles [*de Leeuw et al.*, 1984]. Therefore simultaneous measurements are required of particle and humidity profiles to incorporate humidity effects in an analysis of the particle concentration profiles and in a model describing particle dynamics in the vicinity of the sea surface.

6. Comparison With Other Experiments

Previously, measurements of particle concentration profiles were made in the North Atlantic [*de Leeuw*, 1986*a*]. As in the present experiments these data also revealed that at low winds all structure is absent except for a negative gradient very close to the surface due to the limited ejection velocity and the action of gravitation. On the other hand, in the North Atlantic the double maximum was not observed. This may be due to the low vertical resolution in these measurements. Alternatively, the longer wave periods in the North Atlantic (about 10 s as opposed to wave periods of about 3 s in the North Sea) may be responsible for this difference. Also, the strong tidal currents in the North Sea may be of influence. More measurements in the North Atlantic, with improved vertical resolution, are required to decide on the origin of the mentioned differences.

Other data on vertical profiles of particle concentrations near the sea surface are not available from the literature. A comparison of the North Atlantic data with results from the few field measurements on profiles higher above the surface and with laboratory experiments was presented previously [*de Leeuw*, 1986*b*].

7. Discussion and Conclusions

The observed vertical variation of particle size distributions has been explained qualitatively in terms of a wave-rotor model. Because of the temporary trapping of particles in eddy airflow, the particle concentration profiles show maxima at heights corresponding with the most likely particle trajectories in the periphery of the eddy.

This vertical variation of particle concentrations will be more clearly observed when wind speed, and thus the number of eddies, increases. This explains the more distinct structure observed in the concentration profiles on November 21 ($u = 12.4$ m/s).

The influence of turbulence (as distinguished here from the eddy airstream by its random structure) has only briefly been indicated in the proposed qualitative model, although obviously this will contribute to vertical transport of sea spray particles. Numerical calculations are required to decide on the relative importance of both mechanisms (turbulence versus wave-rotor) for particle transport near the sea surface and to explain quantitatively the observed particle profiles.

Humidity effects could not be considered in the above analysis because of the lack of experimental data on the vertical variation of humidity near the sea surface. In a numerical model these effects can be introduced for comparison with results from future experiments.

The similarity with respect to the upper maximum in the profiles shown in Figures 1 and 2 is somewhat surprising, since wave heights differ by a factor of 2. From the measurements in the North Atlantic it was concluded that the height at which maximum concentrations occur depends on wind speed and/or wave height. The present results reveal that wind speed will probably be the most important parameter that influences both production rate and vertical mixing. Through the dependence of turbulence intensity on wind speed [cf. *Werner et al.*, 1985] there is an influence of wind speed on the variation in the vertical structure of the particle concentrations as well. Apparently, the height of wind-induced waves is not of major influence. On the other hand, the scale of the wave-rotor mechanism will be determined by wave height and swell.

With respect to the assumed model, different results are expected from measurements at fixed height and from measurements with a wave follower. However, no abnormal transitions in the particle profiles were observed in the overlapping region between the two sampling approaches. Obviously, this is due to the integration over many waves, resulting in similar data from both approaches. Faster sampling methods are being considered for future experiments to investigate temporal behavior.

The processes governing the vertical variations of particle concentrations are the subject of continued investigations. Parallel field measurements of profiles of particle size distributions, relative humidity, and small-particle scattering are in preparation to achieve better insight into the generation, removal, and vertical transport of aerosol particles near the sea surface. In addition, information on other air-sea interaction processes can be obtained from such experiments, such as humidity exchange or the interaction between wind and waves, as can the scale heights for these processes. The present results indicate that the scale height for particle concentration profiles is 1–2 times the wave height.

Acknowledgments. The author wishes to acknowledge the stimulating insights of C. W. Lamberts and the practical assistance of M. M. Moerman and P. Piket in the preparation of the measurements and the processing of the particle data. HEXOS publication 3.

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(Received October 15, 1986;
accepted May 27, 1987.)