Papers

Aerosol masses and mass gradients in the marine boundary layer over the breaker zone^{*}

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Abstract

The aerosol size distribution function and aerosol concentration in the marine boundary layer depends strongly on wind speed, direction and duration. Measurements were therefore carried out in various seasons of the year, with all the above factors and different types of sea bottoms being taken into consideration. By employing the size distribution function and concentrations of aerosol particles, which were determined in the coastal zones of the southern Baltic Sea by means of the lidar method, the aerosol masses and their gradients in the marine boundary layer were calculated. It was found that for onshore winds the concentrations and gradients were substantially lower when compared with those for winds blowing off the land.

1. Introduction

In coastal areas, aerosols in the zone of direct interaction between the air and the ocean surface are characterised by rapid time-space changes of concentrations (Dovgolyuk and Ivlev, 1977; Nielson, 1979). The continual

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changes in aerosol concentrations are due both to external changes, such as variations in particle input, and internal changes like coalescence, coagulation and evaporation (Kaufman, 1993). These processes result in variations in aerosol sizes, which also influences particle removal from the marine boundary layer (Bertrand *et al.*, 1974; Kondratev *et al.*, 1974). This means that the changes in aerosol concentration, and thus in mass, in the marine boundary layer are concerned with the changes in the spectrum of marine aerosol size distribution functions. This spectrum is complex and depends strongly on weather conditions in the marine boundary layer (Doran *et al.*, 1995). The shape of the size distribution function depends primarily on wind speed, duration and direction, and additionally on relative humidity (Van Eijk and de Leeuw, 1992).

Active remote methods can be employed to investigate short-term changes in marine aerosol dynamics (Zieliński and Zieliński, 1994). The aerosol size distribution function can be accurately obtained from lidar backscattered data. Marine aerosol particles are particularly well suited for such research: their scattering characteristics can be represented by the Mie theory as the particles are almost spherical and their complex index of refraction is well known (Piskozub, 1994).

2. Experimental site and instrumentation

Measurements have been made since 1992 at several stations on the Polish coast of the Baltic Sea (17–19°E and 54–55°N) (Zieliński, 1995). Their locations are shown in Fig. 1. The measurements were carried out in different seasons in order to detect the influence of the type of wind on the level of aerosol mass in the coastal zone. Therefore, wind speed and direction as well as wet and dry-bulb temperatures were recorded, in addition to other supporting data. The air temperature ranged from 278 to 293°K, the wind



Fig. 1. Location of measuring stations

speed from 0 to 19 m s⁻¹ and the wind direction from NE to SW (Zieliński and Zieliński, 1994, 1996).

The FLS–12 lidar system was installed in a van and stationed on the top of the dunes at a fixed distance from the sea. The inclination of the lidar was readily changed, which enabled the marine boundary layer to be sounded at various altitudes.

The FLS-12 is a tunable laser system designed for remote sensing of the air in the VIS spectral range (400–670 nm). The source of UV pumping for the dye laser is a XeCl excimer laser (308 nm). The backscattered energy from various distances is collected by a Cassegranian configured telescope, which has a 280 mm diameter primary mirror and is registered by the separate channels of a multichannel (8 channels) photoreceiver. A detailed description of the FLS-12 lidar is given elsewhere (Zieliński *et al.*, 1994). During the experiments the lidar collected aerosol backscattered data at wavelengths in the range from 440 to 670 nm. The lidar measurements were calibrated by simultaneous measurements with six stage cascade impactors and a laser particle counter (PMS – Particle Measurement System).

3. Theory

For the calculations, the procedure for deriving the size distribution function and aerosol concentration described in Zieliński *et al.* (1994) was employed to determine aerosol concentration and mass, as well as mass gradients in the marine boundary layer over the breaker zone. The ensemble of aerosols was assumed to consist of non-absorbing spherical water droplets, and that the total geometric cross-section of aerosols has a form corresponding to the power law for size distribution. Three different sounding-light wavelengths (444, 567 and 665 nm) were used in the computations.

The concentration of particles of sizes ranging from r to $r + \Delta r$ in unit volume is described as follows:

$$\Delta N = N_c e^{-br} (1 - e^{-b\Delta r}), \tag{1}$$

where

$$N_c(z_i, h_i) = \int_{0.5}^r n_r dr = \frac{a(z_i, h_i)}{b(z_i, h_i)} e^{-0.5b(z_i, h_i)},$$
(2)

is the normalised total concentration of aerosol particles at an arbitrary distance z and altitude h for particles in the 0.5 μ m to 5 μ m radius size range, and a, b – distribution parameters.

By assuming that Δr is small and dividing eq. (1) by this factor, the following equation describing the gradient of the number of the particles in unit volume with size variations is obtained:

$$\frac{dN}{dr} \approx \frac{\Delta N}{\Delta r} = \frac{N_c}{\Delta r} e^{-br} (1 - e^{-b\Delta r}).$$
(3)

On the basis of concentrations of the aerosol measured and employing eqs. (1) and (2), the total mass of aerosols for the particle range $r \in [0.5, 5 \ \mu m]$ can be expressed by

$$M_c = 4\pi\rho \frac{a}{b} \int r^2 e^{-br} dr,\tag{4}$$

where

r – particle radius,

 ρ – particle density.

Aerosol mass gradients related to the mean aerosol concentration can be described by

$$\frac{dM}{dr} = 4\pi\rho \int_{r+\Delta r}^{r} r^2 \frac{dN}{dr} dr.$$
(5)

4. Results

Eqs. (3) and (5) enable the gradients to be determined by employing experimentally obtained mean aerosol concentrations and masses in unit volume of the marine boundary layer. It was shown that values of distribution parameters b varied with wind speed; nevertheless, the best



Fig. 2. Dependence of aerosol mass on wind speed and direction

results were obtained for $b = 2 \text{ m}^{-1}$. Fig. 2 shows the dependence of aerosol mass on wind speed and direction: aerosol mass increases with rising wind speed. However, for southerly winds these masses are higher when compared with northerly winds of the same or similar speed. For both wind directions, situations occur where aerosol masses vary, even though the wind speed is the same. Such a distribution of data is due to the fact that these results refer to situations when winds blew at the same speed for a different length of time – from ca 48–75 to a few hours. For northerly winds (*i.e.* onshore) the aerosol ensemble comprised mainly marigenic particles, while for southerly winds (*i.e.* offshore) the major contribution was from terrigenous particles. Additionally, it was found that marine aerosol masses varied with offshore distance over the breaker zone and that there were distinct maxima associated with the higher production in this area as a result of breaking waves. In the case of southerly winds no very insignificant changes in aerosol masses were detected with respect to offshore distance over the breaker zone (Fig. 3).



Fig. 3. Variations in aerosol mass with offshore distance for winds from two directions: northerly winds (a), southerly winds (b)

In Fig. 3b it is practically impossible to determine any variations in the value of the mass over the measuring distance for southerly winds. Offshore winds blew for up to 35 hours at a time, and the marine boundary layer was therefore well mixed. Such winds carried a significant amount of pollution, thus giving rise to larger masses than those carried by northerly, onshore



Fig. 4. Dependence of aerosol mass gradients on wind speed and direction: northerly winds (a), southerly winds (b)

winds, regardless of the fact that northerly wind speeds are higher than those of southerly winds. However, for northerly winds (Fig. 3a) it is possible to determine the limits of the breaker zone (from ≈ 40 m to ≈ 130 m). This is due to the production of marine aerosols as a result of breaking waves, which are significantly higher in this area than beyond it. Fig. 4 shows the changes in the mass gradients of aerosols for two kinds of winds derived from eq. (5). For the size range $r \in [0.5, 5 \ \mu m]$ for both kinds of wind, aerosol mass gradients increase with increasing wind speed. For offshore winds the values are higher when compared with those for onshore winds. In both cases, aerosol mass gradients decrease with increasing particle size due to the increase in deposition velocity, and are higher for the former than for the latter. However, for both wind directions there are maximum mass gradients that occur for particles smaller than 0.5 μ m. The maxima were found to vary with wind speed. For winds of speeds lower than 2 m s^{-1} , the maximum radius $r_{\rm max} \approx 0.4 \ \mu {\rm m}$ and for wind speeds over 7 m s⁻¹, $r_{\rm max} \approx 0.1 \ \mu {\rm m}$. Similar results were reported by Hoppel et al., (1985). Such a result means that in both cases, the aerosol size distribution function and masses in the marine boundary layer varied over the coastal zone.

Fig. 5 shows the dependence of aerosol mass gradients on wind speed and direction with sensing distance. Like the situation shown in Fig. 3, it is possible to determine the limits of the breaker zone in the case of northerly



Fig. 5. Variations in aerosol mass gradients with offshore distance for winds from two directions: northerly winds (a), southerly winds (b) winds (Fig. 5a), something which is impossible to do where southerly winds are concerned (Fig. 5b). Furthermore, aerosol mass gradients are higher for

southerly winds than those for northerly winds. Finally, vertical aerosol mass gradients were determined for winds of various speeds and directions. The results are shown in Fig. 6. These



Fig. 6. Variations in aerosol mass gradients with altitude for winds from two directions

gradients steepen with altitude above sea level and with decreasing wind speed. This confirms the fact that over the breaker zone with increasing altitude above the sea surface, the aerosol mass decreases and is strongly dependent on the wind speed. The aerosol mass is again higher in the case of southerly winds when compared with the results obtained for northerly winds. It was found that for offshore winds, aerosols greater than 4 μ m in radius were undetectable at altitudes around 40 m, while for onshore winds this altitude was 80 m. As the wind speed increases in unit volume at a given altitude in the marine boundary layer, so does the number of large particles. This is due to the increase in the deposition velocity of large particles that fall from higher layers of the atmosphere (Zieliński and Zieliński, 1996). Such particles are also entrained from lower parts of the atmosphere owing to the vertical component of wind speed and turbulent diffusion.

5. Conclusions

The aerosol size distribution function and aerosol mass in the marine boundary layer over the breaker zones of the southern Baltic Sea depend on wind speed, direction and duration. The lidar-based results obtained for the size distribution are comparable to those obtained by Smith et al. (1991), Jennings et al. (1991) and Rongshuo et al. (1991). It can therefore be assumed for northerly winds that the particles present in the marine boundary layer above the breaker zones of the southern Baltic Sea are marine aerosols. In other cases, especially when winds are southerly, the particles are a mixture of marine aerosols and particles of terrestrial origin. With southerly winds the aerosol concentrations, masses and mass gradients in the marine boundary layer above the breaker zones are significantly higher than is the case with northerly winds. Moreover, they do not differ substantially with offshore distance, even though the wind speeds are lower for southerly winds. It was found that with northerly winds the breaker zone was readily distinguishable from aerosol concentrations, masses or fluxes, which were higher when compared with those from the open sea. With southerly winds it is not as easy to determine the breaker zone range because the mean aerosol concentration is constant along the sounding path.

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