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Modelling of the aerosol size distribution over the Baltic Sea

Marine aerosol Baltic Sea Aerosol size distribution Empirical orthogonal function

> Aérosol marin Mer Baltique Répartition en taille des aérosols Fonctions empiriques orthogonales

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The results of applying empirical orthogonal functions (EOF) to decomposition and approximation of aerosol size spectral distribution are presented. The experimental measurements were done during two research cruises in the Baltic Sea and during a coastal experiment in Lubiatowo in 1994. The main finding is that aerosol size distribution can be presented as a superposition of a slowly varying background of dust-like particles and a further distribution of sea-spraysized particles multiplied by amplitude functions reflecting the contribution of marine aerosols. The amplitude is dependent on wind speed, wind direction and fetch. Decrease in aerosol size distribution amplitude with increasing wind speed was found when air fetch over the sea exceeded 400 km. The opposite tendency of increase in aerosol size distribution amplitude was found when air fetch over the water was not longer than 50 km.

RÉSUMÉ

ABSTRACT

Modélisation de la répartition en taille des aérosols sur la mer Baltique.

La répartition spectrale de la taille des aérosols au-dessus de la mer Baltique a été analysée à l'aide des fonctions empiriques orthogonales (EOF). Les mesures ont été effectuées en 1994 au cours de deux campagnes à la mer et pendant une expérience sur la côte de Lubiatowo. La répartition en taille des aérosols se présente comme la superposition de deux spectres : celui, fondamental et lentement variable, de particules de type poussières, et celui des embruns, multipliée par des fonctions d'amplitude reflétant la contribution des aérosols marins. L'amplitude dépend de la vitesse et de la direction du vent, ainsi que de la distance sur laquelle il agit. L'amplitude diminue lorsque la vitesse du vent augmente et lorsque le fetch dépasse 400 km. A l'inverse, l'amplitude augmente lorsque le fetch reste inférieur à 50 km.

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INTRODUCTION

Investigations of marine aerosols by field and theoretical methods have been intensified since the 1950s, when Kientzler *et al.* (1954) completed a photographic study on the projection of droplets by bubble bursting at a water surface. Subsequently, many other investigations conducted, for example, by Blanchard and Woodcock

(1957), Monahan *et al.* (1982), Garbalewski and Marks (1985), Bowyer (1986), Cipriano *et al.* (1987), de Leeuw (1987), Stramska *et al.* (1990), and Marks (1990), have enriched our knowledge of marine aerosol generation and transport processes.

One of the recently studied processes relates to the influence of atmospheric aerosols on the radiative balance

of the atmosphere. Aerosol particles and, in particular, the marine aerosol affect the Earth's radiative transmission, both directly through the backscatter of radiation and indirectly as cloud condensation nuclei. Among the optical properties, the extinction coefficient of the atmosphere over the different sea regions is one of the important factors affecting the Earth's radiative balance.

Preliminary findings on the influence of sea-salt aerosol concentration and size distribution on the extinction coefficient of the atmosphere over the Baltic Sea were presented in the paper by Kuśmierczyk- Michulec (1994), where two aerosol size distributions, both represented by the modified gamma function, were calculated on the basis of sea-salt aerosol experimental data derived from measurements by six stage impactors.

In this study, not only the cases where relatively clean marine aerosols are present but also the influences of continental aerosols are investigated.

This article presents the results of applying the empirical orthogonal functions (EOF) method (Lorentz, 1956; Nielsen, 1979; Preisendorfer, 1988; Jankowski, 1994) to decomposition and approximation of aerosol size distributions. In particular, the intention is to demonstrate the application of this method to the modelling of aerosol size distributions over the Baltic Sea.

MEASUREMENTS

The experimental data were collected during two research cruises in the Baltic Sea and during a coastal experiment in Lubiatowo. The first cruise took place on board s/y "*Pogoria*" on 14-21 April 1994, followed by a cruise on board r/v "*Oceania*" on 7-15 May 1994. The Polish Academy of Science Laboratory in Lubiatowo was chosen for the coastal experiment performed on 19-29 September 1994, as a part of the Baltic Aerosol Experiment (BAEX).

Recorded data consisted of continuous marine aerosol concentration and size distribution data, together with the set of meteorology data. Aerosol observations within sixty size ranges from $0.25 \,\mu\text{m}$ to $23.5 \,\mu\text{m}$ in radius were recorded using PMS Classical Scattering Aerosol Spectrometer Probe, Model CSASP-100-HV. A He-Ne high-order multimode laser probe of 5 mW was mounted at 5 m elevation above the water surface, always on the windward side on a ship. During the coastal experiment, however, the probe was mounted on the beach at 2 m elevation above the ground at a distance of about 20 m from the water line. The air flow through the probe was 32.8 m s⁻¹. Both communication with the probe and data assembly were provided by PC computer and developed software.

Meteorological data were collected every three hours and included wind speed and direction, air temperature and humidity, atmospheric pressure, precipitation and current weather observations.

RESULTS

In order to analyse the spatial and temporal variability of aerosol size spectral distributions and to extract their main components, the method of empirical orthogonal functions (EOF) was applied.

Firstly, all aerosol size distributions were normalized to RII = 80% according to Fitzgerald (1975), and among all experimental distributions only data corresponding to relative humidities not greater than 85% were chosen. Data collected at higher humidities as well as under rainy conditions were not analysed. Secondly, the calculated empirical orthogonal functions were used to analyse the aerosol size distributions n(r).

Results of the EOF method show that all spectra may be defined by means of one general equation, taking into account only the first mode h_1 , because the contribution of the first eigenvalue \wp_1 to the total variance is 95.34%. Such a large value makes it feasible to neglect the remaining modes (Nielsen, 1979; Wróblewski, 1986; Jankowski, 1994) and write $n_i(r_j)$ as:

$$n_i(r_j) = \langle n(r_j) \rangle + h_1(r_j)\beta_{i1}$$

$$j = 1, \dots, 15; \quad i = 1, \dots, 32;$$
(1)

where the subscript *i* symbolizes each successive measurement, the subscript *j* corresponds to the radius ranges (r_j is the mean value for each 0.25 µm singlewindow width, within 0.25 - 4.0 µm radius ranges). A total of 32 successive profiles were averaged in two hours and refer to almost stable conditions of wind and humidity. In order to interpret the components of the equation 1 and to obtain the positive fluctuations of aerosol concentration, the above expression can be written in the following form:

$$n_{i}(r_{j}) = \langle n(r_{j}) \rangle' + h_{i}(r_{j})\beta'_{i1}$$

j = 1,...,15; i = 1,...,32; (2)

where the transformed mean value $\langle n(r_i) \rangle'$ is defined by:

$$\langle n(r_j) \rangle' = \langle n(r_j) \rangle + h_l(r_j) \beta_{minl}$$
 (2a)

and the positive fluctuations β'_{i1} are:

$$\beta_{i1}' = \beta_{i1} - \beta_{min1} \tag{2b}$$

Figure 1 presents two functions: one is plotted based on the normalized size distribution given for the dust-like particles; the term "dust-like" refers to the portion of collected aerosol which is insoluble in water (International Association, 1984), the second function is the normalized size distribution calculated for the basic spectral distribution $\langle n(r_j) \rangle'$ (see, eq. 2a). The total number of particles is defined in terms of the final extinction coefficient normalized to unity at 0.55 µm. It appears that $\langle n(r_j) \rangle'$ might be interpreted as the slowly varying continental background. This conclusion may be regarded as a confirmation of the experimental results, presented in Van Eijk and de Leeuw, 1992.

Figure 2 again presents two functions: one is the normalized size distribution for the sea-spray particles (particles generated at the sea surface by the action of the wind; they consist of 30% sea salt and 70% liquid water); the second is the normalized size distribution calculated for the mode 1. The total number of particles is defined in terms of the





Comparison of normalized size distribution for dust-like particles insoluble in water (dashed line), with particles determined by the spectral distribution $\langle n(r) \rangle'$ (points).

final extinction coefficient normalized to unity at 0.55 μ m. The dashed line describes the model function (International Association, 1984), assuming the mean radius of particle $\mathbf{r} = 0.3 \ \mu$ m. Because the amplitudes β'_{i1} (see eq. 2b) have positive values, then $h_1(r)\beta'_{i1}$ might be interpreted as an additional aerosol source (sea-spray particles).

The initial analysis shows that the amplitude function β'_{i1} strongly depends on wind speed and direction. To study the influence of these two factors on the amplitude of aerosol size distribution over the Baltic Sea, all collected data were separated into two classes. These classes were established on the basis of wind direction and 1000 mbar air mass





Normalized size distribution for sea-spray particles (dashed line) and for particle size distribution determined by mode 1 (points)

trajectories data. One of them included air advected from the NE - ENE wind sector with air fetch over the water exceeding 400 km; the second related to the WSW - NW wind sector with air fetch over the water not longer than about 50 km, or alongshore advected air in the case of coastal data. The amplitude function associated with the NE - ENE sector was found to decrease with wind speed (*see* Fig. 3*a*). The linear curve fitted to the amplitude data showed good correlation with wind speed as reflected by the regression coefficient of 0.95. Then, the amplitude β'_{i1} due to the data from NE - ENE wind direction may be written as a function of wind speed v

$$\beta_{i1}' = (-27.5v + 86.8)10^4 \tag{3}$$





Amplitudes of aerosol size distribution β'_{i1} plotted against wind speed: a) for NE - ENE wind sector, b) for WSW - NW wind sector.

From the results presented in Figure 3a, it may be concluded that for significant air fetch over the water with increasing wind speed, the amplitude reflecting the contribution of the sea-spray particle production to the total aerosol size distribution decreases. Similar results were reported by Van Ejik and de Leeuw, 1992. The same aerosol amplitude function as associated with the WSW - NW wind sector was found to increase with wind speed (*see* Fig. 3b). Although only ten samples were collected within that wind sector, nevertheless the distribution of points and fitted curve showed relatively good correlation with wind speed as manifested by a regression coefficient of 0.94. The amplitude function for the wind direction WSW - NW may be written as

$$\beta_{i1}' = (22.08v + 15.5)10^4 \tag{4}$$

In this case, the air had to travel over the sea for only a distance of about 50 km. The wind wave field is not in balance with the wind speed, so the amplitude reflecting the contribution of sea-spray particles from breaking waves is high. On the other hand, it is known that with increasing wind speed the production of sea-

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de Leeuw G. (1987). Near-Surface Particle Size Distribution Profiles Over the North Sea. J. Geophys. Res. 92, 14,631-14,635. salt particles also increases (Lovett, 1978; Monahan et al., 1983).

CONCLUSIONS

Amplitude analysis of aerosol size distribution can be useful in studying aerosol transfer processes. The results showed a significant decrease of the aerosol size distribution amplitude with increasing wind speed when the air had to travel more than 400 km distance over the sea. A contrary increase in aerosol size distribution amplitude with increasing wind speed was found for mixed land/marine air when air fetch over the water was not longer than about 50 km. Further application of this method for marine aerosol investigations is currently in progress.

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