Papers

Modelling of the aerosol size distribution and aerosol optical properties over the Baltic Sea*

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Abstract

Some applications of the method of modelling the aerosol size spectral distribution are presented. The single scattering albedo of the aerosol is calculated on the basis of particle counter measurements and the results of the empirical orthogonal functions method. The experimental measurements were done during two research cruises on the Baltic Sea and during two coastal experiments at Lubiatowo in 1994 and 1995. The results confirmed that the aerosol size distribution can be represented by a slowly varying background of dust-like particles superimposed on the distribution of marine particles, multiplied by amplitude functions reflecting the latter's contribution. The amplitude functions were found to be dependent on the speed and direction of the wind.

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1. Introduction

The early interest in marine aerosol formation (Blanchard and Woodcock, 1957) has expanded to include the role of marigenic droplets on radiative forcing and climate changes. Therefore, one of the principal aims of this and other field studies is to provide the necessary data base and to incorporate aerosols into global atmospheric radiative models. To understand the complexity of marine aerosol systems, more interdisciplinary physical, chemical and bio-aerosol observations are needed. These should include aerosol experiments conducted over both oceans and regional seas. An example of such interdisciplinary investigations into the marine aerosol was the Baltic Aerosol Experiment (BAEX). That series of process studies focused on pollution fractionation in coastal waters and in particular on their water-to-air transfer (Petelski *et al.*, 1993). The coastal environment of the southern Baltic Sea provides a good opportunity to study both marine and terrestrial aerosol systems, including their radiative properties.

In order to quantify the perturbations of the marine aerosol size distribution caused by dust or anthropogenic influences, the empirical orthogonal functions (EOF) method (Lorenz, 1956; Nielsen, 1979; Preisendorfer, 1988; Jankowski, 1994) was applied to the resolution and approximation of aerosol size distributions. The aim of the present paper is to develop further the modelling of the aerosol size distribution over the Baltic Sea, the preliminary results of which were presented in Kuśmierczyk-Michulec and Marks (in press).

The intention of this paper is to extend the possible applications of the EOF method to include an indirect determination of the extinction and scattering coefficients, and of the single scattering albedo of the aerosol.

2. Measurements

The experimental data were collected during two coastal experiments on the southern Baltic Sea coast and two research cruises at sea. Fig. 1 shows the location of the measurement stations. The first cruise took place on board s/y 'Pogoria' from 14 to 21 April 1994, and was followed by a cruise on board r/v 'Oceania' from 7 to 15 May 1994. The Polish Academy of Sciences Laboratory at Lubiatowo was chosen for the coastal experiments performed within the framework of the Baltic Aerosol Experiment (BAEX) from 19 to 29 September 1994 and from 9 to 29 September 1995.

The data collected consisted of a continual record of marine aerosol concentrations and size distributions, along with a complete set of meteorological data. The aerosol observations within sixty radius ranges from 0.25 μm to 23.5 μm were recorded using a CSASP–100–HV PMS Classical

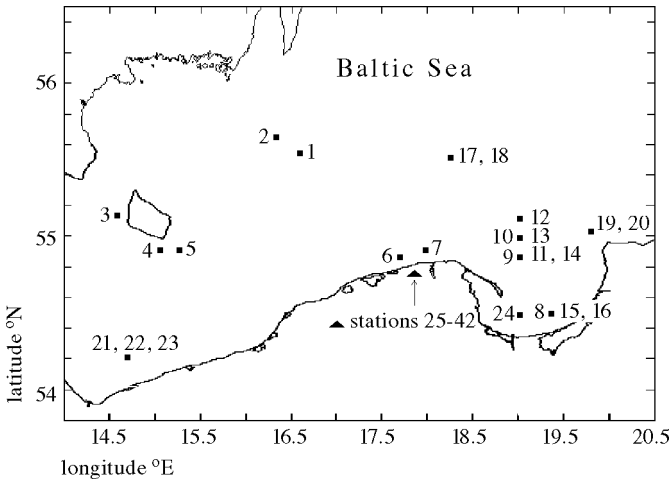

Fig. 1. Location of measurement stations (numbered)

Scattering Aerosol Spectrometer Probe. This aerosol particle counter (He-Ne high-order multimode laser probe of 5 mW) was mounted at a height of 5 m above the water surface, always on the windward side of the ship. During the coastal experiment, however, the probe was mounted on the beach 2 m 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}. Communication with both probe and data assembly was provided by a PC computer and advanced software. General information on the PMS probe used is listed in Tab. 1.

Table 1. General information on the CSASP–100–HV PMS spectrometer probe

Range No.	Radius range [μm]	Number of channels	Width of single channel [μm]
0	1.0 – 23.5	15	1.5
1	1.0 – 16.0	15	1.0
2	0.5 – 8.0	15	0.5
3	0.25 – 4.0	15	0.25

Most of the meteorological data were collected continually by the OMEGA WMS–14 meteorological station and included wind speed and direction, air temperature and humidity, atmospheric pressure, and precipitation. In addition, current weather observations were noted every three hours.

3. Extinction, scattering and single scattering albedo

The single scattering albedo is the fraction of the energy removed from the incident wave reappearing as scattered radiation. According to Twomey (1977), the single scattering albedo of an aerosol is given by

$$\omega_A(\lambda) = \frac{b(\lambda)}{c(\lambda)}, \tag{1}$$

and the total extinction coefficient $c(\lambda)$ by

$$c(\lambda) = \pi \int_{r_1}^{r_2} r^2 Q_{ex} n(r) dr, \tag{2}$$

where r is the particle radius, λ is the wavelength, and Q_{ex} is the extinction efficiency factor.

Analogously, the total scattering coefficient $b(\lambda)$ is given by

$$b(\lambda) = \pi \int_{r_1}^{r_2} r^2 Q_{sc} n(r) dr, \tag{3}$$

where Q_{sc} is the scattering efficiency factor. The efficiency factors Q_{sc} and Q_{ex} can be obtained by means of the model for calculating the respective Mie coefficients for scattering and absorbing spherical particles (Król, 1984, 1985).

4. Results

Initially, all aerosol size distribution data were normalised to $RH = 80\%$ (Fitzgerald, 1975), although in later calculations only the data corresponding to wind speeds not exeeding 6 m s^{-1} and relative humidities no higher than 85% were chosen. The aerosol size distributions $n(r)$ were subsequently analysed with the use of the computed EOFs.

The results of the EOF method show that all distributions are definable by means of one general equation (A10). This takes into account only the first mode h_1 ($l = 1$), because the contribution of the first eigenvalue $\wp_1$ to the total variance is 95.54% :$\aleph(1) = \imath_1 = 0.95$. Since this value is so large, the remaining modes ($l = 2$,..., 15) can be neglected (Nielsen, 1979; Wróblewski, 1986; Jankowski, 1994) and eq. (A10) can be rewritten thus:

$$n_i(r_j) = h_1(r_j)\beta_{i1} + < n(r_j) > \qquad j = 1, ..., 15; \;\; i = 1, ..., 42. \tag{4}$$

The subscript i (see Appendix) stands for each successive measurement $i = 1, ..., N$, the subscript j for the number of spectral channels $j = 1, ..., M$.

Fig. 2 presents the amplitudes β_{il} for the first three modes ($l = 1, 2, 3$) at each measurement station. It is evident that the amplitude of the first mode reflects nearly all the differences between the aerosol size distributions at the various measurement stations. The small second and third mode amplitudes can be regarded as disturbances and neglected.

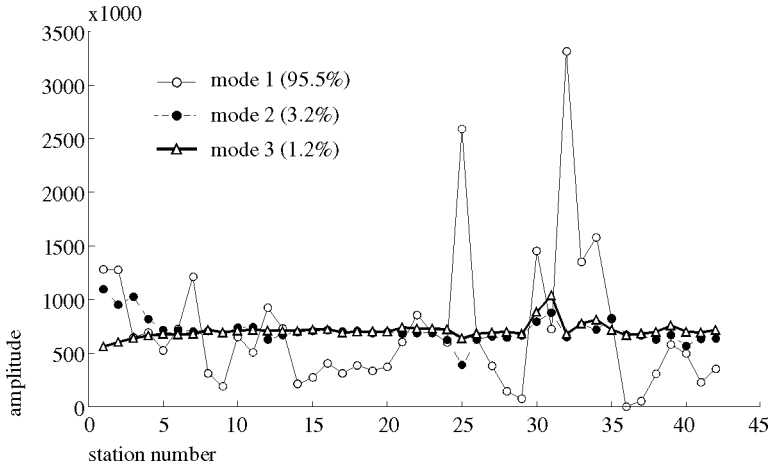

Fig. 2. Amplitudes β_{il} for the first three modes ($l = 1, 2, 3$) for each consecutive measurement station; '×1000' indicates that the amplitude values should be multiplied by 1000

In order to interpret the components of eq. (4), it was rewritten in the following form:

$$n_i(r_j) = < n(r_j) >' + h_1(r_j)\beta'_{i1} \qquad j = 1, ..., 15; \;\; i = 1, ..., 42, \tag{5}$$

where

$$< n(r_j) >' = < n(r_j) > + h_1(r_j)\beta_{\min 1}. \tag{5a}$$

$$\beta'_{i1} = \beta_{i1} - \beta_{\min 1}. \tag{5b}$$

Fig. 3 presents two functions: one is plotted on the basis of the normalised size distribution of dust-like particles (the term 'dust-like' refers to the water-insoluble portion of the aerosol collected (International Association..., 1984)), the other is the normalised size distribution calculated for the basic spectral distribution $< n(r_j) >'$ (see eq. (5a)). The total number of particles is defined in terms of the final extinction coefficient normalised to unity at 0.55 μm. It seems that $< n(r) >'$ could be interpreted as a slowly varying background of terrestrial aerosol. This conclusion is confirmation of the experimental results presented in Van Ejik and de Leeuw (1992).

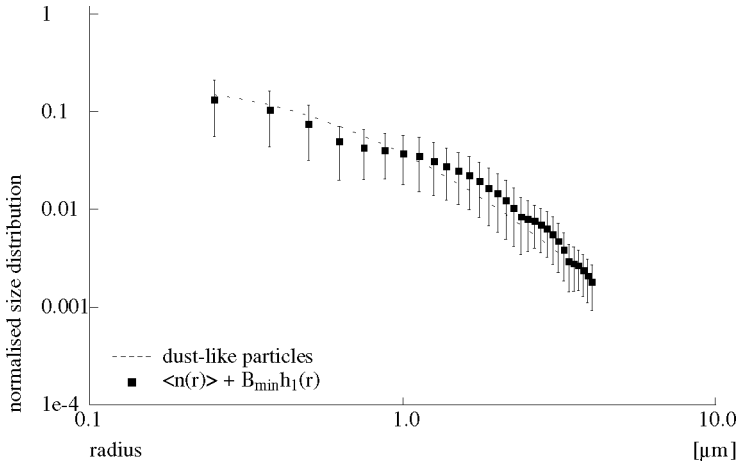

Fig. 3. Comparison of two normalised size distributions for dust-like particles (dashed line) and particles determined by $< n(r) >'$ (points)

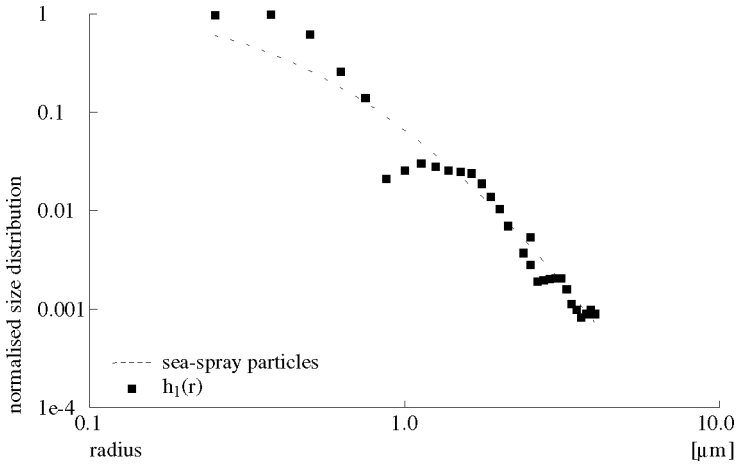

Fig. 4. Normalised size distribution for sea-spray particles (dashed line) and for particles determined by mode 1 (points)

Fig. 4 likewise shows two functions: one reflects the normalised size distribution of 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 other is the normalised size distribution calculated for mode 1. The total number of particles is defined in terms of the final extinction coefficient normalised to unity at 0.55 μm. The dashed line describes the model function (International Association..., 1984) on the assumption that the

mean particle radius $r = 0.3$ μm. Since amplitudes β'_{i1} (see eq. (5b)) have positive values, $h_1(r)\beta'_{i1}$ could be interpreted as an additional aerosol source (sea-spray particles).

Both functions, *i.e.* the mean aerosol size distribution and mode 1, can be represented by the sum of two gamma functions

$$< n(r) >', h_1(r) = \sum_{k=1}^{2} A_k (\frac{r}{r_0})^{\alpha_k} \exp[-b_k(\frac{r}{r_0})], \tag{6}$$

where the parameters $A_k, r, r_0, \alpha_k, b_k$ for each function are listed in Tab. 2.

Table 2. Parameters of modified gamma functions

Function	k	r_0 [μm]	r [μm]	α_k	b_k	A_k [m^{-3}]
mean value	1	1	0.25–0.63	1.35	5.7	3.31×10^6
$< n(r) >'$	2	1	0.64–4.0	1.2	1.75	$2.3\ \times10^5$
mode 1	1	1	0.25–0.88	5.04	16.38	6.28×10^4
$h_1(r)$	2	1	0.89–4.0	3.3	2.97	0.475

Initial analysis shows that the amplitude function β'_{i1} is closely dependent on the wind speed and its direction. To study the influence of these two factors on the aerosol size distribution amplitude over the Baltic Sea, all the data collected were separated into two air-advection classes, established on the basis of wind direction and 1000 mb air-mass trajectories. One of these classes refers to air advected from the NE–ENE wind sector with a wind fetch over the water in excess of *ca* 400 km, the other to the WSW–NW wind sector with a wind fetch over the water of no more than *ca* 50 km, or to air advected along the shore in the case of coastal data. The amplitude function associated with the NE–ENE sector was found to decrease with wind speed (see Fig. 5a). When fitted to the amplitude data, the linear curve showed good correlation with the wind speed – this is borne out by the regression coefficient of 0.97. Then, the amplitude β'_{i1} from the NE–ENE wind direction data can be written as a function of the wind speed v

$$\beta'_{i1(NE-ENE)} = (-27.3v + 93.1)10^4. \tag{7}$$

With regard to the results in Fig. 5a, it can be concluded that for a significant wind fetch over the water and increasing wind speed, the amplitude decreases, reflecting the contribution of sea-spray particles to the total aerosol size distribution. Similar results were reported by Van Ejik and de Leeuw (1992).

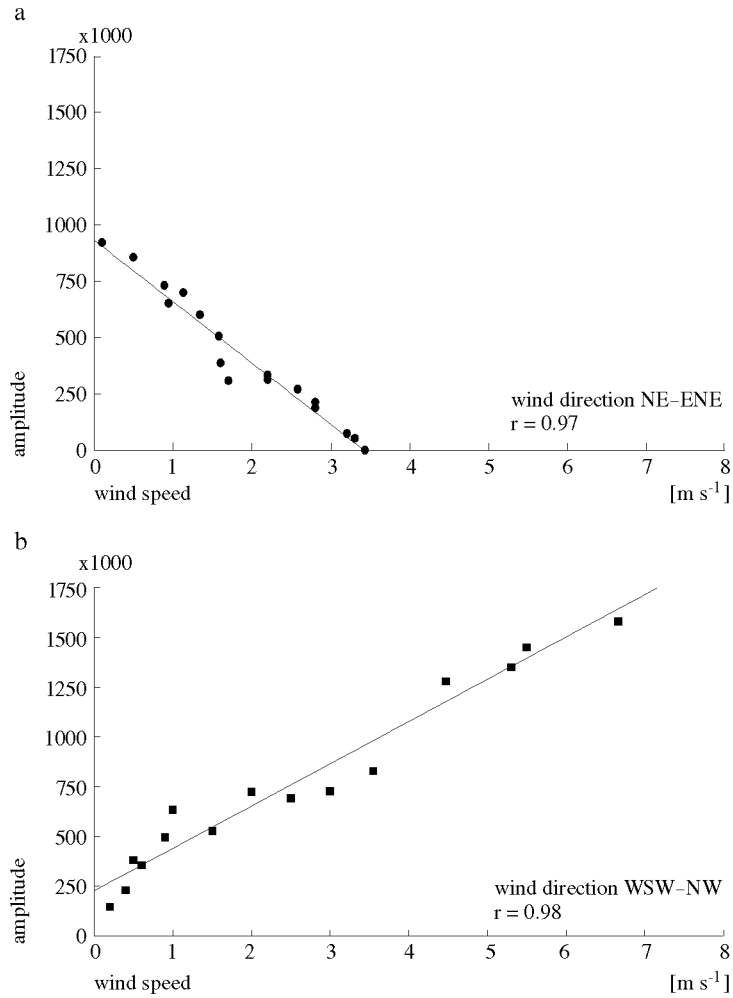

Fig. 5. Amplitudes of aerosol size distribution β'_{i1} plotted against wind speed: for the wind direction sector NE–ENE (a), for the wind direction sector WSW–NW (b); '×1000' indicates that the amplitude values should be multiplied by 1000

By contrast, the same aerosol amplitude functions associated with the WSW–NW wind sector were found to increase with increasing wind speed (see Fig. 5b). Although only 15 samples were collected within that wind sector, the distribution of points and the fitted curve still correlated fairly well with the wind speed, as manifested by the regression coefficient of 0.976. The amplitude function for the WSW–NW wind direction can be written as

$$\beta'_{i1(WSW-NW)} = (21.3v + 81.1)10^4. \tag{8}$$

These results reflect the cases when the air had to travel over the sea a distance of only *ca* 50 km. Therefore wind-wave interactions might not reach a balance with the wind speed, so the amplitude reflecting the contribution of sea-spray particles coming from breaking waves is high. On the other hand, as the wind speed increases, so does the production of sea–salt particles (Lovett, 1978; Monahan *et al.*, 1983).

An additional advantage of resolving the aerosol size distribution into two main components is that modelling the extinction and scattering coefficients becomes relatively simple, especially when both marine and terrestrial aerosols are making themselves felt. It is then possible to rewrite eq. (2) in the following way:

$$c(\lambda) = c_1(\lambda)\beta' + < c(\lambda) >', \tag{9}$$

where $c_1(\lambda)$ is the atmospheric extinction coefficient calculated for the aerosol size distribution determined by mode 1 and the refractive indices of sea-spray particles, $< c(\lambda) >'$ is the extinction coefficient obtained for the mean value of $< n(r) >'$ (eq. (5a), Tab. 2) and the refractive indices of dust-like particles. In both cases the complex refractive indices were taken from International Association... (1984). The total scattering coefficient $b(\lambda)$ can be calculated in like manner.

The ratio of scattering $b(\lambda)$ to extinction $c(\lambda)$ defines the single scattering albedo of an aerosol $\omega_A(\lambda)$. The values of this parameter are listed in Tab. 3 and presented in Fig. 6.

Table 3. The values of $\omega_A(\lambda)$

λ [nm]	$< \omega_A(\lambda) >$	$\delta\omega_A$	$\omega_{A\max}$	$\omega_{A\min}$
400	0.989	±0.011	1.000	0.978
488	0.989	±0.010	0.999	0.979
515	0.989	±0.010	0.999	0.979
550	0.989	±0.010	0.999	0.979
633	0.989	±0.009	0.998	0.980
694	0.990	±0.009	0.999	0.981

where $< \omega_A(\lambda) >$ is the mean value, $\omega_{A\max}$ and ω_{Amin} are the respective maximum and minimum errors determined by the limits of the standard deviation $\delta\omega_A$.

Obviously, the values of $\omega_A(\lambda)$ can be regarded as constant and equal to 0.989 within the range of visible wavelengths. This is confirmed by the values of ω_A for the maritime model of an aerosol published by International Association... (1984) (see Fig. 6).

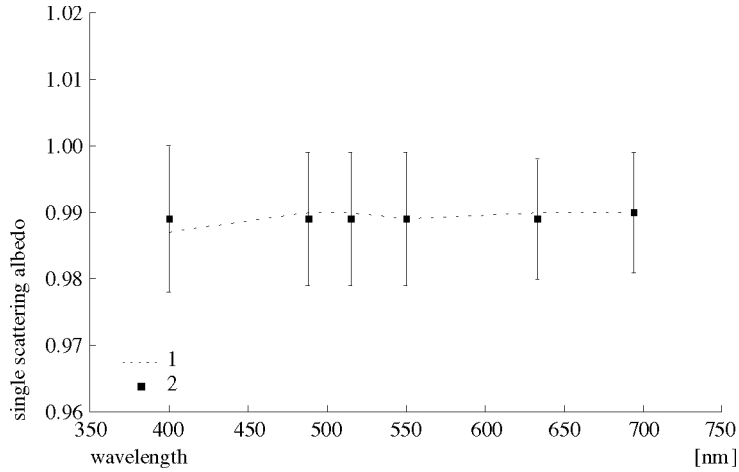

Fig. 6. The single scattering albedo of an aerosol: 1 – mean value of $\omega_A(\lambda)$ based on the experimental aerosol size distribution and the EOF method, 2 – values of $\omega_A(\lambda)$ for the maritime model of an aerosol (International Association..., 1984)

5. Conclusions

The results of this research are of a preliminary nature. The application of the EOF method to aerosol size distribution spectra has enabled us to reach the following conclusions:

- All spectra of the aerosol size distribution over the Baltic Sea can be defined by means of one general equation taking into account only the first mode h_1, because the contribution of the first eigenvalue to the total variance is 95.5%.
- It seems that $< n(r) >'$ could be interpreted as the slowly varying terrestrial background. However, the product $h_1(r)\beta'_{i1}$ could be interpreted as an additional aerosol source reflecting the contribution of seaspray.
- The temporal variability in the aerosol size distribution $n(r)$ is determined by the amplitudes β_{i1}. The direct relationships between these and the optical parameters of the atmosphere have been analysed in depth. However, only wind speed and wind direction turned out to have a significant influence on β_{i1}.
- A significant decrease in the aerosol size distribution amplitude with increasing wind speed was found when the air had to travel a distance of more than 400 km over the sea. Conversely, the aerosol size distribution amplitude for mixed terrestrial/marine air was found to increase

with increasing wind speed when the wind fetch over the water was no longer than *ca* 50 km.

- The resolution of the aerosol size distribution into two main components may be useful in modelling the aerosol extinction, scattering and in the determination of the single scattering albedo.

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Appendix

Let $n_i(r_j)$ be the aerosol size spectral distribution. The subscript i symbolises each successive measurement $i = 1, ..., N$; the subscript j corresponds to the number of spectral channels $j = 1, ..., M$. The mean spectral distribution is determined for this set of experimental data,

$$< n(r_j) >= \frac{1}{N} \sum_{k=1}^{N} n_k(r_j) \qquad j = 1, ..., M \qquad \text{(A1)}$$

as are the spectral distributions of fluctuations ${}^*n_i(r_j)$ with respect to the calculated mean distribution $< n(r_j) >$,

$${}^*n_i(r_j) = n_i(r_j) - < n(r_j) > \qquad i = 1, ..., N; \quad j = 1, ..., M. \qquad \text{(A2)}$$

Fluctuation distributions are approximated by expanding them into a series of functions

$${}^*n_i(r_j) = \sum_{k=1}^{M} h_k(r_j)\beta_{ik} \qquad i = 1, ..., N; \quad j = 1, ..., M. \qquad \text{(A3)}$$

Functions $h_i(r_j)$ should fulfil the conditions of orthogonality and be normalised:

$$\sum_{j=1}^{M} h_i(r_j)h_k(r_j) = M\delta_{ik} \qquad i, k = 1, ..., M, \qquad \text{(A4)}$$

where

$$\delta_{ik} = \begin{cases} 0 & \text{for} \quad \text{i} \neq \text{k} \\ 1 & \text{for} \quad \text{i} = \text{k} \end{cases} .$$

Functions $h_k(r_j)$ satisfying the above condition are called modes or main components, while their corresponding coefficients β_{ik} are known as amplitude functions, or simply amplitudes (Preisendorfer, 1988).

In the EOF method, functions h_k are chosen as eigenfunctions (eigenvectors) of the covariance matrix $\jmath(r_i, r_j)$,

$$\jmath(r_i, r_j) = \frac{1}{N} \sum_{k=1}^{N} {}^*n_k(r_i){}^*n_k(r_j) \qquad i, j = 1, ..., M, \qquad \text{(A5)}$$

which, at the same time, are solutions of the equation

$$\sum_{i=1}^{M} \jmath(r_i, r_j)h_k(r_i) = \wp_k h_k(r_j) \qquad j, k = 1, ..., M, \qquad \text{(A6)}$$

where $\wp_k$ are eigenvalues of the covariance matrix.

Eigenvalues $\wp_k$ and eigenvectors h_k are usually calculated by Jacobi's method (Ralston, 1975).

Amplitudes β_{ik} can be estimated from

$$\beta_{ik} = \sum_{j=1}^{M} {}^{*}n_i(r_j)h_k(r_j) \qquad i = 1, ..., N; \quad j = 1, ..., M. \tag{A7}$$

Amplitudes β_{ik} also have to fulfil the orthogonality condition

$$\tfrac{1}{N} \sum_{k=1}^{N} \beta_{ki}\beta_{kj} = \tfrac{1}{M}\wp_i\delta_{ij} \qquad i, j = 1, ..., M \tag{A8}$$

and their variance – the following dependence:

$$\tfrac{1}{M}\wp_j = \tfrac{1}{N} \sum_{k=1}^{N} \beta_{kj}^2 \qquad j = 1, ..., M. \tag{A9}$$

The above equation shows the connection between amplitudes β_{ik} and the eigenvalues $\wp_i$ of the covariance matrix $\jmath(r_i, r_j)$. Full information about the aerosol size distribution is given by the equation

$$n_i(r_j) = \sum_{k=1}^{l} h_k(r_j)\beta_{ik} + < n(r_j) > \qquad l = 1, ..., L; \; i = 1, ..., N, \tag{A10}$$

where L is the number of modes chosen in accordance with the following criterion:

$$\aleph(L) = \frac{\sum_{i=1}^{L} \wp_i}{\sum_{i=1}^{M} \wp_i} \qquad L = 1, ..., M. \tag{A11}$$

Apart from the parameter $\aleph(L)$, an additional parameter $\imath_i$, defining the contribution of the individual eigenvalues $\wp_i$ to the total variance, is also calculated:

$$\imath_i = \frac{\wp_i}{\sum_{i=1}^{M} \wp_i}. \tag{A12}$$

In practical calculations, values of $\aleph(L)$ are assumed to be of the order of 0.90–0.95 (Nielsen, 1979; Wróblewski, 1986; Jankowski, 1994). Mode h_1 contains the maximum energy of the entire data set, the next one contains the maximum energy of what is left after the first mode has been hived off, *etc.* More details about this method may be found in Kundu *et al.* (1975), Nielsen (1979) and Preisendorfer (1988).