Marine aerosol fluxes in the coastal zone – BAEX experimental data

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Abstract

This paper presents the results of investigations designed to determine the exchange processes between the sea and the air above the coastal zone. The calculations were carried out using experimental data collected by impactor measurements of the marine aerosol above the coastal zone during the Baltic Aerosol Experiment – I and II (BAEX–I, II).

1. Introduction

The determination of marine aerosol fluxes is a necessary step in attempts to parametrize the mass exchange between sea and atmosphere, particularly as regards the emission of pollutants from the sea surface. Studies concerning aerosol fluxes are rather infrequent, though the first attempt to estimate the production of aerosols from the sea surface was undertaken nearly half a century ago by Woodcock (1952). The majority of the references cited deal with the mean macroscale emission fluxes calculated on the basis of marigenic aerosol concentrations in the atmosphere. Some of the articles discuss aerosol fluxes from the point of view of particular emission mechanisms; for instance Monahan (Monahan et al., 1983) parametricates emission on the basis of the emission flux produced by a single whitecap measured in a laboratory tank.

This article presents a method for calculating the mean aerosol emission flux in the coastal zone and the evaluation of emission in the coastal zone from experimental data collected during the BAEX experiment (Petelski et al., 1993).
2. Measurements

The data were collected in the coastal zone of the southern Baltic Sea at the Lubiatowo station (BAEX–I and II) and on board r/v ‘Oceania’ (BAEX–I). The aerosol formation was measured with six-stage impactors (Stramska, 1988) located at different heights. At the land station, the lowest impactor was 2 m above sea level, the next ones 3 and 4 m above sea level, and the topmost one 5 m above sea level. The impactors on the ship were situated 0.5 m, 2 m and 6 m above sea level and 2 m from the side in order to reduce the measurement error resulting from drops generated by the waves washing off the ship’s side. The impactors worked simultaneously on land and on the ship. The impaction time was kept constant at 30 min, the air was sucked in at a rate of 800 \( \text{lh}^{-1} \). The concentration of the marine aerosol was expressed in terms of concentration of sea salt per \( \text{m}^3 \) air, since it is a significant component of the aerosol.

Simultaneously, the measurements of turbulent fluxes were made on the beach, 3 m above sea level, by a group of scientists from the Institute of Water Problems in Moscow. Fluctuations in the horizontal and vertical components of the wind velocity were measured with a sonic anemometer, humidity with a Lyman alpha hydrometer and air temperature with a resistance sensor. The momentum, heat and humidity turbulent fluxes were calculated from these measurements. Their methodology was described in the paper by Kanemasu and Panin (1992).

The average wind speed, temperature and humidity were measured at the land station and also on board the ship, located 2 miles off shore, facing the wind. Fig. 1 shows the position of the measurement sites.

The data used in this study comprise 20 measurement series from beach and sea (BAEX–I) and 18 series from the beach (BAEX–II).

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Fig. 1. Location of measurement sites: ● – land station at Lubiatowo \((\phi = 54^\circ 47'N, \lambda = 17^\circ 48')\), ○ – on board r/v ‘Oceania’ \((\phi = 54^\circ 52'N, \lambda = 17^\circ 54')\)
3. The method of calculating the aerosol emission flux

Calculation of aerosol fluxes from experimental data is a complex task. For the open sea area, where significant horizontal homogeneity occurs, the calculation of aerosol production can be based on turbulent and imission fluxes of the marine aerosol. In the case of the coastal zone, horizontally unhomogeneous, advective fluxes of marigenic aerosol have to be incorporated into the calculations. Emission can be calculated from the aerosol balance in the air layer near the sea over the coastal zone.

The aerosol balance in a hypothetical cuboid, constructed in the near-water air layer over the coastal zone (Fig. 2) can be expressed by the equation

\[ F_{u_1} + F_E - F_{I_1} + F_{I_2} - F_T - F_{u_2} = 0, \]  

where

- \( F_{u_1}; F_{u_2} \) – advective fluxes through the side walls of the cuboid,
- \( F_{I_1}; F_{I_2} \) – gravimetric imission flux at the top and bottom walls respectively,
- \( F_T \) – turbulent diffusion flux through the top side,
- \( F_E \) – emission flux from the bottom of the cuboid.

Fig. 2. Diagram of the aerosol balance over the coastal zone
3.1. Assumptions introduced into the calculations

The initial assumption applied in the calculations is the hypothesis of horizontal quasihomogeneity. It was assumed that over the surf zone a homogenous boundary layer is formed different from that over the open sea and over the land. Thus, all fluxes along the longer wall ($x$-axis) of the cuboid in Fig. 2 are constant:

$$\frac{\partial}{\partial x} F_i = 0. \quad (2)$$

The assumption that the boundary layer in the coastal zone is quasihomogeneous does not include horizontal homogeneity in general, but does include advection:

$$F_{u_1} \neq 0, \quad F_{u_2} \neq 0, \quad F_{u_1} \neq F_{u_2}. \quad (3)$$

(In our method of calculations the boundary layer above the surf zone is treated as horizontally quasihomogeneous). Such an assumption can be introduced because the experiment at Lubiatowo (Panin, 1994) showed that nearly all the turbulent transport is accomplished by disturbances with frequencies greater than $f_{\text{min}} = 0.1 \text{ Hz}$, which means that the linear dimensions of these disturbances $L$ have to be smaller than $L_{\text{max}}$:

$$L_{\text{max}} \geq \frac{\bar{u}(z)}{f_{\text{min}}}, \quad (4)$$

where

$\bar{u}$ – mean wind speed.

Since the wind speed was assumed to range from 0 to 10.0 m s$^{-1}$, $L_{\text{max}} \leq 100$ m, the width of the surf zone $D$ was taken to be from 20 to 1000 m and $D$ is greater than $L$ by about one order of magnitude. During the measurements the condition that

$$D \gg L_{\text{max}}, \quad (5)$$

was always valid.

Taking into account these assumptions, eq. (1), the total flux, takes the form

$$\frac{1}{D} F_{u_1} + F_E - F_{I_1} + F_{I_2} - F_T - \frac{1}{D} F_{u_2} = 0, \quad (6)$$

where

$F_{u_1}$ – advective flux of marine aerosols [µg m$^{-2}$ s$^{-1}$],
$F_{u_2}$ – on-shore advective flux [µg m$^{-2}$ s$^{-1}$],
$F_T$ – turbulent flux [µg m$^{-2}$ s$^{-1}$],
$F_I$ – imission flux [µg m$^{-2}$ s$^{-1}$],
$F_E$ – emission flux [µg m$^{-2}$ s$^{-1}$],
$D$ – length of breaker zone [m].
During the experiment the size of the sea-salt particles measured was included in the spectrum of the aerosol, which can be regarded as a mixture of substances (Leeuw, 1987). The calculations presented in this paper were carried out to evaluate the flux emitted from the seawater sublayer in the direct sphere of wave activity (Petelski, 1978), not from the sea surface. Thus, the bottom of the cuboid in Fig. 2 was not at sea level but was set 2 m above, and the highest measuring level was 5 m above sea level. This arrangement of the measuring levels allowed for a more precise evaluation of imissive and advective fluxes than was possible in calculations from sea level, which would require the effect of waves on the wind profile to be incorporated into the equation. This cuboid arrangement fitted well with the aerosol concentration measuring levels located on the beach (2 m, 3 m, 4 m and 5 m above sea level).

3.2. Turbulent aerosol flux

The turbulent aerosol flux $F_T$ can be calculated from gradient measurements on the beach. Analysis of the boundary level structure (Panin and Krivitski, 1992) shows that the measurement equipment should be placed at an appropriate level above the beach and the parameters characteristic of the boundary layer over the coastal zone can then be obtained. This conclusion was confirmed by impactor measurements carried out on the beach during the experiment at Lubiatowo (Petelski et al., 1994). The profiles measured during that experiment displayed negative vertical concentration gradients in the upper part, which is an indication of upward aerosol transport.

As the layer under investigation is horizontally quasihomogenous, the turbulent fluxes in it can be described to a first approximation by the theories of Monin and Obukhov (1953). According to these, non-dimensional gradients of physical parameters in the near-water atmospheric layer are functions of the single dimensionless parameter $z/L$:

$$\frac{\chi z \partial u}{U_* \partial z} = \varphi_u(z/L),$$

$$\frac{z \partial \Theta}{T_* \partial z} = \varphi_\theta(z/L),$$

where

$$L = \frac{-u_*^3}{\chi \alpha Q},$$

$L$ – length scale after Monin and Obukhov,
$Q$ – sensible heat flux,
$\chi$ – Karman constant,
\( \alpha = \frac{g}{T_p} \) – buoyancy parameter,
\( g \) – acceleration due to gravity,
\( T_p \) – air temperature,
\( u_* \) – wind speed scale – the friction velocity,
\( T_* \) – temperature scale,
\( u \) – wind speed,
\( \Theta \) – potential temperature,

\[
u_* = \left(\frac{\tau}{\rho}\right)^{0.5}, \quad (9)
\]

\[
T_* = -\frac{Q}{\chi u_*}, \quad (10)
\]

where
\( \tau \) – shearing stress.

In view of the fact that the properties of the near-water atmospheric layer depend not only on the fluxes of momentum, heat and humidity but also on the aerosol flux, the parameters characterising this layer should include this flux \( F_N \) or \( F_M \). Thus the new scales \( M_* \) and \( N_* \) can be expressed as follows:

\[
M_* = \frac{F_M}{u_*}, \quad (11)
\]

\[
N_* = \frac{F_N}{u_*}. \quad (12)
\]

The non-dimensional aerosol concentration gradient comes to

\[
\frac{z}{M_*} \frac{\partial M}{\partial z} = \phi_M \left( \frac{z}{L} \right), \quad (13)
\]

thus

\[
M(z_2) - M(z_1) = M_* \left[ f \left( \frac{z_2}{L} \right) - f \left( \frac{z_1}{L} \right) \right], \quad (14)
\]

\[
N(z_2) - N(z_1) = N_* \left[ f \left( \frac{z_2}{L} \right) - f \left( \frac{z_1}{L} \right) \right], \quad (15)
\]

where

functions \( f_N(\xi) \) and \( f_M(\xi) \) are the respective primary functions \( \xi^{-1} \phi_N(\xi) \) and \( \xi^{-1} \phi_M(\xi) \),

\( z, z_1, z_2 \) – measuring level above sea level,

\( M \) – mass concentration of aerosol in one cubic metre of air,

\( N \) – number of sea-salt particles in one cubic metre of air.

If the marigenic aerosol can be regarded as a mixture of substances, then it seems probable that the function \( \phi_M \) has a form similar to \( \phi_T \).
and $\phi_u$. Zilitinkevitch et al. (1978) suggested that, for practical purposes, the advective flux can be described by the following expression:

$$f_u(\xi) = \begin{cases} 
\ln \xi + 10\xi & 0 < \xi \\
\ln |\xi| & -0.07 \leq \xi \leq 0 \\
0.25 + 1.2\xi^{-1/3} & \xi < -0.07.
\end{cases} \quad (16)$$

Assuming that $f_N = f_M = f_T = f_u$, the practical determination of the aerosol gradient can be conducted using the function given in eq. (16).

The scales $M_*$ and $N_*$ can be determined on the basis of the aerosol concentrations measured at several levels and eqs. (14) and (15). These measurements were carried out under nearly neutral stratification conditions (see Tab. 1). For this reason the calculations of $M_*$ were done using only the logarithmic branch of the function $f(z/L)$ – from eq. (16). $M_*$ was calculated using the least squares method, under the assumption that $M_*$ is the regression coefficient of the line formed by impactor measurements in the plane $M(z), \ln(z/L)$. Thus the new scales $M_*$ and $N_*$ were used to calculate the turbulent fluxes of the aerosol $F_T$:

$$F_T = M_* u_*, \quad \text{or} \quad (17)$$

$$F_T = N_* u_*, \quad (18)$$

where $u_*$ was calculated from measurements of pulsations of wind speed components on the beach.

**Table 1.** Selected parameters characterising the dynamics of the near-water atmospheric layer during the BAEX–I (1993) experiment

<table>
<thead>
<tr>
<th>Date Time</th>
<th>$u_*$ [m s$^{-1}$]</th>
<th>$Q$ [J s$^{-1}$ m$^{-2}$]</th>
<th>$L$ [m]</th>
<th>$z/L$ $z = 3$ m</th>
<th>$z/L$ $z = 4$ m</th>
<th>$z/L$ $z = 5$ m</th>
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<td>0.15</td>
<td>18.00</td>
<td>-17.04</td>
<td>-0.17</td>
<td>-0.23</td>
<td>-0.29</td>
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<td>-0.37</td>
<td>-0.47</td>
</tr>
<tr>
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<td>-0.01</td>
<td>-0.01</td>
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<td>0.05</td>
<td>0.07</td>
</tr>
<tr>
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<td>0.09</td>
<td>0.12</td>
<td>0.15</td>
</tr>
<tr>
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<td>0.06</td>
<td>0.08</td>
<td>0.10</td>
</tr>
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Table 1. continued

<table>
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<tr>
<th>Date Time</th>
<th>( u_\ast ) [m s(^{-1})]</th>
<th>( Q ) [J s(^{-1}) m(^{-2})]</th>
<th>( L ) [m]</th>
<th>( z/L ) ( z = 3) m</th>
<th>( z/L ) ( z = 4) m</th>
<th>( z/L ) ( z = 5) m</th>
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<tr>
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<td>0.12 0.11</td>
<td>-10.00 5.00</td>
<td>-24.20 -72.70</td>
<td>-0.12 -0.04</td>
<td>-0.16 -0.05</td>
<td>-0.20 -0.06</td>
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<td>10.00</td>
<td>-72.70 -142.30</td>
<td>-0.04 -0.02</td>
<td>-0.05 -0.03</td>
<td>-0.06 -0.04</td>
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<tr>
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<td>0.28</td>
<td>7.00</td>
<td>-284.60 -124.50</td>
<td>-0.01 -0.02</td>
<td>-0.01 -0.03</td>
<td>-0.02 -0.04</td>
</tr>
<tr>
<td>22.10 13.55</td>
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<td>25.00</td>
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<td>-0.03 -0.07</td>
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<tr>
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<td>0.42</td>
<td>32.00</td>
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<td>-0.01 -0.03</td>
<td>-0.01 -0.05</td>
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</tr>
<tr>
<td>23.10 23.00</td>
<td>0.25</td>
<td>21.00</td>
<td>-71.10 -34.00</td>
<td>-0.04 -0.03</td>
<td>-0.05 -0.05</td>
<td>-0.06 -0.07</td>
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<td>23.10 09.00</td>
<td>0.31</td>
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<td>-0.06 -0.07</td>
</tr>
<tr>
<td>23.10 13.30</td>
<td>0.29</td>
<td>31.00</td>
<td>-71.10 -71.10</td>
<td>-0.04 -0.03</td>
<td>-0.05 -0.05</td>
<td>-0.06 -0.07</td>
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<tr>
<td>23.10 16.30</td>
<td>0.28</td>
<td>26.00</td>
<td>-76.30 -76.30</td>
<td>-0.03 -0.03</td>
<td>-0.05 -0.05</td>
<td>-0.06 -0.07</td>
</tr>
<tr>
<td>23.10 19.30</td>
<td>0.28</td>
<td>26.00</td>
<td>-76.30 -76.30</td>
<td>-0.03 -0.03</td>
<td>-0.05 -0.05</td>
<td>-0.06 -0.07</td>
</tr>
</tbody>
</table>

\( u_\ast \) – friction velocity,
\( Q \) – sensible heat flux,
\( L \) – length scale.

\[
 u_\ast = \sqrt{u'w'},
\]  

where
\( u' \) – pulsations of the horizontal component of the wind speed,
\( w' \) – pulsations of the vertical component of the wind speed,
and the dash over the symbols designates the averaging over physical realisations; this means that this averaging was substituted by averaging over time by applying the ergodity of the wind pulsation field.
3.3. Advective fluxes

Advective fluxes can be calculated from the aerosol concentration data and wind speed values at the entrance to and exit from the cuboid studied by applying the following equations:

\[ F_{u_1} = \int_{0}^{z_1} M_1(z) u_1(z) dz, \]  
(20)

\[ F_{u_2} = \int_{0}^{z_2} M_2(z) u_2(z) dz. \]  
(21)

As the wind profile at the measuring levels can be approximated to a logarithmic profile with a suitable confidence level, eqs. (20) and (21) can be transformed into

\[ u(z) = \frac{u_*}{\chi} \ln \left( \frac{z}{z_0} \right). \]  
(22)

This equation contains two parameters \(z_0\) and \(u_*\), which must be determined before the defined wind profile can be estimated; \(u_*\) is described by eq. (19).

Besides the turbulent fluxes, the mean wind speed at a height of 3 m above sea level was measured during the experiment. The results obtained served to determine \(z_0\):

\[ z_0 = 4 \exp \left( \frac{\chi u(3)}{u_*} \right). \]  
(23)

In calculations conducted for the open sea region, \(u(z)\) was applied in the form of eq. (22), where \(z_0\) – the friction factor – was calculated in accordance with the Charnock equation (Druet et al., 1976):

\[ z_0 = m \frac{u_*^2}{g}, \]  
(24)

where

\(m = 0.035,\)  

\(g\) – acceleration due to gravity.

The friction velocity was calculated from the mean wind speed measured at a height of 10 m.

\[ u_*^2 = c_{10} u_{10}^2, \]  
(25)

where

\(c_{10}\) – drag coefficient.

The vertical profile of the aerosol concentration \(M(z)\) (eqs. 14, 15) can be presented in the form

\[ M(z) = M_* \ln z + C_c. \]  
(26)
The function $M(z)$ on the beach was determined from the results of measurements at four levels and function (25) was resolved by the least squares method. For the open-sea region $M(z)$ was considered in the form

$$M(z) = M_* \ln \left( \frac{z}{z_0} \right).$$ (27)

The values of $z_0$ were the same as for the wind profile calculations, and $M_*$ was estimated from measurements of aerosol concentrations at 2 m elevation (carried out on board r/v ‘Oceania’). Having incorporated all these elements into eq. (21), the advective flux in the open sea region was described by

$$F_{u_1} = \int_2^5 \frac{M_* u_{10}}{\chi \sqrt{C_{10}}} \ln^2 \left( \frac{z g c_{10}}{m u_{10}^2} \right) dz.$$ (28)

The advective flux on the beach (eq. 20) was calculated from the following expression:

$$F_{u_2} = \frac{u_*}{\chi} \int_2^5 (M_* \log z + C_c) \ln \left( \frac{z}{z_0} \right) dz.$$ (29)

The data obtained simultaneously on board the research vessel and on the beach served to calculate the following: advective fluxes using eqs. (28) and (29); turbulent fluxes (17, 18); the difference between imission fluxes at 5 m and 2 m above sea level, i.e. the imission fluxes were calculated from eq. (30) taking into account eq. (29); the effective emission flux was calculated from eq. (5) as the sum of these parameters. The calculated values are given in Tab. 2.

**Table 2. Aerosol fluxes over the coastal zone – BAEX–I**

<table>
<thead>
<tr>
<th>Date</th>
<th>Time</th>
<th>$\frac{1}{D}F_{u_1}$</th>
<th>$\frac{1}{D}F_{u_2}$</th>
<th>$F_T$</th>
<th>$\Delta F_I$</th>
<th>$F_E$</th>
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<tr>
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$\frac{1}{D}F_{u_1}$ – marigenic advective flux [$\mu g m^{-2} s^{-1}$],

$\frac{1}{D}F_{u_2}$ – shore advective flux [$\mu g m^{-2} s^{-1}$],

$F_T$ – turbulent flux [$\mu g m^{-2} s^{-1}$],

$\Delta F_I$ – imission flux [$\mu g m^{-2} s^{-1}$],

$F_E$ – emission flux [$\mu g m^{-2} s^{-1}$],

$D$ – length of breaker zone [m].
3.4. Immission flux

Immission flux $F_I$ is generated by aerosol particles precipitating from the air and is given by

$$F_I = MV_D,$$

(30)

where

- $M$ – particle concentration,
- $V_D$ – deposition velocity.

The precipitation rate was introduced after Zieliński (1993)

$$V_D = \frac{V_T}{1 - \exp[-V_r/c_{10}u_{10}]} ,$$

(31)

where

- $c_{10}$ – drag coefficient:
  - $c_{10} = 1.14 \times 10^{-3}$ for $u_{10} < 10 \text{ m s}^{-1}$,
  - $c_{10} = (0.49 + 0.065 u_{10}) \times 10^{-3}$ for $u_{10} > 10 \text{ m s}^{-1},$
- $u_{10}$ – wind speed,
- $V_T$ – Stokes’ velocity.

The values of the immission fluxes calculated from the above equations are presented in Tab. 2.

4. Results

The aerosol fluxes over the coastal zone calculated from the experimental data by the method described are presented in Tab. 2. The last column (5) gives the results calculated from the emission flux balance eq. (6).

The emission flux took values between 3.2 and 384 $\mu g \text{m}^{-2} \text{s}^{-1}$. Such a wide range corresponds well with flux ranges over oceans (Garbalewski, 1983). The mean value of the calculated fluxes is also in accordance with the results obtained by other authors, e.g. Blanchard (1985), Erickson et al. (1986). Assuming that the Polish coastline of the Baltic is 500 km long and that the mean width of the coastal zone is 50 m, the mean annual emission from this coastal zone would, according to our calculation, amount to $1.5 \times 10$ tons per year. This result indicates the importance of the surf zone in the exchange processes between the sea and atmosphere. Stramska (1984) estimated the total emission from the Baltic Sea aerosol at $1.7 \times 10$ tons per year.

The turbulent aerosol flux was calculated on the basis of data from both experiments. The values of this flux ranged from 1 $\mu g \text{m}^{-2} \text{s}^{-1}$. The turbulent aerosol flux as a function of wind speed is presented in Fig. 3. The correlation coefficient between wind speed and turbulent aerosol flux was found to be 0.71. The correlation coefficient between the wave parameters
and the turbulent aerosol flux (0.89) was even higher than between the wind speed and the turbulent aerosol flux.

Fig. 4 shows the ratio of the calculated aerosol flux to the difference between the zero moments of the wave spectra \((M_{01} - M_{02})^{3/4}\), where \(M_{01}\) and \(M_{02}\) are the respective zero moments of the wave spectra obtained from wavegraphs situated 90 m and 30 m from the shoreline. The relation between changes in wave energy and aerosol flux in the coastal zone is very clear. As the aerosol emission fluxes are much greater in the surf zone than in the open-sea area, it becomes evident that aerosol concentrations are influenced more strongly by horizontal transport than by the vertical component.

This finding is illustrated in Fig. 5 in the form of a ratio (%) between the advective fluxes and emission fluxes. In most cases, the advective flux and emission flux were independent of the wind speed. The figure indicates that the fluxes are not correlated with each other, although the values of the advective flux are well correlated with wind speed. As regards the vertical
Fig. 4. Relation between calculated turbulent flux $F_T$ and the difference of zero moments of the wave spectrum $(\delta M_0)^{3/4}$

Fig. 5. Ratio between advective flux and emission flux as a function of wind speed

fluxes, the turbulent flux and imission flux, they are much better correlated with the wave parameters than with the wind speed, which indicates that emission derived from the swell makes a greater contribution in the coastal zone. Imission fluxes are the least important component of the aerosol balance over the coastal zone, their levels being three orders of magnitude lower than those of the emission fluxes.
The method presented here of calculating emission fluxes from the coastal zone can be applied under meteorological conditions which satisfy the inequality (4), i.e. in most meteorological situations except for convection in windless weather.

The precision of the method is determined by the precision of aerosol measurement on the beach and at sea. All the fluxes included in the balance were calculated from aerosol concentration data. The turbulent flux is connected directly with aerosol measurements by eqs. (14) and (15), advective fluxes by eqs. (20) and (21), and imission fluxes by eq. (30). Because this method of determining aerosol concentration is liable to ±25% error, the error of flux calculation is of the same order.

5. Conclusions

This paper presents the first step towards balancing aerosol fluxes above the breaker zone. The results obtained point to the very important role of the coastal zone in the aerosol exchange between the sea and atmosphere. The emission fluxes obtained from the coastal zone are much greater than those calculated for the open-sea region.

The vertical turbulent flux of aerosol in the coastal zone depends greatly on the dissipation of wave energy.

Imission fluxes can be neglected in aerosol balance calculations in the coastal zone.

Advective fluxes are the most important for the coastal zone: it is there that they attain the highest values.

The results also indicate the necessity of including an advective component in studies of aerosols over the coastal zone and of carrying out simultaneous measurements on shore and at sea.

The relation between the fluxes obtained speaks in favour of the method of emission flux calculation from the coastal zone presented in this paper.

References

Garbalewski C., 1983, Mean spatial distribution of basic physical characteristics and source regions of particle emission from the ocean surface, Oceanologia, 14, 139–165.


