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Marine aerosol fluxes over open sea calculated from vertical concentration gradients

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Abstract

Sea spray emission fluxes were calculated on the basis of vertical gradients of the aerosol concentration from experimental data collected with a laser particle counter during two polar scientific cruises carried out in July and August 2000 and 2001, and during cruises in the Baltic Sea in October 2000, and May and October 2001. Calculated fluxes range between 10^3 and 10^7 m^{-2} s^{-1}.

1. Introduction

Studies of production and transport of aerosol over the sea are very important for many areas of knowledge like cloud physics, atmospheric optics, and environmental pollution research or sea–atmosphere interaction. Intensive development of satellite methods in geophysical studies creates demand for information on near-water atmospheric layer aerosol.

Mechanisms of sea salt particle transport from the sea surface to the atmosphere are well understood thanks to the efforts of many researchers, among others Blanchard (1954, 1989), Cipriano, Blanchard, Hogan, and Lala (1983). Emission of drops from breaking wave whitecaps and sea foam is quite well-known thanks to laboratory studies of, e.g. Wu (1973) and Monahan, Spiel, and Davidson (1982). However, the problem of parameterization of aerosol emission from the sea surface has not been solved yet.

Without precise determination of the flux of drop emissions from the sea surface, it is not possible to decide whether sea spray affects the air–sea fluxes of heat and latent heat. This problem has been addressed for many years (Bortkovskii, 1972, 1987; Borisenkov, 1973; Petelski, 1986, 1996;

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Mestayer, Edson, Fairal, Larsen, & Spiel, 1989; Andreas, 1992; Petelski & Chomka, 1996b, 2000; Edson, Anquentin, Mestayer, & Sini, 1996).

Existing models of drop emission from the sea surface are based on laboratory studies (Monahan, Spiel, & Davidson, 1986; Monahan & Van Patten, 1989; Iida et al., 1992), or on measurements of aerosol size distributions under an assumption that flux of the particles falling out of the near-water atmospheric layer is equal to drop emission from the sea surface (Smith, Park, & Consterdine, 1993). Andreas (1998) reviewed the published functions of sea spray generation models. The conclusion is that the authors of models do not agree even on the order of magnitude of aerosol generation flux from the sea surface. Most of these models has been developed using speculations, not measurements.

According to Andreas (1998), the best existing model is the model by Smith et al. (1993), on which he based his own model. Smith and co-authors based their model on aerosol measurements carried out on 14 m high tower situated on an island coast. Smith et al. calculated emission flux from a simple flux balance equation based on an assumption of horizontal uniformity and stationary

$$\frac{\mathrm{d}F}{\mathrm{d}r} = V_{\mathrm{d}} \frac{\mathrm{d}N}{\mathrm{d}r}, \tag{1}$$

where $\mathrm{d}F/\mathrm{d}r$ in Eq. (1) is the emission source function (the number of particles of radii from r to $r + \mathrm{d}r$, emitted in a unit time from a unit sea surface area), V_{d} is the deposition velocity, and r is the particle radius.

Eq. (1) is fulfilled extremely rarely. Hence, an estimation of aerosol emission fluxes based on this equation is burdened with an error.

There are also estimations of aerosol emission based on measurements of aerosol concentration and mixing layer thickness (Fairall, Davison, & Schacher, 1983, 1984). Reid, Jonsson, Smith, and Smirnov (2001) used a similar method. However, this method is correct only for mezoscale averages.

Petelski and Chomka (1996a) gave a method for calculation of the mean emission fluxes in the coastal zone based on the aerosol balance in this zone. Their method was used to calculate emission fluxes from BAEX experiment data. The results allowed them to formulate aerosol emission model for the coastal zone (Chomka & Petelski, 1997) which relates aerosol emission flux with wave energy dissipation. The proportionality of aerosol emission flux to the mean wave energy dissipation has been confirmed by TABEX experiment results (Petelski & Chomka, 1996b).

The present work is the first step towards formulating an analogous model for the open sea. The model should relate vertical aerosol fluxes in the near-water atmospheric layer to the flux origin, i.e. wave energy dissipation through whitecaping. To achieve this, vertical aerosol fluxes should be determined from measurements. We propose a method for the flux estimation on the basis of measurements of vertical gradients of aerosol concentration in the near-water atmospheric layer. Sections 2 and 3 define and specify the area of our interest, in particular the aerosol flux and aerosol particle size range we take into account in our study. Further, we present a method for the flux computation based on the similarity theory of Monin and Obukhov (1953). In Sections 5 and 6 we apply this method to the data we collected during cruises to the Baltic and polar seas. Section 7, Conclusions, summarizes our findings and discusses possible future research arising from this study.

2. Aerosol fluxes in marine atmospheric surface layer

Drops of sea spray emitted from the sea surface are transported to higher atmospheric layers by turbulent diffusion. Advection and diffusion are responsible for horizontal transport of aerosols. Simultaneously, a portion of aerosol particles is removed from the air by gravitational fallout.

Let $n(r)$ denote $\mathrm{d}N/\mathrm{d}r(r)$, i.e. aerosol particle concentration with a radius r. $n(r)$ can be expressed as $n(r) = \overline{n(r)} + n'(r)$, where $\overline{n(r)}$ is a mean particle concentration with a radius r, and $n'(r)$ a momentary deviation from the mean.

After an analogous decomposition of the velocity vector $V_i = \overline{V_i} + V_i'$ into the mean and a momentary deviation, the turbulent flux of particles with a radius r may be expressed as follows:

$$F_{T_i}(r) = \overline{n'(r) \cdot V_i'}. \tag{2}$$

The bar in Eq. (2) symbolizes averaging over a set of realizations. In practice, the time averaging is usually applied (in our case over $\frac{1}{2}$ h), which is justified by the assumption of ergodicity of physical parameter fields in the near-water atmospheric layer.

The fallout flux of particles with a radius r may be formulated as follows:

$$F_{\mathrm{d}}(r) = V_{\mathrm{d}}(r) \cdot \overline{n(r)}. \tag{3}$$

The transport equation for aerosol particles of a size r can be expressed as

$$\frac{\mathrm{D}}{\mathrm{D}t}\bar{n}(r) = \frac{\partial}{\partial x_i}(\overline{n'(r)u_i'} + \overline{n(r)}V_{\mathrm{d}}(r)), \tag{4}$$

where $\bar{n}(r)$ is the mean particle concentration of radius r, and $\mathrm{D}/\mathrm{D}t = \partial/\partial t + \overline{u_i}(\partial/\partial x_i)$ the substantial derivative.

In the case of open seas we often deal with horizontally uniform conditions. Then Eq. (4) takes the form of

$$\bar{w}\frac{\partial\overline{n(r,z)}}{\partial z} = \frac{\partial}{\partial z}\overline{n'(r,z)w'} + \frac{\partial}{\partial z}(V_{\mathrm{d}}\overline{n(r,z)}), \tag{5}$$

where w is the vertical component of the velocity vector.

Because the mean vertical velocity is negligible in the near-surface atmospheric layer, Eq. (5) can be transformed into

$$\overline{n'(r)w'} + V_{\mathrm{d}}(r,z)\overline{n(r,z)} = \mathrm{const}. \tag{6}$$

Eq. (6) expresses the balance of the vertical fluxes in the near-water atmospheric layer under conditions of horizontal uniformity. It differs from Eq. (1), which is widely used for this layer. Eq. (6) shows that in order to study marigenic aerosol over open seas we need to determine only two fluxes. However, we must not forget about the horizontal uniformity assumption we made; that we assumed horizontal gradients are negligible. In the case of aerosol, this assumption is not fulfilled even over the apparently uniform horizontal sea surface, particularly for small particles, which can

travel over long distances. Therefore in the marine aerosol studies, it must be remembered that for particles of different sizes, different temporal and spatial scales should be taken into account.

3. Choice of the particle size range

The aim of our research is the quantitative determination of droplet emission from the sea surface. However, because of the measurement restrictions, we must limit our study to only a part of the sea spray size spectrum. Our measurements were conducted in the open sea from a ship, which entails a relatively high measurement height in order to avoid the ship hull influence. The measurements were taken in the layer of 5–20 m above sea level, which allowed us to avoid all the complexity of near-surface measurements.

The 5–20 m layer is reached by the drops transported by the means of turbulent diffusion. Therefore we will concentrate on the droplets of sizes for which turbulent diffusion is the main transport process. Transformation of Eq. (5) to the non-dimensional form yields

$$\frac{(\bar{w} - V_{\mathrm{d}})z}{N_* U_*} \frac{\partial \bar{n}}{\partial z} = \frac{z}{F_*} \frac{\partial}{\partial z} \overline{n'w'}, \tag{7}$$

where asterisks indicates scales of concentration N_*, velocity U_*, and flux F_*.

Eq. (7) indicates that the term for droplet fallout can be omitted in the aerosol transport equation when:

$$\frac{V_{\mathrm{d}}}{U_*} \ll 1. \tag{8}$$

If V_{d} is the sphere falling speed in viscous medium, Eq. (8) gives the condition for the aerosol particle size when gravitational fallout can be neglected in the turbulent diffusion equation

$$r \ll \left(\frac{9\rho_{\mathrm{p}} v U_*}{2\rho_{\mathrm{w}} g} \right)^{1/2}, \tag{9}$$

where ρ_{p} is the air density ≈ 1.29 [kg/m^3], ρ_{w} is the water density $\approx 10^3$ [kg/m^3], v is the air viscosity $\approx 1.8 \times 10^{-5}$ [Ns/m^2], and g the acceleration due to gravity ≈ 9.81 [m/s^2].

As the first approach, the friction velocity can be substituted with wind speed at 10 m above the sea level by the means of the following equation:

$$u_* = \sqrt{C_{10}} \cdot u_{10} \approx 0.04 u_{10}. \tag{10}$$

Substituting Eq. (10) into Eq. (9) we obtain the inequality that is presented graphically in Fig. 1. The area under the dotted curve represents the ranges of the parameters where the inequality (9) is satisfied. Fig. 1 indicate that for particles of a radius below 30 μm inequality (9) is satisfied for the whole range of wind speed relevant for droplet emission from the sea surface, i.e. $U_{10} > 3$ m/s.

The lower limit of the analyzed range of particle size is determined by the condition of horizontal uniformity, that is, horizontal gradient vanishing. Seeking a relationship between sea surface state and aerosol fluxes in the near-water atmospheric layer, we must select such sizes of the droplets for analysis for which the condition on horizontal uniformity of both aerosol concentration and sea

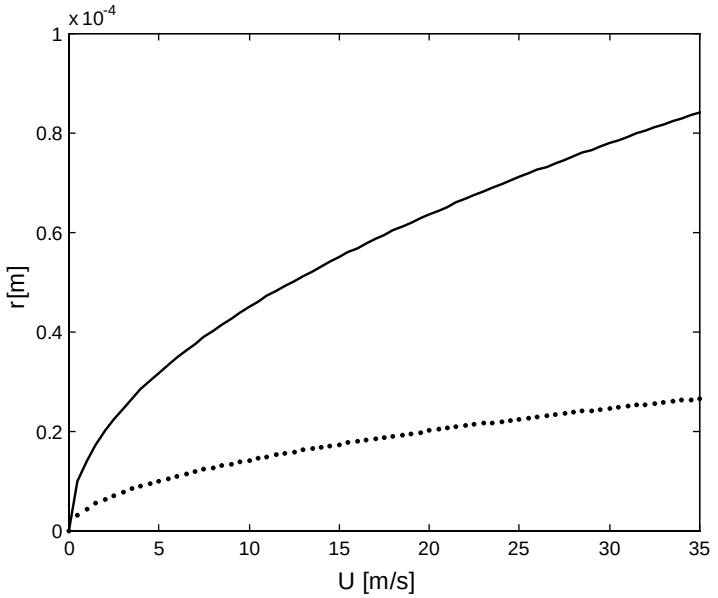

Fig. 1. Turbulent diffusion dominated particle sizes as a function of wind speed. Solid line $r = (9\rho_p \nu U_*/2\rho_w g)^{1/2}$; dotted line $r = 0.1(9\rho_p \nu U_*/2\rho_w g)^{1/2}$.

surface were satisfied in the same scales. $L_s = U_g(H/V_d(r))$ can be adopted as the horizontal advection scale for particles of a radius r, where U_g is geostrophic wind speed, H mixing layer height, and $V_d(r)$ denotes deposition velocity for particles of the radius r. Deposition velocity for particles of radius r from 0.1 to 1.0 μm is a very sharply increasing function whose values strongly depend on wind speed (Slinn & Slinn, 1981). In the radius range mentioned above, the function value increases from 0.01 to 3 cm/s because the wind speed is of order of 10^3 cm/s, and mixing layer height is of order of 10^2–10^3 m; thus $L_s = 10^7$–$10^8 \sim 10^5$–10^6 m. For particles of radius $r = 1$ μm, L_s takes values from 100 to 1000 km, but for particles of $r = 0.1$ μm, L_s can be as high as 10^4–10^5 km. The above reasoning shows that studies on aerosol emission from the sea surface need to be limited to the aerosol size range of 1 μm, especially when the studies are conducted on a small sea, such as the Baltic sea.

4. Flux parameterization by the means of Monin–Obukhov (M–O) theory

In studies of aerosol particle transport for sizes of 1–30 μm, the particle concentration can be treated as a scalar property of the air and thus the transport equation (4) can be expressed as

$$\frac{\partial}{\partial z}\overline{(n'w')} = 0. \tag{11}$$

Eq. (11) shows that for this range of sizes and under the condition of horizontal uniformity, vertical turbulent flux equals the emission from the sea surface

$$\frac{\partial F(r)}{\partial r} = \overline{n'(r)w'}. \tag{12}$$

The turbulent aerosol flux can be parameterized by the means of the similarity theory by Monin and Obukhov (1953). The structure of this horizontal uniformity over a water layer is fully defined be momentum flux τ, sensible heat flux Q, buoyancy parameter $\beta = g/T$. These parameters allow the following scales to be defined by

Velocity:

$$u_* = (\tau/\rho)^{1/2}\text{—friction velocity} \tag{13}$$

Temperature:

$$T_* = \frac{-Q}{\kappa u_*} \tag{14}$$

Length:

$$L = \frac{-u^3}{\kappa\beta Q}\text{—scale Monin–Obukhov,} \tag{15}$$

where κ is the Karman constant.

According to M–O theory non-dimensional gradients of physical parameters in the constant fluxes' layer can be expressed by means of a function of the non-dimensional parameter z/L:

$$\frac{\kappa z}{u_*}\frac{\partial u}{\partial z} = \varphi(z/L), \quad \frac{z}{T_*}\frac{\partial \theta}{\partial z} = \varphi(z/L). \tag{16}$$

If we define the scale of particle concentration by

$$N_* = \frac{F_N}{u_*}, \tag{17}$$

non-dimensional aerosol concentration gradient can be expressed by the universal function z/L:

$$\frac{z}{N_*}\frac{\partial N}{\partial z} = \Phi(z/L). \tag{18}$$

Thus

$$N(z_2) - N(z_1) = N_*[f(z_2/L) - f(z_1/L)], \tag{19}$$

where f is primary function $z^{-1}\Phi(z/L)$.

The exact form of function f is not known, although in the literature there are several different formulae. However, the M–O theory provides its asymptotic forms ($z/L \to 0$ gives $f \to \ln|z/L|$). Therefore, in most cases, for the atmospheric surface layer over sea, the following formula can be used:

$$N(z) = N_* \ln(z) + C. \tag{20}$$

5. Measurements

The aerosol vertical concentration measurements were performed during two Arctic scientific cruises carried out in July and August 2000 and 2001, and during cruises to the Baltic sea in October 2000, and May and October 2001. The Baltic cruises were especially planned to study marine aerosol. The aerosol concentrations were measured by a laser particles counter (PMS model CSASP-100_HV) deployed on a mast of r/y Oceania in a way so that the instrument could be moved vertically.

The measurements were taken at 5 levels, 8, 11, 14, 17, and 20 m above the sea level. A measurement at each level lasted 2 min. Every measuring cycle consisted of at least 4 measurements at each level. Results have been averaged at every level summing data from 4 measurements, so the averaging time at each level was about 8 min and each aerosol concentration profile was based on about 1 h measurements. Thus, we averaged over nearly the whole spectrum of eddies that contribute to the turbulent transport.

Considering the power spectrum of horizontal wind speed, first time published by Van der Hoven (1957), and later confirmed by many researchers, we can see that turbulent fluxes are only weakly sensitive to a change in the averaging time ranging from several to tens of minutes. The same data allow us to conclude that the optimum averaging time is 67 min (Laihtman, 1970). An application of the averaging time shorter than the optimum results in an error in the flux determination. However, this error is relatively small and does not exceed 20%. Because during the experiment, I had a single particle counter, I had to use relatively short averaging time.

The instrument detected particles of diameters from 0.5 to 36 μm. The instrument bin width equals 1 μm.

6. Results

Results have been averaged at every level summing data from 4 measurements. Mean concentration profiles $n(r,z)$ have been calculated, then $n(r,z)$ was converted to $n_{80}\ (r,z)$ size distribution at 80% relative humidity using Fitzgerald's (1975) formula. Examples of mean aerosol concentration vertical profiles are presented in Fig. 2 (data from 21st May 2001, Baltic Sea, wind speed $U_{10} = 9.5$ m/s). The lines connect points of the concentration value at given level. Each line represents a different concentration range. The largest stars represent particles concentration in the whole given range e.g. 0.5–10 μm. The bold straight line on the Fig. 2 is the function $N(z) = N\ln(z) + C$ determined by least squares method. As one may assume (according to Formula (20)), it clearly follows from Fig. 2 that vertical concentration profiles may be approximated by straight lines on plane $\ln(z); n(z)$. The smaller the particles are, the easier the approximation by a straight line is. The increase of measurement points dispersion with increasing size of particles may also be due to the short time of averaging. Out of necessity, as we had only one particle counter, the averaging time was relatively short and it could have been too short for big particles of concentrations few orders smaller than 1 μm particles.

The N_* scale calculated on the basis of four cruises are shown in Fig. 3. The vertical axis on all 3 graphs represents N_* scale value, the horizontal one represents a size r and the serial number of the measurement. As it can be seen from the Fig. 3 almost in every case N_* is negative meaning

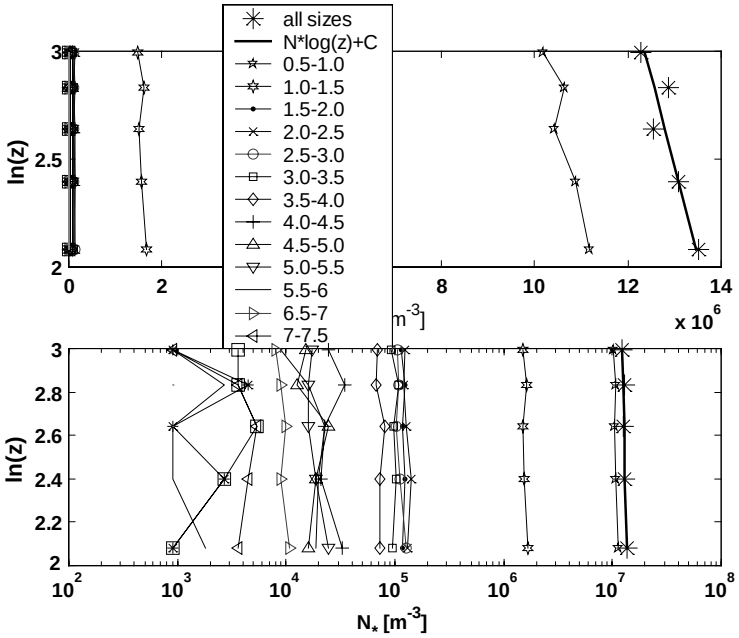

Fig. 2. Vertical Profiles of aerosol concentration. The lines connect points of concentration value at given level. Each line and kind of marker represents concentration of particle different size (in μm). The largest marker (star) represent total particles concentration in the given range, e.g. 0.5–10 μm.

that there are negative gradients of aerosol particle concentration in the atmospheric surface layer caused by the emission from sea surface. Individual departures from this rule can be explained by measurement error or advection impact. One can also notice that the biggest variation of N_* has been achieved for the smallest particles (from 0.5 to 1 μm range), which for conditions of horizontal homogeneity are the most difficult to find.

Aerosol fluxes have been calculated after selection of N_* scales and rejection from data file of values from which $n(z)$ profile could not be presented by formula (20). Based on definitions (17) and (10), aerosol flux can be calculated by the function

$$F = N_* \sqrt{C_{10}} U_{10}. \tag{21}$$

The obtained values are characterized by significant point dispersion and low correlation with wind speed. Their values are situated below the centre of the convergence range of sea aerosol generation functions known from literature (Andreas, 1998). An example of calculated aerosol fluxes for the Baltic Sea and Arctic seas is presented in Fig. 4. For comparison, aerosol concentrations at 5 m a.s.l. based on 4 polar cruises data are shown in Fig. 5. One can see that aerosol concentrations are less correlated with wind speed than fluxes presented in Fig. 4. Large dispersion of values in Figs. 4 and 5 is probably caused by the fact that wind speed is not the only factor influencing the marine aerosol concentration. The relationship between wind speed and volume flux $(4\pi r^3/3\mathrm{d}F/\mathrm{d}r)$ of the particles of size 1–10 μm is presented in Fig. 6. Mass flux of the particles from this range is shown in Fig. 7. Assuming that the particles consist of sea salt, fluxes in the above described

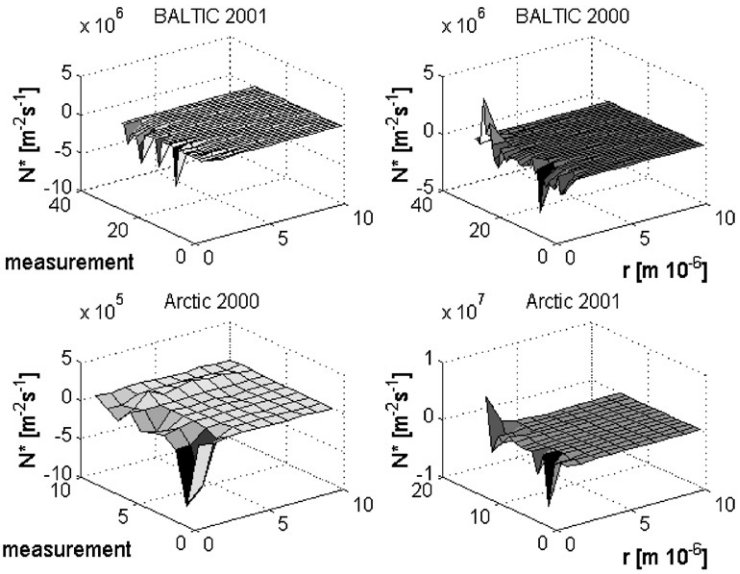

Fig. 3. Scale N_* of aerosol concentration. Each graph represents result from one cruise. The vertical axis on all 4 graphs represents N_* scale value, as a function of particle radius for the all measurements in single cruise (left horizontal axis is the sequential number of the measurement).

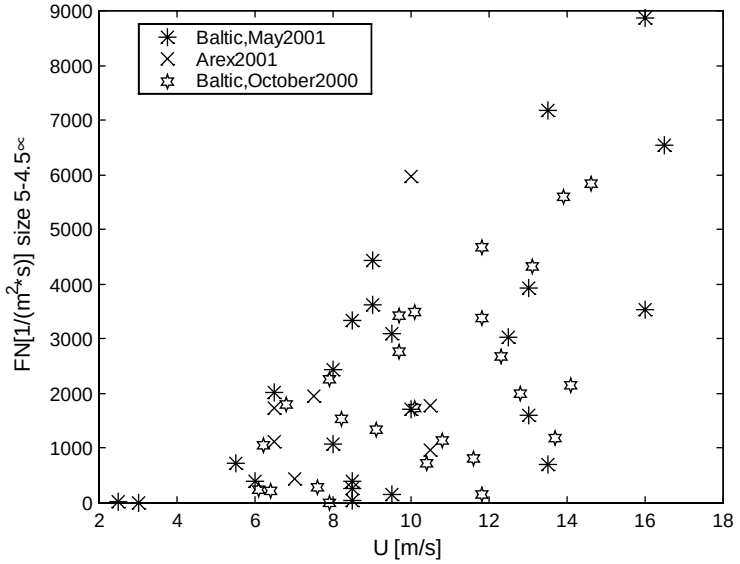

Fig. 4. Number particle flux versus wind speed (for size range 4.5–5.5 μm, Arctic Seas and Baltic Sea).

experiments did not exceed 50 μg/(sm^2) while sea salt fluxes emitted from the coast zone in the BAEX experiment reached 500 μg/(sm^2). The obtained fluxes converted into particles surface fluxes $FS = 4\pi \int_1^{10} r^2(dF/dr)dr$ are shown in Fig. 8. Andreas (1998) showed that FS fluxes obtained by

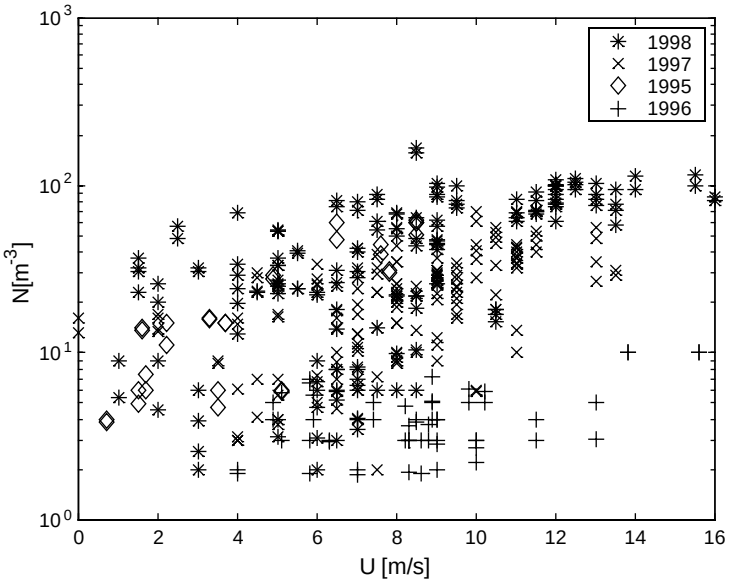

Fig. 5. Particle concentration versus wind speed (for size range 4.5–5.5 μm, Arctic Seas).

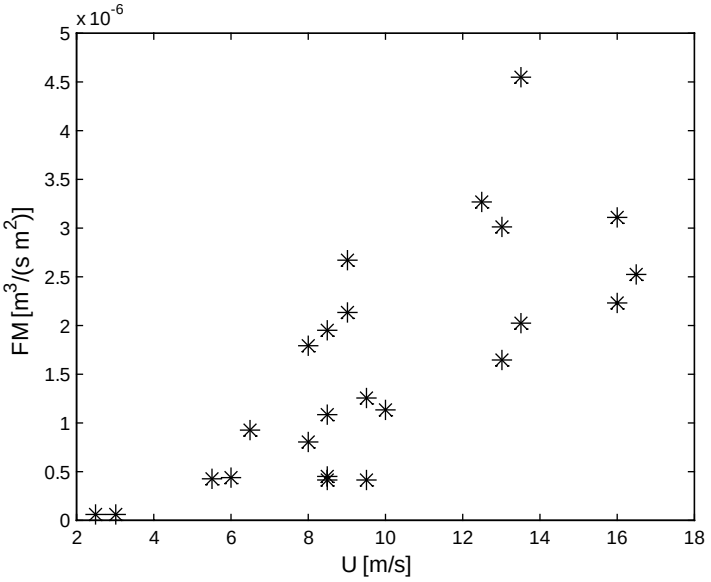

Fig. 6. Particle Volume Flux versus wind speed (Baltic 2000/2001).

many authors are proportional to the third power of friction speed. Similarly, our results may be approximated by function proportional to U_*^3 (curve in Fig. 8). As expected, resultant $\mathrm{d}F/\mathrm{d}r$ values are smaller than the results from Smith et al. (1993) model. Data from the Baltic experiments,

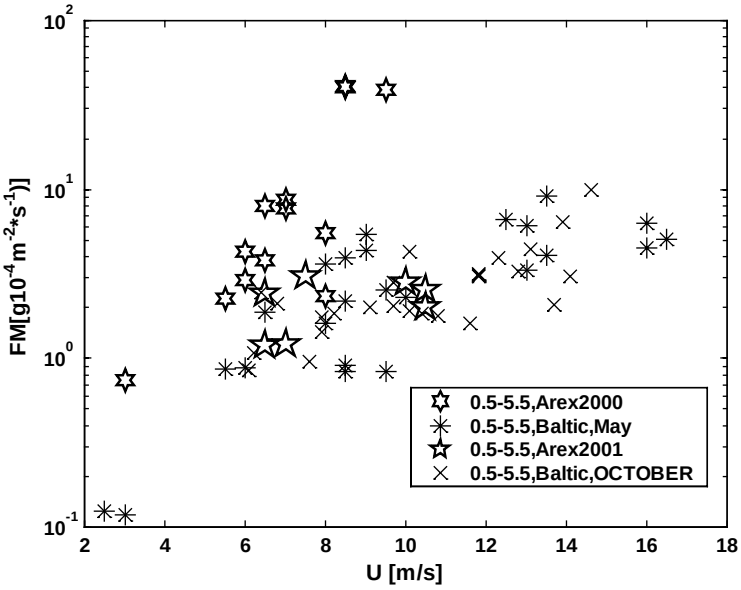

Fig. 7. Particle mass flux versus wind speed.

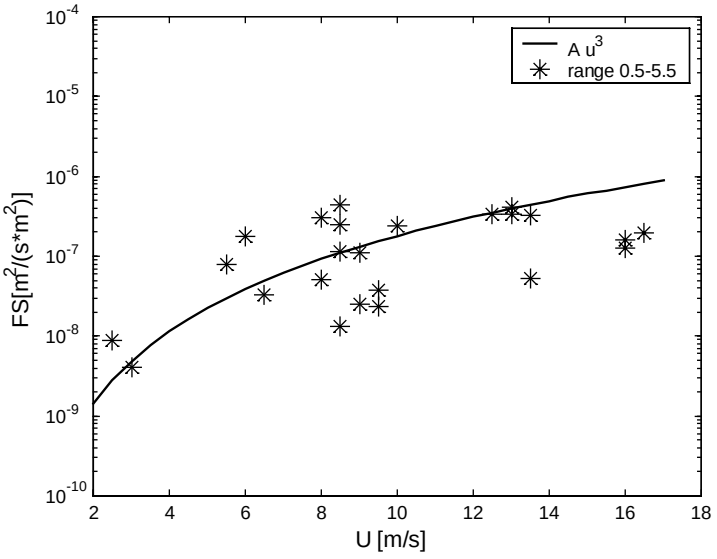

Fig. 8. Particle Surface Area Flux (Baltic 2000/2001).

calculated at different wind speed are shown in Fig. 9. Stars represent fluxes values, red line represents Smith's model results at wind speed 10 m/s, green line connects mean values of the fluxes calculated by this author.

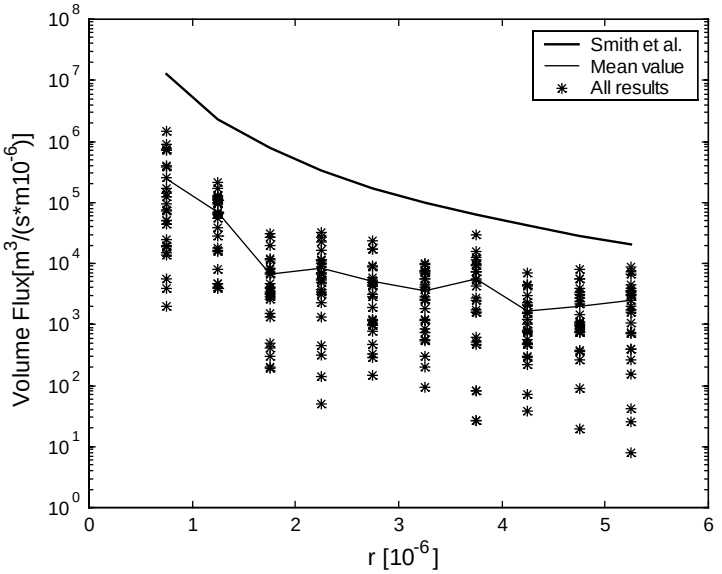

Fig. 9. Sea spray generation function as a volume flux, that is, as $4\pi r^3/3\,\mathrm{d}F/\mathrm{d}r$.

7. Conclusions

Breaking of the wind waves can be regarded as the main reason of the aerosol emission from the sea surface. This process is directly related to the wind speed in the case of the deep sea. Therefore aerosol emission from the sea surface should be well correlated to the wind speed.

On the other side, results of our measurements indicate that the correlation between an aerosol concentration in the atmospheric surface layer over sea and the wind speed is rather weak (Fig. 5). Thus, we conclude that it is not possible to obtain a good estimation of aerosol emission only on the basis of the aerosol concentration and deposition velocity.

An aerosol flux from the sea surface can be determined from the vertical gradient of aerosol concentration. As it was shown, there is a range of aerosol sizes for which this vertical gradient is entirely defined by the aerosol emission from the sea surface.

Fluxes of the aerosol emission, calculated from the measured vertical gradients of the aerosol concentration, range between 10^3 and 10^7 [$\mathrm{m}^{-2}\ \mathrm{s}^{-1}$]. They are one order of magnitude lower than the ones estimated by the model by Smith et al. (1993), which for the same range of dimensions results in fluxes between 10^4 and 10^8 ($\mathrm{m}^{-2}\ \mathrm{s}^{-1}$).

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