Studies of vertical coarse aerosol fluxes in the boundary layer over the Baltic Sea*

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Abstract

The results of studies of the vertical gradient of aerosol concentration measurements made during cruises of r/v 'Oceania' between 2008 and 2012 are presented. Using the results from those experiments, sea spray emission fluxes were calculated for all particles of sizes in the range from 0.5 μm to 8 μm, as well as for particles of sizes from fifteen channels of 0.5 μm width. The information obtained was further used to calculate the Sea Salt Generation Function (SSGF) for the Baltic Sea depending on the wind speed and the aerosol size distribution.

1. Introduction

The latest reports on Sea Spray Aerosols (SSA) indicate that the level of knowledge in this field is still insufficient (Vignati et al. 2010, de Leeuw et al. 2011, Tsigaridis et al. 2013). New findings have been reported practically every year: e.g. the influence of the organic fraction on SSA has been suggested in recent years (Modini et al. 2010, Westervelt et al. 2012). The development of computer models of the global climate requires more detailed information about the importance of SSA in these models. One of the parameters that describes the generation of SSA in the atmosphere is the Sea Salt Generation Function (SSGF).

The dependence of SSA on parameters such as wind speed or particle radius has been studied by many authors (Monahan 1988, Smith et al. 1993, Andreas 1998, Zieliński & Zieliński 2002, Gong 2003, Zieliński 2004, Petelski & Piskozub 2006, Keene et al. 2007, Kudryavtsev & Makin 2009, Long et al. 2011, Norris et al. 2012). One of the methods for investigating aerosol fluxes involves the Gradient Method (GM) (Petelski 2003, Petelski 2005, Petelski et al. 2005, Petelski & Piskozub 2006). Very little research has been done on the topic of SSGF from the surface of the Baltic Sea (Chomka & Petelski 1996, Chomka & Petelski 1997, Massel 2007) and thus any new insights based on aerosol studies in this region are of great importance to global studies.

A new approach to the SSGF was suggested by Andreas et al. (2010). On the basis of a reliable estimate of the SSGF, they calculated the effective production rate for droplets with initial radii from 5 to 300 μm. Most publications associated SSA flux with wind speed or friction velocity. Later, Veron et al. (2012) described a sea spray concentration function for spume droplets under high wind speed conditions. This work suggests that supra-millimetre droplets are more important than had been earlier predicted. What is more, this work describes the observation of liquid sheets forming at the crests of breaking waves, which is an earlier unreported SSA generation mechanism. Another interesting parameterisation was proposed

by Ovadnevaitte et al. (2014), where the friction velocity was replaced by the Reynolds number for a multimodal aerosol flux.

The vast majority of the SSA flux literature relates to measurements in the open ocean. Aerosol measurements in the Baltic Sea are valuable since its waters differ substantially from oceanic waters. The Baltic is one of the largest inland brackish seas by area, where major inflows of oceanic waters are rare. Waves on the Baltic Sea surface have relatively shorter lifetimes compared with ocean waves. The SSA coarse mode is produced by wave crashing and bubble bursting, and these mechanisms are strongly correlated with wind speed. The influence of wind speed and air masses on SSA concentrations in the Baltic region have been studied by a number of researchers (Zieliński & Zieliński 2002, Petelski & Piskozub 2006, Lewandowska & Falkowska 2013).

The prevailing winds in the southern Baltic Sea are westerlies. Such a circulation is determined by the transport of fresh maritime polar air masses (Leppäranta & Myrberg 2009), creating strong wind conditions related to the movements of low pressure systems from the Atlantic Ocean. Zdun et al. (2011) showed wind direction to have a strong influence on aerosol optical properties in the Baltic Sea region. Byčenkienė et al. (2013) demonstrated that the marine boundary layer is not seriously affected by long-range transport but that local transport of air pollution is an important factor. Thus, averaged SSA concentrations and size distributions in the Baltic Sea region are very valuable.

All processes responsible for SSA emission from the sea surface, like bubble bursting (Blanchard 1963) or the direct tearing of wave crests (Monahan et al. 1986), are related to the composition of sea surface water. Surface active agents (surfactants) significantly reduce surface tension (Rosen et al. 2012). The role of surfactants in SSA flux has been widely described, inter alia by Sellegri et al. (2006), Modini et al. (2010) and Long et al. (2011). The Baltic Sea is a drainage basin for a large area, which is why the composition of surfactants differs significantly from that of ocean areas (Drozdowska et al. 2013, Drozdowska & Fateyeva 2013). This is indicated by coloured dissolved organic matter (CDOM) measurements (Schwarz et al. 2002, Kowalczuk et al. 2003, Kowalczuk et al. 2010). There are also significant differences between the waters of the Baltic Sea and the Nordic Seasin laser-induced fluorescence spectra (Drozdowska 2007).

In this study we calculated profiles of vertical sea spray fluxes in the near-water layer on the basis of the averaged vertical concentration and the Monin-Obukhov theory. Using these fluxes we calculated the Sea Salt Generation Function (SSGF) over the Baltic Sea. This function provides

information on the emission of particles of different sizes, depending on environmental parameters.

2. Study area and measurement method

Data were collected during fourteen measurement days over the period between 2008 and 2012. Figure 1 shows the location of the measurement stations. A CSASP-100-HV laser particle counter was used to measure vertical aerosol concentrations (Petelski 2005). A detailed description of the CSASP-100-HV probe is given by Zieliński (2004). The measurements were made at five elevations: 8, 11, 14, 17 and 20 m above sea level, with a single measurement at each level lasting 2 minutes. The vertical aerosol concentration gradient was obtained from a minimum of 4 measurement series. Thus each result consists of a 1 hour series with an average sampling time at each elevation of 8 minutes. This gives 40 minutes of sampling; the other 20 minutes were lost on moving the probe from one level to another.

With regard to the vertical wind speed spectrum one sees that the magnitude of the turbulent flux is not very sensitive to the averaging time in ranges from a few to several dozen minutes (Van der Hoven 1957). Using the van der Hoven data one can deduce that the optimal averaging time is 67 minutes (Leihtman 1970). Using a shorter than optimal time, it causes a maximum error of 20% in the flux calculation. Based on the average vertical aerosol concentration profiles, the vertical aerosol fluxes in the boundary layer can be calculated using the Monin-Obukhov (M-O)

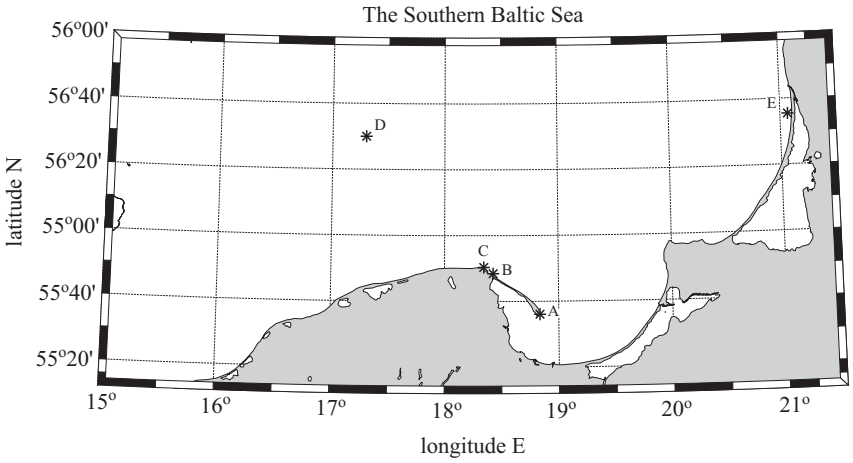

Figure 1. Locations of the measurement stations (5 points in the Baltic Sea). A: off the Hel Peninsula, B: off the town of Władysławowo, C: off Cape Rozewie, D: in the Southern Middle Bank region, E: off Klaipeda

theory (1953). A comprehensive methodology for such calculations has been presented by Petelski (2003).

To calculate the aerosol flux based on the M-O theory, we assumed that the particle concentration is a scalar property of the air. On this basis, for our range of aerosol sizes (0.5–8 μm) under the condition of horizontal uniformity, the vertical flux is equal to the emission from the sea surface. One can fully describe horizontal uniformity by using such parameters as momentum flux τ (expressed in [kg ms^{-2}]), sensible heat flux Q [W m^{-2}] and buoyancy parameter $\beta = g/T$ (g is the acceleration due to gravity $\approx$9.81 [m s^{-2}], T is the air temperature [K]). These parameters make it possible to define the following scales: Velocity (friction velocity): $u_* = (\tau/\rho)^{1/2}$, Temperature: $T_* = -Q/\kappa u_*$ and Length: $L = -u^3/\kappa\beta Q$ (ρ is the density of the air $\approx$1.29 [kg m^{-3}], $\kappa = 0.4$ is the dimensionless von Kármán constant). The scale of the particle concentration [1/m^3] is defined as:

$$N_* = F_N/u_*, \tag{1}$$

where F_N is the aerosol flux, defined as particle emission by the surface in time [1/m^2 s].

It is then possible to express the non-dimensional aerosol concentration gradient by the universal function of z/L:

$$(z/N_*)\partial N/\partial z = \Phi(z/L). \tag{2}$$

Using this formula the final equation can be derived using asymptotic forms from the M-O theory ($z/L \to 0$ gives $f \to \ln|z/L|$):

$$N(z) = N_* \ln(z) + C. \tag{3}$$

Measurements of the aerosol concentration at 5 elevations enabled N_* and thus the aerosol fluxes to be calculated.

The SSGF should describe SSA emission when the near-water boundary layer stratification is neutral, i.e. when a logarithmic profile of the SSA concentration exists ($z/L \to 0$). In such conditions positive (upward) fluxes can be measured. These fluxes were used in the subsequent parameterisation (see Figure 2).

In the literature both approaches for harmonising particle size are commonly used: the dry particle diameter (D_{dry}) and the wet radius (R_{80}) at 80% relative humidity (RH) (Ovadnevaitte et al. 2014). All the results presented in this paper were corrected to R_{80} (Fitzgerald 1975, Petelski 2005).

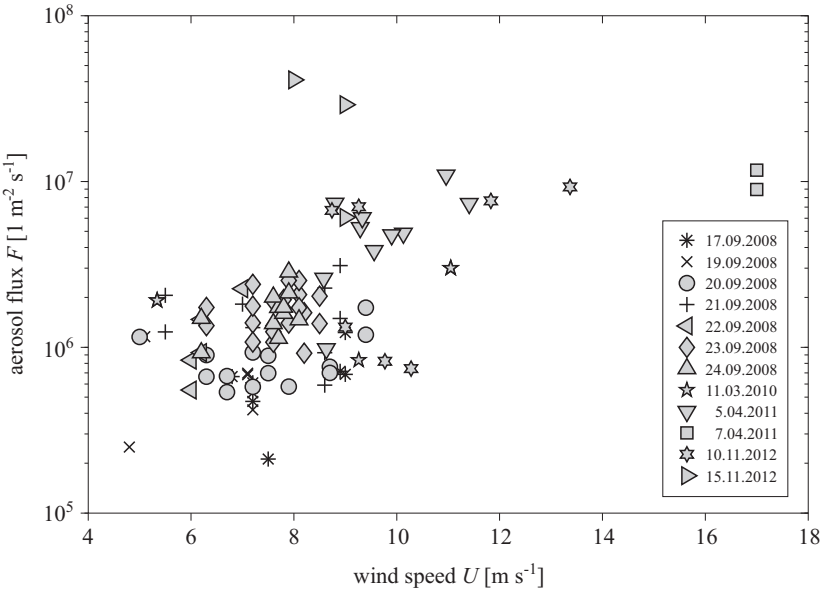

Figure 2. Estimated SSA fluxes. Each symbol represents a different day of measurements

3. Sea Salt Generation Function at the surface of the Baltic Sea

The purpose of determining the source functions is to show the correlation between the value of the marine aerosol emission and particle diameter: it depends on different environmental parameters. The sea salt emission depends on the amount of energy wind waves dissipate in the breaking process. This phenomenon is difficult to parameterise (Massel 2007), but as a first approximation one can use wind speed at 10 m elevation (U_{10}) for this. Hence, the designated function depends on the particle radius r and the wind speed U_{10}.

To derive the equation from the data gathered, fluxes not fulfilling the following criteria were rejected. Firstly, if during the daily measuring series we encountered both positive and negative fluxes, such a series was considered to be unreliable. Episodes with a negative flux may be caused by advection of local air pollution (Byčenkienė et al. 2013). Secondly, data gathered when the relative humidity was higher than 95% also were rejected. Finally, the correlation coefficient between the vertical gradient of SSA and the logarithm of the height provides information about the prevailing conditions similar to the regime of the Monin-Obukhov theory (Petelski 2003). Fluxes with correlation coefficients higher than or equal to 0.9 were accepted for further analysis.

The generation function $F(U, r)$ can be presented as the product of two functions $f_1(U)$ and $f_2(r)$:

$$F(U, r) = f_1(U) f_2(r), \tag{4}$$

where $f_1(U)$ represents the overall particle emission [1/m^2 s] and $f_2(r)$ represents particle sizes [1/μm].

Function $f_1(U)$ was found, using the least squares method, by fitting the aerosol flux values to the function $AU^2 + B$. The function was fitted to the values of total aerosol fluxes, i.e. to the mean flux for the full range of measured particle diameters. The use of the quadratic function of wind speed resulted from the fact that the highest correlations between aerosol fluxes and wind speeds were found for the quadratic power (Petelski et al. 2005); another commonly used dependence of power 3.51 was first presented by Monahan et al. (1986). The result of the fitting is shown in Figure 3. Here we see that the quadratic form has a higher coefficient of determination. The quadratic function has a zero for $U \approx 2.7$, whereas function $f(U^{3.41})$ has a zero for the negative value of the domain and intersect with the OY axis in $f(u^{3.41} = 0) = 1.2 \times 10^6$, which is why applying $f(U^2)$ is more realistic. The next argument in favour of using

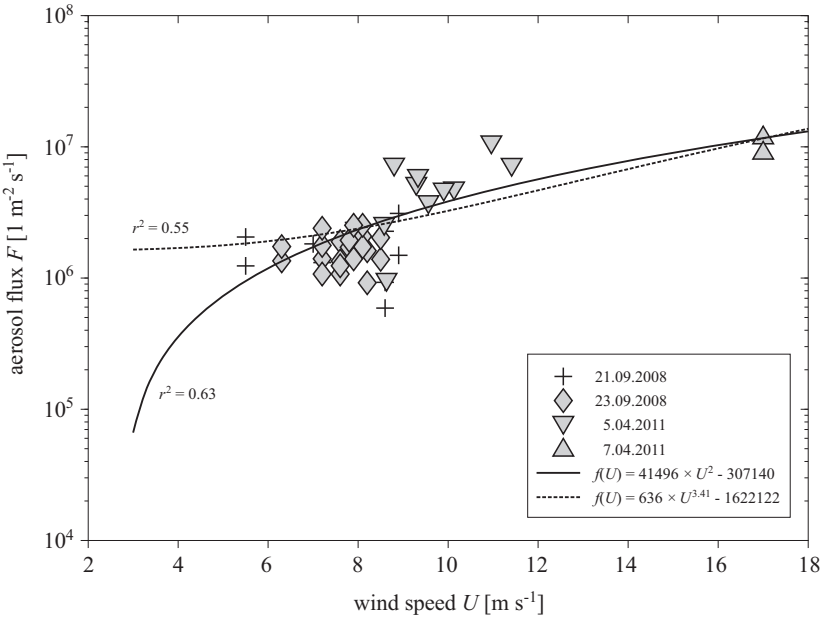

Figure 3. Aerosol flux versus wind speed after data selection (see text), with quadratic and 3.41 power functions fitted. For both functions the p-value is much less than 0.01; the functions are therefore statistically significant

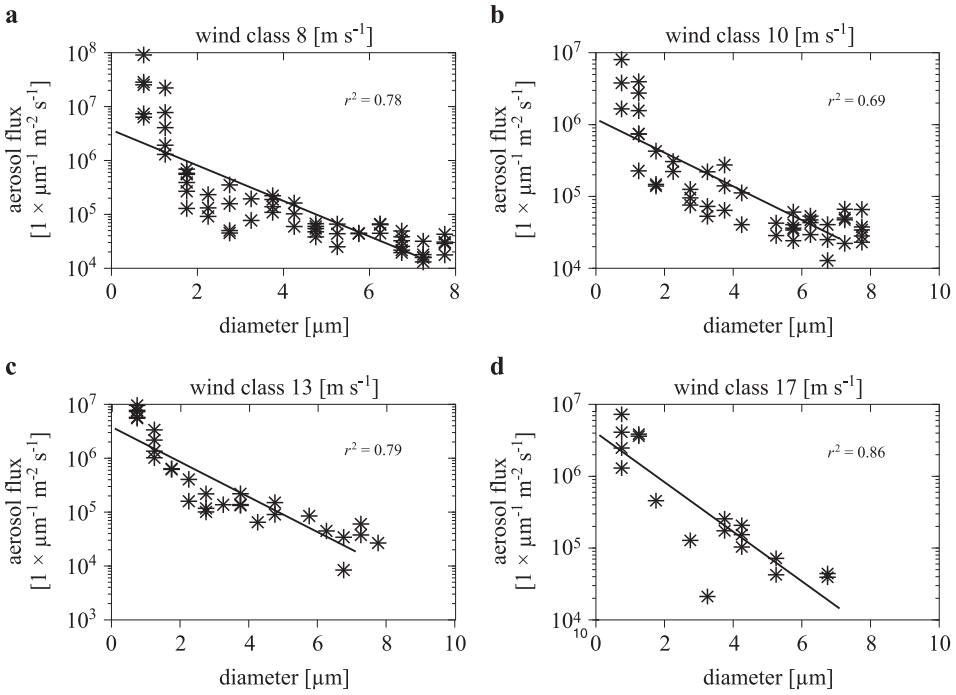

Figure 4. Examples of aerosol flux vs. particle diameter with the log-linear approximation applied for four wind classes (see text)

the quadratic dependence is the quadratic relation between aerosol optical depth (AOD) and wind speed with a strong correlation ($r^2 \sim 0.97$), as reported by Mulcahy et al. (2008) for clean marine conditions.In the following we will use the quadratic function.

The flux values presented in Figure 3, confirm the usefulness of the quadratic function for the fit. In this case as the first part of SSGF we propose:

$$f_1(U) = 41496 \times U^2 - 307140. \tag{5}$$

The next step in calculating SSGF is to find the dependence of the flux on the particle radius. In order to obtain function $f_2(r)$ the method suggested by Petelski & Piskozub (2006) was applied. The fluxes were classified into ten different wind speed ranges. Each series from the range of $U - 0.5$ m s^{-1} to $U + 0.5$ m s^{-1} was assigned to an integer wind speed U class. Figure 4 shows four examples for wind speeds of 8, 10, 13 and 17 m s^{-1}. In order to find the $f_2(r)$ equation for each class, a linear approximation in the $\ln(f_2)$, $2r$ space was used. For each wind speed the following function was fitted:

$$\ln[f_2(r)] = a2r + b, \tag{6}$$

where $f_2(r) = \exp(a2r + b)$, a and b are fitting coefficients. For each wind class there is one pair of coefficients.

In the subsequent calculations the average value of coefficient a was used ($a = -0.62$ μm). Factor b increases with wind speed, and this increase can be approximated with a linear function, although the results are rather scattered. In this case we have to change our approach. Data for the total fluxes of aerosol particles are statistically more reliable than each flux for one diameter range separately. Thus, instead of a linear function $b(U)$, we used a first-order fit of function $(AU^2 + B)$:

$$AU^2 + B = \int_{r_{\min}}^{\infty} \exp(-a2r + b)dr, \tag{7}$$

where $r_{\min} = 0.25$ μm is the radius of the smallest particle that is measureable with the instrument used in the study. From equation (6) one can obtain:

$$\exp(b) = [AU^2 + B]/[-2a\exp(a2r_{\min})]. \tag{8}$$

In this equation b is present as a function of wind speed. Using equation (8) in the exponential form of function f_2 in equation (6), we can derive a new form of the SSGF in which

$$\begin{aligned} f_1(U) &= AU^2 + B, \\ f_2(r) &= (-1/2a)\exp[2a(r - r_{\min})], \end{aligned} \tag{9}$$

where $A = 41496$ s m^{-4}, $B = -307140$ 1/m^2 s.

Hence, the function we are looking for is

$$F(U, r) = f_1(U)f_2(r) = (-\kappa/2a) \times (AU^2 + B) \times \exp[a2(r - r_{\min})]. \tag{10}$$

This function is valid for $U \geq 3$ m s^{-1}. In order to introduce the Andreas (2007) correction, the equation is multiplied by the von Kármán constant.

Finally, we propose the SSGF formula in the following form:

$$F(U, r) = 1.83 \times 10^4 \times U^2 - 1.35 \times 10^4)\exp(-1.24 \times r). \tag{11}$$

4. Conclusions

We present the results of calculations of the Sea Spray Generation Function (SSGF) for the Baltic Sea. The function depends on particle diameter and wind speed. Figure 5 shows particle fluxes and the SSGF for selected diameters. The SSGF fits well at both low and high wind speeds.

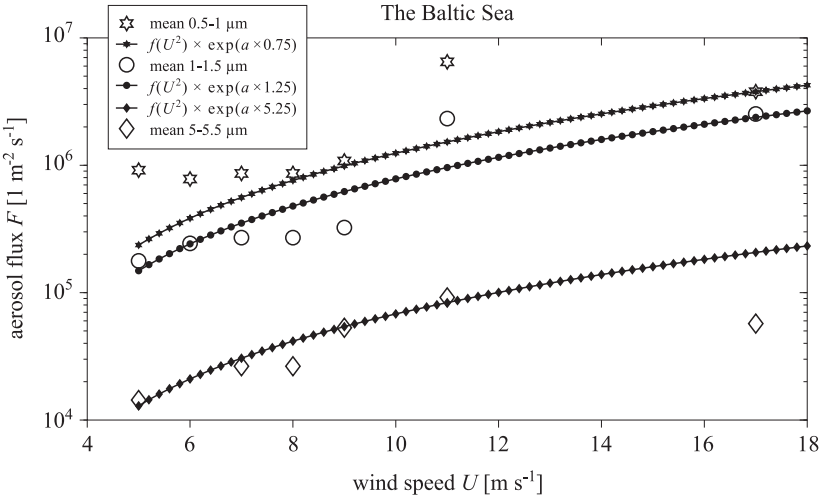

Figure 5. The final result of the SSGF fitting. The lines represent the predicted values of the SSGF for each particle diameter. The symbols represent the average fluxes from the measurements

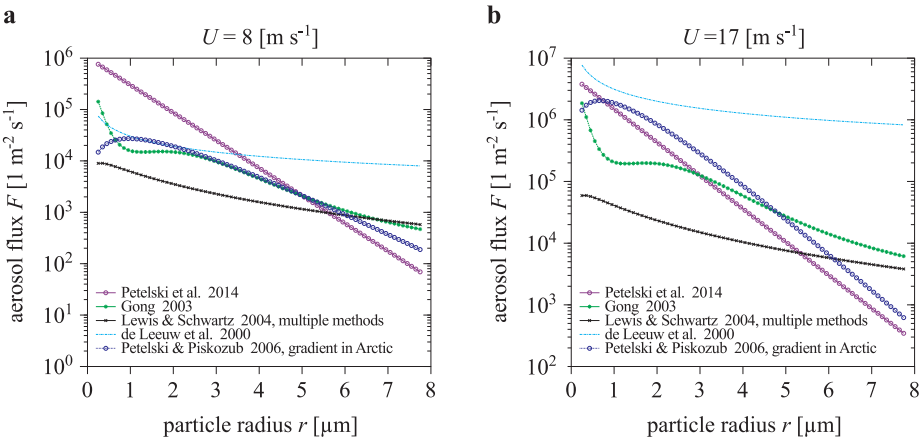

Figure 6. Comparison of our results with those obtained by other authors for two wind speed classes. The selected functions come from the work by de Leeuw et al. (2011)

The function $F(U, r)$ was also compared with other Sea Spray Generation Functions which were likewise expressed as functions of particle radius and wind speed (Figures 6a and b).

In order to avoid too much information in one graph, Figures 6a and b present only selected SSGFs: the de Leeuw et al. (2000) SSGF determined from the micrometeorological method (eddy correlation), Gong's function

(Gong 2003), which is based on Monahan's research, and the Lewis and Schwartz function (Lewis & Schwartz 2004), a function based on multiple methodologies.

Figure 6 also shows the Petelski & Piskozub (2006) function (with the Andreas (2007) modification) based on gradient measurements in the Arctic region. Here we see that there are differences between both gradient measurements, which are closely associated with the region where the measurements were made. That is why a separate function for the Baltic Sea is important for improving the quality of regional atmospheric and air-sea interaction models.

Most of the functions based on Monahan's work from 1986 were based on the Whitecap Method. The SSGF is independent of that method and is based on the micrometeorological method. The postulated quadratic dependence seems to be more justified with regard to AOD measurements (Mulcahy et al. 2008).

Since there has not been much research carried out to date on Sea Surface Generation Functions for marine basins like the Baltic Sea, our findings represent a significant contribution to the field of air-sea interaction studies, and should prove especially valuable for local use.

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