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SHORT COMMUNICATION

Sea spray aerosol flux estimation based on long-term variation of wave statistics[☆]

Dag Myrhaug^{*}, Hong Wang, Lars Erik Holmedal

Department of Marine Technology, Norwegian University of Science and Technology (NTNU), Trondheim, Norway

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Bivariate distributions

Summary This note provides estimates of the mean sea spray aerosol flux based on long-term wave statistics using the whitecap method based on the limiting steepness and threshold vertical acceleration criteria. The aim is to present a procedure demonstrating how global wave statistics can be used to give estimates of the long-term aerosol flux. These estimates are obtained by using bivariate distributions of significant wave height and characteristic wave period, representing open ocean deep water waves in the Northern North Sea and the North Atlantic.

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1. Introduction

The air-sea interface couples the atmosphere and the ocean by exchanging heat, momentum and water, and is crucial for understanding the Earth's climate. For highly energetic flow conditions the surface waves become unstable, and they will break. Wave breaking plays a major role in sea surface spray aerosol production. The ocean is a main source for water and

aerosols in the atmosphere, hence playing a key role in the climate control of the Earth. There are two main aerosol production mechanisms; the bursting of bubbles formed and dispersed primarily by breaking waves, and droplets directly driven by the wind from the spume off the wave crest (see e.g. [Massel \(2007\)](#), Figs. 8.1 and 8.2, respectively); the first production mechanism is considered in the present work.

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^{*} Corresponding author at: NTNU, Otto Nielsens vei 10, NO-7491 Trondheim, Norway. Tel.: +47 73 59 55 27.

E-mail addresses: dag.myrhaug@ntnu.no (D. Myrhaug), hong.wang@ntnu.no (H. Wang), lars.erik.holmedal@ntnu.no (L.E. Holmedal).

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Different methods of determining the sea spray aerosol production fluxes exist: The steady state dry deposition method, statistical method, deposition method, micro-meteorological methods (eddy correlation and gradient methods), and the whitecap method (see de Leeuw et al. (2011) for more details and a critical review). Despite a better understanding of the sea spray aerosol production fluxes, there is a large uncertainty in the predictions. Present applications of these different methods in global climate models have revealed a spread of nearly two orders of magnitude in the estimates of global sea spray aerosol emissions (de Leeuw et al., 2011). Moreover, de Leeuw et al. (2011) found that sea spray contains organic particles in addition to sea salt, especially for very small particles. More details and reviews are given in Massel (2007) and de Leeuw et al. (2011).

This note applies the whitecap method to give estimates of the sea spray aerosol fluxes based on long-term wave statistics, using the link between the whitecap coverage and the sea state parameters significant wave height and spectral peak frequency as given by Massel (2007). This makes it possible to relate the aerosol fluxes to specific sea locations and to seasonal variations. Here the results are exemplified by using long-term wave statistics from deep water sites in the Northern North Sea and the North Atlantic. The whitecap method used herein is based on the limiting steepness and threshold vertical acceleration criteria (Massel, 2007). The main purpose is to provide a procedure which can be applied to systematically compare the aerosol fluxes at different locations based on the long-term statistical information of the wave climate.

2. Background

Here the sea spray aerosol flux is estimated by using the whitecap (wc) method based on the limiting steepness and threshold vertical acceleration criteria. Following Massel (2007, Ch. 10.5), the sea spray generation function according to the whitecap method, $f_{sgf}^{(wc)}$, is given by

$$f_{sgf}^{(wc)}(r) = f_{prod}^{(wc)}(r) \cdot F_{cov}, \quad (1)$$

where $f_{prod}^{(wc)}(r)$ is the size-dependent aerosol production flux, and F_{cov} is the whitecap coverage. The Woolf et al. (1988) formula for $f_{prod}^{(wc)}(r)$ is adopted

$$f_{prod}^{(wc)}(r) = \exp[16.1 - 3.43 \log_{10} r - 2.49(\log_{10} r)^2 + 1.211(\log_{10} r)^3], \quad (2)$$

where the droplet radius r is taken to represent r_{80} , i.e. the droplet radius in equilibrium with the atmosphere at a given ambient relative humidity of 80% given in μm ($=10^{-6}$ m). Here Eq. (2) is valid for r in the range 0.8–10 μm . It should be noted that for sea salt particles originating from sea water with typical salinity 0.034–0.036, the droplet radius is also approximated by the radius at its formation, $r_0 = 2r_{80}$, and by its volume-equivalent dry radius, $r_{dry} = 0.5r_{80}$ (de Leeuw et al., 2011; Massel, 2007). Here Eq. (2) represents an upper estimate of the size-dependent aerosol production flux compared with those given e.g. by Monahan et al. (1986) and Monahan (1988) (see Massel (2007), Figs. 9.3, 9.4). Moreover,

the parameterizations of F_{cov} given in Massel (2007, Ch. 10.5) are adopted:

Limiting steepness criterion

$$F_{cov} = \exp \left[-0.1933 \left(\frac{H_s \omega_p^2}{g} \right)^{-2} \right]. \quad (3)$$

Threshold vertical acceleration criterion

$$F_{cov} = 1 - \Phi \left[0.447 \left(\frac{H_s \omega_p^2}{g} \right)^{-1} \right]. \quad (4)$$

Here H_s is the significant wave height, $\omega_p = 2\pi/T_p$ is the spectral peak frequency, T_p is the spectral peak period, g is the acceleration due to gravity, and Φ is the standard Gaussian cumulative distribution function (*cdf*) given by

$$\Phi(x) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^x \exp\left(-\frac{1}{2}t^2\right) dt. \quad (5)$$

Further details are given in Massel (2007).

Thus the sea spray generation function in Eq. (1) is defined in terms of r and the sea state parameters H_s and T_p . If, for example, the mean zero-crossing wave period T_z is given, then T_p is related to T_z by e.g. $T_p = \alpha T_z$ where α is a constant. Here Eq. (1) is written as

$$f_{sgf}^{(wc)}(r, H_s, T) = f_{prod}^{(wc)}(r) F_{cov}(H_s, T), \quad (6)$$

where T represents T_p or T_z .

The results presented in Section 4 are based on taking $T = T_p$ and for $T = T_z$, $T_p = \alpha T_z$ with $\alpha = 1.28$ in Eqs. (3) and (4), where the latter is taken from Myrhaug and Kjeldsen (1987, Fig. 11) for a JONSWAP spectrum with peakedness factor $\gamma = 3.3$. However, it should be noted that this relationship between T_p and T_z is not necessarily valid for the data sets used here; these might contain mixed swell and wind sea for which other relationships exist.

3. Long-term aerosol flux estimation

A quantity of interest is the expected (mean) value of the sea spray generation function based on long-term variation of H_s and T_p (or T_z), i.e.

$$E[f_{sgf}^{(wc)}(r, H_s, T)] = f_{prod}^{(wc)}(r) E[F_{cov}(H_s, T)], \quad (7)$$

where

$$E[F_{cov}(H_s, T)] = \int_{H_s} \int_T F_{cov}(H_s, T) p(H_s, T) dH_s dT. \quad (8)$$

Here $p(H_s, T)$ is the joint probability density function (*pdf*) of H_s and T (i.e. T_p or T_z).

Different parametric models for the joint *pdf* of H_s and T_p or H_s and T_z , are given in the literature. Examples are Haver (1985) and Moan et al. (2005) (hereafter referred to as MGAU05) for H_s and T_p ; Mathisen and Bitner-Gregersen (1990) and Bitner-Gregersen and Guedes Soares (2007) (hereafter referred to as BGG07) for H_s and T_z . In the present note, the aerosol flux estimation is exemplified by using the joint *pdf* of H_s and T_p proposed by MGAU05 based upon 29 years of wave data in the Northern North Sea, and the

joint *pdf* of H_s and T_z proposed by BGG507 based upon five data sets from the North Atlantic. These *pdfs* are given as

$$p(H_s, T) = p(T|H_s)p(H_s), \quad (9)$$

where $p(H_s)$ is the marginal *pdf* of H_s , which for the MGAU05 model is given by the following combined lognormal and Weibull distributions (this type of distribution was first suggested by Haver (1985))

$$p(H_s) = \begin{cases} \frac{1}{\sqrt{2\pi\kappa}H_s} \exp\left[-\frac{(\ln H_s - \theta)^2}{2\kappa^2}\right], & H_s \leq 3.25 \text{ m} \\ \beta \frac{H_s^{\beta-1}}{\zeta^\beta} \exp\left[-\left(\frac{H_s}{\zeta}\right)^\beta\right], & H_s > 3.25 \text{ m} \end{cases}. \quad (10)$$

Here $\theta = 0.801$, $\kappa^2 = 0.371$ are the mean value and the variance, respectively, of $\ln H_s$ and $\zeta = 2.713$, $\beta = 1.531$ are the Weibull parameters.

For the BGG507 model, $p(H_s)$ is given by the following three-parameter Weibull distribution

$$p(H_s) = \frac{r}{s} \left(\frac{H_s - t}{s}\right)^{r-1} \exp\left[-\left(\frac{H_s - t}{s}\right)^r\right], \quad H_s \geq t, \quad (11)$$

where r , s and t are the Weibull parameters given in BGG507, see Table 1.

$p(T|H_s)$ is the conditional *pdf* of T given H_s , which for both models is given by the lognormal distribution

$$p(T|H_s) = \frac{1}{\sqrt{2\pi\sigma T}} \exp\left[-\frac{(\ln T - \mu)^2}{2\sigma^2}\right], \quad (12)$$

where μ and σ^2 are the mean value and the variance, respectively, of $\ln T$. For the MGAU05 model, $T = T_p$ and μ , σ^2 are given by Gao (2007) as

$$\mu = a_1 + a_2 H_s^{a_3}; \quad (a_1, a_2, a_3) = (1.780, 0.288, 0.474), \quad (13)$$

$$\sigma^2 = b_1 + b_2 e^{b_3 H_s}; \quad (b_1, b_2, b_3) = (0.001, 0.097, -0.255). \quad (14)$$

For the BGG507 model $T = T_z$ and

$$\mu = a_1 + a_2 H_s^{a_3}, \quad \sigma = b_1 + b_2 H_s^{b_3}, \quad (15)$$

where the parameters in μ , σ are given in BGG507, see Tables 2 and 3. All these data represent wave conditions in the North Atlantic. Data sets 1, 2 and 3 are numerically generated wave data taken from global databases representing 44 years (1958–2004) at 59°00'N, 19°00'W. Data set 4 refers to Global Wave Statistics (GWS) zone 9 (the zone located south of Iceland and west of UK) representing visual observations collected from ship in normal service all over the world in the period 1949–1986. Data set 5 refers to Juliet Shipborne Wave Recorder (SBWR) representing data

Table 1 Weibull parameters for H_s , see Eq. (11).

Data	s	r	t
Data set 1	3.104	1.357	0.906
Data set 2	2.848	1.419	1.021
Data set 3	2.939	1.240	0.896
Data set 4	2.857	1.449	0.838
Data set 5	2.420	1.169	1.258

Table 2 Mean value of $\ln T_z$, see Eq. (15).

Data	a_1	a_2	a_3
Data set 1	1.350	0.366	0.392
Data set 2	1.365	0.375	0.453
Data set 3	0.790	0.805	0.292
Data set 4	0.835	1.139	0.119
Data set 5	1.952	0.168	0.499

Table 3 Standard deviation of $\ln T_z$, see Eq. (15).

Data	b_1	b_2	b_3
Data set 1	0.020	0.165	−0.166
Data set 2	0.033	0.285	−0.752
Data set 3	0.055	0.195	−0.269
Data set 4	0.140	0.030	−0.958
Data set 5	0.070	0.066	−0.081

registered at the Ocean Weather Station Juliet during 13 years since 1952 at 52°00'N, 20°00'W. More details are given in BGG507.

The main differences between the parametric models used to exemplify the aerosol flux estimation (MGAU05 and BGG507) are the fraction of “steep sea states” contained in them, as will be discussed further in Section 4.

4. Results and discussion

Here the long-term aerosol flux is estimated based on the results given in Section 3. Thus the results are limited to aerosols produced by the bursting of bubbles formed and dispersed primarily by breaking waves, and for droplets with radii in the range 0.8–10 μm .

Figs. 1 and 2 show the expected (mean) aerosol volume flux, $E[f_{vol}(r)]$, versus the droplet radius $r = r_{80}$ in the range 0.8–10 μm for the six different distributions based on the limiting steepness criterion (criterion 1) (Fig. 1) and the threshold vertical acceleration criterion (criterion 2) (Fig. 2). The volume flux is obtained by multiplying the results in Eq. (7) by the factor $(4\pi/3)r^3$. As expected it appears that the volume flux increases as r increases. The corresponding values of the total expected volume aerosol flux (i.e. the values obtained by integrating the fluxes $E[f_{vol}(r)]$ given in Figs. 1 and 2 over the droplet radii (0.8–10 μm) are given in Table 4. These values confirm the visual impression provided by Figs. 1 and 2 with respect to the effect of using criteria 1 and 2, and to the relative ranking of the different sites. First, the results based on criterion 1 give larger fluxes than using criterion 2 except for BGG507 data set 5. Second, BGG507 data set 1 corresponds to the highest flux, BGG507 data set 5 gives the lowest flux, while the other data sets give intermediate volume flux values.

The results in Figs. 1 and 2 and Table 4 are obtained by multiplying the aerosol production flux, $f_{prod}^{(wc)}(r)$, with the expected value of the whitecap coverage, $E[F_{cov}(H_s, T)]$ (see Eqs. (7) and (8)). It should be noted that these results are

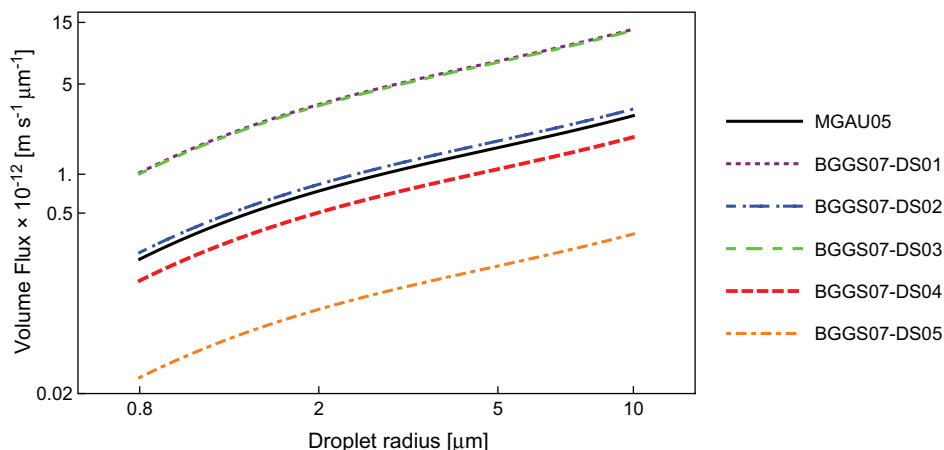


Figure 1 The expected (mean) value of the aerosol volume flux function, $E[f_{vol}(r)]$, versus r for the six different distributions based on the limiting steepness criterion (criterion 1), where r is r_{80} .

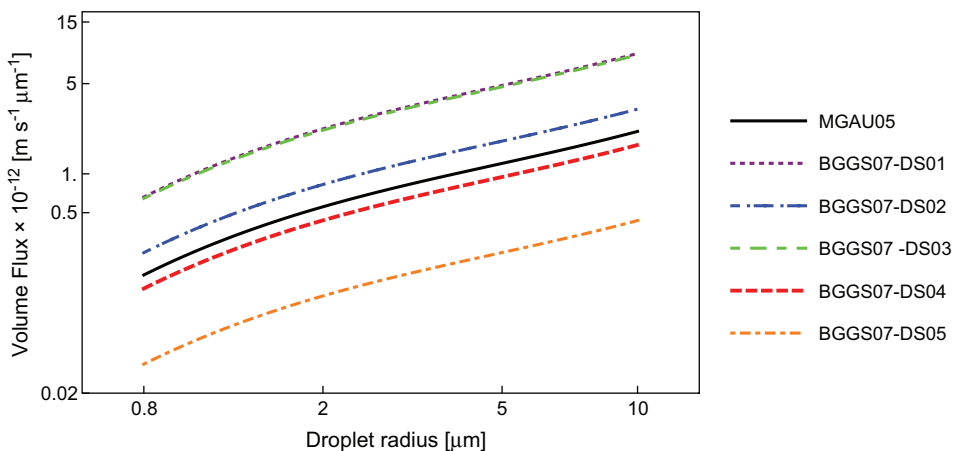


Figure 2 The expected (mean) value of the aerosol volume flux function, $E[f_{vol}(r)]$, versus r for the six different distributions based on the threshold vertical acceleration criterion (criterion 2), where r is r_{80} .

criteria- and site-dependent. Values of $E[F_{cov}(H_s, T)]$ are given in Table 5, showing the consistency with the results in Figs. 1 and 2 and Table 4. First, criterion 1 gives higher whitecap coverage than criterion 2 except for BGG07 data set 5. Second, BGG07 data set 1 corresponds to the highest

whitecap coverage, BGG07 data set 5 gives the lowest whitecap coverage, while the other data sets give intermediate values of the whitecap coverage.

The main differences between the parametric models (MGAU05 and BGG07) used to exemplify the application

Table 4 Comparison of total expected volume aerosol flux of r_{80} at six different sites.

Long-term distribution at site	$E[f_{vol}^{(tot)}] [\times 10^{-12} \text{ m s}^{-1}]$	
	Criterion 1	Criterion 2
MGAU05	15.3	11.5
BGG07 Data set 1	71.5	46.0
BGG07 Data set 2	17.2	17.1
BGG07 Data set 3	70.0	44.9
BGG07 Data set 4	10.4	9.0
BGG07 Data set 5	1.9	2.3

Table 5 Expected value of the whitecap coverage, $E[F_{cov}]$, at six different sites.

Long-term distribution at site	$E[F_{cov}(H_s, T)]$	
	Criterion 1	Criterion 2
MGAU05	7.67×10^{-3}	5.75×10^{-3}
BGG07 Data set 1	3.59×10^{-2}	2.31×10^{-2}
BGG07 Data set 2	8.64×10^{-3}	8.55×10^{-3}
BGG07 Data set 3	3.51×10^{-2}	2.25×10^{-2}
BGG07 Data set 4	5.23×10^{-3}	4.51×10^{-3}
BGG07 Data set 5	9.29×10^{-4}	1.17×10^{-3}

of the present procedure are those associated with the fraction of “steep sea states” contained in the models. Here “steep sea states” mean the combination of relatively large values of H_s and relatively low values of T (T_p or T_z) resulting in relatively large values of H_s/T^2 , which is directly related to $H_s\omega_p^2$ in Eqs. (3) and (4). As reflected in the results in Figs. 1 and 2 and Tables 4 and 5 it appears that BGG07 data set 1 contains the largest fraction of “steep sea states”, followed by, in decreasing order, BGG07 data sets 3, 2, MGAU05, BGG07 data sets 4 and 5.

5. Summary

Estimates of the long-term sea spray aerosol flux by using the whitecap method based on the limiting steepness and threshold vertical acceleration criteria are provided. This is obtained by adopting the criteria given in Massel (2007) and by using joint *pdf* models of significant wave height and characteristic wave period representing open ocean deep water wave data sets at six different sites in the Northern North Sea and the North Atlantic. The example estimates of the total mean volume aerosol flux at these sites ranges from about $2 \times 10^{-12} \text{ m s}^{-1}$ to $72 \times 10^{-12} \text{ m s}^{-1}$, where the droplet radius is r_{80} .

Overall, this work provides a procedure which can be applied to systematically compare the aerosol fluxes at different locations based on the long-term statistical information of the wave climate.

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