

The Discrete Whitecap Method for Estimating Sea Salt Aerosol Generation: A Reassessment

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1. Introduction

The Discrete Whitecap Method (DWM) for combining measurements made during, and immediately after, the decay of a laboratory whitecap with field measurements of oceanic whitecap coverage to estimate the rate of sea salt aerosol generation as a function of wind speed introduced by Monahan et al (1982,1986) has recently been described as “the most widely used source function” by Grythe et al (2014) in an extensive review of such sea-spray aerosol source functions. In the more than 30 years since this marine aerosol generation function was first described, numerous authors have made constructive suggestions as to how it could be improved, and many other authors have inadvertently misconstrued the significance of the three terms that make up this model (Eq.1).

$$\partial F_o / dr = W_B \cdot \tau^{-1} \cdot dE / dr \quad \text{Eq.1}$$

Here $\partial F_o / dr$ is the rate of marine aerosol generation, per unit area of sea surface, per unit increment of spray droplet radius (at 80% R.H.), and W_B represents the simple fraction of the sea surface covered by Stage B whitecaps, i.e. decaying foam patches in the usage of Bondur and Sharkov (1982). The symbol tau represents the e-folding time for the exponential decay of such a whitecap, and dE/dr is the laboratory-determined number of marine aerosol particles produced, per increment droplet radius, as a consequence of the demise of all the bubbles initially associated with a whitecap of known initial area.

It should be stressed at the outset that dE/dr encompasses the total production of sea spray droplets resulting from the decay of a laboratory whitecap, and that this production was observed to continue for many seconds beyond the time when the whitecap was no longer visible (Woolf et al, 1987). Thus the suggestion by some that dE/dr might represent in some fashion the average production during the lifetime of the optically resolvable whitecap is not accurate.

This reassessment of this discrete whitecap method begins with a brief clarification of the most-widely used variations of the general whitecap method, namely the continuous whitecap method and the discrete whitecap method. This

is followed by a review of the three terms that appear on the right-hand side of Eq. 1, taken one at a time, to consider to what extent, in light of recent advances, each of these terms can be improved.

Aerosol production (and its chemical composition) from breaking waves is an extremely complex function of wave and bubble physics, thin film fluid dynamics, foam dynamics, bulk water chemistry and biology, the physical, biological and chemical properties of the sea surface microlayer and its rate of formation and disruption, atmospheric chemistry, and turbulent atmospheric transport, and we do not attempt to address any of these factors in depth. Rather, the goal is to explore the conceptual interpretation of the discrete whitecap method because, as stated in the opening paragraph, it remains a widely used method to predict the global flux of aerosol particles from the ocean surface, with significant implications for global climate modelling.

2. Continuous Whitecap Method and Discrete Whitecap Method

The continuous whitecap method (CWM) and the discrete whitecap method (DWM) are two primary variants of the general whitecap method, and as their names suggest, they differ in terms of the mechanism whereby whitecaps are generated in the laboratory.

2.1 CWM

The CWM combines a measure of the number of aerosol particles produced per unit whitecap area per unit time as a function of particle radius ($\partial F_{wc}/\partial r$, $m^{-2} s^{-1} \mu m^{-1}$), with an estimate of the area of whitecap foam per unit sea surface area, also known as whitecap coverage (W , $m^2 m^{-2}$). As such, the total particle flux per unit sea surface is determined from knowledge of these two quantities, and can be written as

$$dF_o / dr = W \cdot \partial F_{wc} / \partial r \quad \text{Eq. 2}$$

Values of $\partial F_{wc}/\partial r$ have been determined by several investigators in the laboratory by measuring the flux of aerosol particles produced by (i) a continuous air-entraining waterfall, and by (ii) forcing air through sintered glass frits (e.g. Cipriano and Blanchard, 1981; Martensson et al., 2003). There are many whitecap coverage parameterizations in the literature, which can be used to scale laboratory results to oceanic conditions with the wind speed parameterization of Monahan and O'Muircheartaigh (1980) being the most widely used.

As noted in deLeeuw et al., (2011), one of the challenges inherent to the CWM is replicating the correct volume flux of air per unit whitecap area so that it adequately represents the rate of air degassing per unit whitecap area

representative of oceanic breaking waves. The fundamental assumptions inherent in the implementation of the CWM is that only those parts of the ocean surface covered in whitecap foam are capable of producing aerosol particles, that all whitecap areas are equally efficient producers of aerosol particles, and that the production rate is equal to that determined in the laboratory.

2.2 DWM

While somewhat similar in construction, the DWM is fundamentally different from the CWM, yet we believe that these differences have been inadvertently misconstrued, obscuring the interpretation of the individual terms in Eq. 1 above, and thus requiring a further clarification of the original concept behind the DWM.

Beginning with Monahan (1971), one underlying assumption of the DWM as proposed by Monahan et al. (1982), is that under steady state conditions, the rate of change of oceanic whitecap coverage is by definition equal to zero. The goal was therefore to formulate a model of aerosol production flux that combined measurements of the total number of size-resolved aerosol particles (dE/dr) produced by the total degassing of a laboratory bubble plume with initial Stage B whitecap area A_o , with the rate of production of whitecap area per unit sea surface area. The term dE/dr therefore represents the total number of size-resolved particles per whitecap area produced during the degassing of a bubble plume with associated initial whitecap area A_o , and has units $m^{-2} \mu m^{-1}$.

Following the assumption of steady state whitecap coverage, the rate of formation of Stage B whitecap area per unit area sea surface is equal to the rate of decay of Stage B whitecap area per unit area sea surface, and a first-order estimate of this exponential decay timescale (τ) was made in the experiments of Monahan et al. (1982). The rate of decay of stage B whitecap area per unit sea surface area was approximated as $W_B \tau^{-1}$, as written in Eq. 1 above.

To find a functional form for dE/dr , discrete individual laboratory whitecaps were generated in a whitecap simulation tank, which allowed the associated time-evolving bubble plume to form and decay in an attempt to replicate the inherently transient nature of individual whitecap evolution found in the open ocean. Measurements of the aerosol particles were made during the evolution of the discrete breaking wave, and continued to be made in the minutes after the optically resolvable whitecap foam had effectively disappeared from the water surface, but while small bubbles of radius roughly $100\mu m$ and less continued to rise to the surface and produce aerosol particles.

An important distinction therefore arises between the CWM and the original formulation of the DWM: the timescale in Eq. 1 is not associated with the timescale of aerosol particle production from bubble bursting within a breaking wave, but rather it quantifies the rate of production of whitecap area, for which

the total number of aerosol particles that will ultimately be produced per unit whitecap area is assumed to be known.

However, in light of recent advances and newly available data on the timescales associated with oceanic whitecaps, a revised whitecap timescale is now proposed to replace τ in Eq. 1 above. This is discussed more fully in section 5 below.

3. The dE/dr term, specifying the number of sea spray droplets, per droplet radius increment, that are produced are a consequence of a single laboratory breaking wave

Subsequent to the introduction of Eq. 1 at a conference in Galway in September 1983 (Monahan et al, 1986) a complementary expression had been presented in Bombannes in September 1985 (Monahan, 1986). This latter expression was cast in terms of the laboratory-determined number of marine aerosol particles produced, per increment droplet radius, as a result of the eventual destruction of all the bubbles associated with the new β -plume of known initial (maximum) volume, normalized by that initial volume, $\partial G/\partial r$.

This approach, as developed step by step in Monahan (1986), can be recast in terms of dE/dr , yielding Eq.3. where D is the e-folding depth associated with the attenuation with depth of the cross-sectional area of an oceanic β -plume, and D_L is the e-folding depth defining the attenuation with depth of the horizontal cross-sectional area of the β -plume as it initially appeared in the laboratory "whitecap simulation tank".

$$\partial F_o / dr = (W_B \cdot \tau^{-1}) (D / D_L) dE / dr \quad \text{Eq.3.}$$

One notes that the right-hand side of Eq.3 is the same as the right-hand side of Eq.1, but now multiplied by the ratio D/D_L , i.e. by the ratio of the appropriate scale depth for the field situation to the scale depth of the bubble plume generated in the laboratory whitecap simulation tank.

The authors who proposed Eq.1 (Monahan et al, 1986) as an approximation whereby $\partial F_o/\partial r$ could be calculated inferred from the work of Thorpe (1982) that a bubble plume could be described as a cloud that attenuated exponentially with depth, also were aware that this same author found that the e-folding depth of such a cloud, or certainly of the bubble population in the surface waters of the ocean, increased modestly with wind speed. Specifically, Fig. 14 in Thorpe (1982) indicates that the e-folding depth, D , of the acoustic cross-section per unit volume of the bubble cloud rich waters in the Firth of Lorne off shore at Oban increases by a only factor of 2.96 as the wind speed goes from 3.25 m/sec to 14.51 m/sec, while the Stage B whitecap coverage, assuming a cubic

dependence on wind speed, can be expected to increase by a factor of 89.0, for the same wind speed modification. Since the dependence of $D(U)$ on wind speed appeared to be insignificant when compared to the increase of $W_B(U)$ with wind speed, they ignored this dependence of D on U in their formulation of $\partial F_o/\partial r$.

The $\partial F_o/\partial r$ expression (Eq.3) found in Monahan (1986) allows for the inclusion of an explicit wind-dependent $D(U)$ in the estimation of sea surface marine aerosol flux. It should be noted that both Eq.1 and Eq.3 reflect the implicit assumption that the bubble size spectrum, and the bubble numbers per unit volume in the β -plume beneath a new Stage B whitecap, are constant regardless of the initial area of the Stage B whitecap, the initial volume of the β -plume, or the characteristic e-folding depth of that plume. It is likewise assumed in this treatment that the same size distribution and number of bubbles ultimately results in the production at the sea surface of the same number, and size distribution, of sea salt aerosols, but this is only an approximation, as the same set of bubbles can produce at the sea surface a varying population of sea salt aerosols, depending on the concentration of the major atmospheric gases in solution in the surface layer of the ocean, as was demonstrated in a laboratory study by Stamska, et al (1990).

The reader might ask why Eq.1 has never, for climate modeling applications, been modified to take into account the varying aspect ratio, or e-folding depth, of bubble plumes, or to reflect the influence on bubble survival, and hence on aerosol production, of changes in the saturation level of the major gases in the surface waters, as suggested in this section and described respectively in Monahan (1986) and Stamska et al (1990). The answer to this question is probably two-fold; first, most modellers quite understandably are hesitant to add these complexities until the dependence of D on wind speed and other environmental variables, and of bubble survival on gas concentration levels, are more rigorously quantified, and secondly, the impact of these effects on $\partial F_o/\partial r$ are assumed to be modest when compared to the effect of even slight changes in W_B .

The quotient W_B/τ was originally intended to represent the rate at which optically resolvable whitecap area disappears, and, if a near dynamic equilibrium pertains, also the rate at which new whitecap area appears, on the sea surface. However, recent field and laboratory studies that things are more complex than these approximations would suggest.

4. The W-term, representing the fraction of the sea surface covered at any moment by optically resolvable whitecaps

Often in the development of these equations, and elsewhere, W and W_B , were used interchangeably. For example, in Monahan (1971), Monahan and O’Muircheartaigh (1980,1986), W actually represents the sum of both Stage A whitecap coverage (W_A) and Stage B whitecap coverage. The substitution of W for W_B , and visa versa, was deemed an appropriate approximation, since for the same wind conditions, etc., W_A was found to be typically only slightly more than 10% of W_B , or W (see, e.g., Monahan and Lu, 1990). (The conceptual image animating this model is one where the spilling wave crest, i.e. Stage A whitecap, with its bubble-rich sub-surface α -plume, settles down and transforms into a larger Stage B whitecap, with its dependent β -plume, in a brief enough time that only a sub-set of the largest bubbles have had time to break on the ocean surface, producing a limited number of film-droplets, in this pre-transformation interval. In this construct most of the bubble-mediated aerosol production (essentially all of the jet-droplet-production, and the overwhelming bulk of the film-drop production) occurs during the decay of the optically-resolvable Stage B whitecap, and for tens of seconds, perhaps minutes, afterward.)

The majority of whitecap studies record variations in the total amount of whitecap foam as a function of environmental forcing conditions, most notably wind speed at a height of 10 m above the mean water surface. This is because the separation of optically resolvable whitecap foam signal into the portion generated during active breaking, and the decaying portion on a wave-by-wave basis is not straightforward, thereby making such an image processing task time-consuming and not readily amenable to routine measurements. Therefore the use of W over W_B in Eq. 1 represents a practical choice. However making such a change then requires a re-evaluation, both conceptually and practically, of the characteristic whitecap timescale that is used Eq. 1. A discussion of this timescale, including data presented in a recent study (Callaghan, 2013) is discussed in more detail in the following section.

From the earliest discussion of the discrete whitecap method, there were attempts to use parameterizations of W in terms of U , the 10-meter elevation wind speed. In Monahan, Spiel, and Davidson (1986) a $W(U)$ expression from Monahan and O’Muircheartaigh (1980), obtained from the analysis of primarily trade wind observations, was introduced. Many of the discrepancies between recent satellite-derived W , and those obtained using a $W(U)$ expression of Monahan and O’Muircheartaigh were a result of applying a $W(U)$ inappropriate for certain high latitudes. These matters are discussed in Monahan, Hooker, and Zappa (2015), which appears on the same AMS Confex website as the present paper.

5. Characteristic Whitecap Timescales

Developments in digital image acquisition and image processing techniques have allowed much greater numbers of sea surface images to be

collected and analysed in whitecap studies, when compared to the original pioneering techniques developed by Monahan and co-workers. Consequently, the task of assembling and processing large data sets of digital images of the sea surface has become less laborious, thus yielding further data on the spatial and temporal evolution of whitecap foam on scales larger than previously possible. This has allowed further in-depth analysis of properties of individual whitecaps such as their timescales, with important implications for the discrete whitecap method.

Time-series of the spatial evolution of whitecap area reveal an approximate linear growth phase in whitecap area, followed by a close-to-exponential decay phase. From a remote sensing point of view, the peak in the area time-series may be used to effectively separate the evolving whitecap into a growth phase and a decay phase, each with its own characteristic timescale. Following Callaghan (2013), the whitecap formation timescale (τ_{form}) can be estimated as

$$\tau_{form} = A_o^{-1} \int_{-\infty}^0 A(t) dt \quad \text{Eq. 5}$$

where the limit $t = 0$ represents the time of maximum whitecap area. Similarly, the whitecap decay timescale (τ_{decay}) can be estimated as

$$\tau_{decay} = A_o^{-1} \int_0^{\infty} A(t) dt \quad \text{Eq. 6}$$

Together, the sum of these two whitecap timescales make up the characteristic whitecap lifetime (τ_{wcap}) such that

$$\tau_{wcap} = \tau_{form} + \tau_{decay} \quad \text{Eq. 7}$$

Recent field measurements have shown that in some circumstances the whitecap growth phase persists for considerably longer than the $\sim 10\%$ of the e-folding time for the decaying whitecap (see, e.g., Callaghan et al, 2012; Callaghan, 2013). Indeed, values of τ_{form} have been shown to be from 20% up to, in some instances, 100% of whitecap decay timescales for individual breaking waves, and therefore make a significant contribution of the overall whitecap timescale. Furthermore, when using W in place of W_B in Eq. 1, it has been shown that the effective whitecap timescale for the discrete whitecap method can be more accurately described as the area-weighted mean whitecap lifetime (τ_{DWM}) for an ensemble of M breaking waves in any given time period (Callaghan, 2013), which is written as

$$\tau_{DWM} = \sum_{i=1}^M A_{o,i} \tau_{wcap,i} \left(\sum_{i=1}^M A_{o,i} \right)^{-1} \quad \text{Eq. 8}$$

Field observations suggest τ_{DWM} cannot be expected to remain constant between different observational periods at a given location (Callaghan, 2013). Environmental factors expected to influence the value of τ_{DWM} include the severity of wave breaking and the injection depth of the bubble plume, the concentration and solubility of surfactants in the water column and the surface microlayer, and the scale of the breaking wave. Furthermore, since whitecap coverage is in part a function of the lifetime of whitecap foam along with the breaking rate and mean whitecap area, not choosing the correct value of τ_{DWM} will introduce systematic biases in the estimated sea spray aerosol production flux.

Developing approaches to parameterize whitecap timescales as a function of appropriate forcing variables, (e.g., wind speed, wave age, surfactant concentration, bubble injection depth) has the potential to lead to improvements in the accuracy of the DWM (Callaghan, 2013). Additional field data on the distributions of maximum whitecap area for individual breaking waves, and a scale-resolved breaking rate, will provide valuable information that can also be used to constrain observational measurements of total whitecap coverage. Such field data would lead to new insights on the natural variability of whitecap coverage, ultimately leading to improved parameterizations with commensurate improvements in the prediction of sea spray aerosol production flux. Finally, as stated in Monahan et al., 1982, it is likely that the quantity dE/dr in the DWM varies from spilling to plunging breaking waves, and further laboratory work is thus required to characterize this potential effect in a well-controlled laboratory setting.

Conclusions

This short paper has been motivated by observations of the authors of an inadvertent conceptual misunderstanding of the role the whitecap timescale plays in the discrete whitecap method, and also as an opportunity to highlight recent field data that has resulted in a refinement of this whitecap timescale. To re-iterate, this timescale is not related to the characteristic timescale of aerosol production per unit whitecap area, but rather is a fundamental property of the lifetime of optically resolvable whitecap foam. This distinction is important, especially in light of comparisons made between the continuous and discrete variants of the whitecap method. The re-evaluation of the discrete whitecap method is warranted as it remains widely used as evidenced in a recent review of SSA production flux parameterizations. Indeed, 9 of the 22 source functions

discussed in Grythe et al. (2014) are based upon the discrete whitecap method as introduced in Monahan et al. (1982;1986).

A correct interpretation of all the terms in the DWM is necessary if further improvements are to be incorporated into existing implementations of the DWM. Several of these have been highlighted here, such as incorporating information of bubble plume depth and water chemistry. Indeed, having available an improved parameterization for the rate of sea salt aerosol generation on the surface of the world's oceans will also lead to more accurate estimates of the sea surface heat, moisture, and gas fluxes, and thus to improved global climate modeling. However, it is important to recognize that accurate regional and global modelling of all the physical and chemical complexities that influence aerosol production flux will likely require a much more complex modelling approach than the DWM can currently offer. Also, we recognize that many research groups around the world are constantly advancing our knowledge of the coupled physical, chemical, and biological factors that influence the size, number and composition of oceanic bubbles, and of the resulting marine aerosol particles.

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