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Wind speed dependent size-resolved parameterization for the organic enrichment of sea spray

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Received: 25 February 2011 – Accepted: 21 March 2011 – Published: 4 April 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

For oceans to become a significant source of primary organic aerosol (POA), sea spray must be highly enriched with organics relative to the bulk seawater. We propose that organic enrichment at the air-sea interface, chemical composition of seawater, and the aerosol size are three main parameters controlling the organic mass fraction of sea spray aerosol (OM_{SS}). To test this hypothesis, we developed a new marine POA emission function based on a conceptual relationship between the organic enrichment at the air-sea interface and surface wind speed. The resulting parameterization is explored using aerosol chemical composition and surface wind speed from Atlantic and Pacific coastal stations, and satellite-derived ocean concentrations of chlorophyll-*a*, dissolved organic carbon, and particulate organic carbon. Of all the parameters examined, a multi-variable logistic regression revealed that the combination of 10 m wind speed and surface chlorophyll-*a* concentration ($[Chl-a]$) are the most consistent predictors of OM_{SS} . This relationship, combined the published aerosol size dependence of OM_{SS} , resulted in a new parameterization for the organic carbon fraction of sea spray. Global marine primary organic emission is investigated here by applying this newly-developed relationship to existing sea spray emission functions, satellite-derived $[Chl-a]$, and modeled 10 meter winds. Analysis of model simulations show that global annual submicron marine organic emission associated with sea spray is estimated to be from 2.8 to 5.6 Tg C yr⁻¹. This study provides additional evidence that marine primary organic aerosols are a globally significant source of organics in the atmosphere.

1 Introduction

Indirect radiative forcing of anthropogenic aerosols is the major source of uncertainty in climate projections today (IPCC, 2007). A large fraction of this uncertainty may be related to the number concentration and size distribution of marine aerosol that are prescribed or diagnosed in global climate models (GCMs) (Hoose et al., 2009), and

ACPD

11, 10525–10555, 2011

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the uncertainties associated with forcings and feedbacks involving marine clouds and precipitation (Bony and Dufresne, 2005; Clement et al., 2009). Marine aerosols are particularly important as they contribute considerably to the global aerosol load and are emitted from a large area of the Earth's surface underlying an atmosphere with low aerosol concentration. The lower bounds (typically from 10 to 40 cm^{-3}) of background aerosol concentration often prescribed in GCMs can vary the simulated aerosol indirect effect by over 80%, from -1.8 W m^{-2} to -0.2 W m^{-2} (Hoose et al., 2009).

Although sea-salt typically dominates total marine aerosol mass burden and emission rates, organic aerosols of marine origin can contribute a considerable fraction of the submicron aerosol mass concentration near biologically productive waters (O'Dowd et al., 2004). Organic aerosol concentrations of up to $3.8 \mu\text{g m}^{-3}$ have been observed on the Atlantic coast of Ireland during periods of onshore flow (Ovadnevaite et al., 2011) where on average $\sim 80\%$ of the carbonaceous material has been directly linked to plankton emissions (Ceburnis et al., 2011). Cloud condensation nuclei (CCN) over the remote oceans typically range from a few tens per cm^3 over biologically inactive regions (seasons) to a few hundred per cm^3 under biologically active conditions (Andreae, 2007). Since cloud properties are most sensitive to the addition of particles when the background concentration is low (Platnick and Twomey, 1994), ambient measurements, remote sensing, and modeling studies indicate that ocean biology could influence marine cloud microphysical properties (Shaw et al., 1983; Charlson et al., 1987; Middlebrook et al., 1998; O'Dowd et al., 2004; Meskhidze and Nenes, 2006, 2010; Bigg and Leck, 2008; Sorooshian et al., 2009, 2010; Thomas et al., 2010).

Organic aerosols in the marine boundary layer are proposed to have different sources that can be broadly classified as primary, produced from the mechanical process of bubble bursting, and secondary (SOA), derived from precursor biogenic volatile organic compounds (BVOCs) emitted by phytoplankton and macroalgae or by photolysis of chromophoric dissolved organic matter (CDOM) in the water column (Zhou and Mopper, 1997; O'Dowd and de Leeuw, 2007; Sellegri et al., 2008). In addition, SOA can also be derived from the chemical transformation of primary or secondary components

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present in the condensed phase. Such complex transformations could take place at the particle surface or in the aqueous phase, and may also involve a further step through the gas phase in which semivolatile aerosol components can be oxidized to form new condensable products (Rinaldi et al., 2010). Due to the potentially important contribution of marine organic aerosol to the CCN budget over the remote ocean, improvement of the fundamental process-level understanding of marine primary and secondary aerosol production mechanisms is needed to develop more reliable parameterizations that can be confidently applied in GCMs. These new parameterizations must capture the total mass of marine organic aerosol emission as well as their cloud nucleating properties (i.e., number concentration and size dependent chemical composition of submicron sea spray). Here, we examine the factors that affect the emission of marine primary organic aerosols (POA).

Laboratory and ambient measurements have revealed that the organic fraction of submicron sea spray aerosols can be enriched (relative to the underlying seawater) by as much as 2 to 3 orders of magnitude (Blanchard, 1964; Gershey, 1983a; Hoffman and Duce, 1974, 1976; Oppo et al., 1999; Keene et al., 2007; Facchini et al., 2008; Russell et al., 2010). The organic enrichment of sea spray is thought to be controlled by the accumulation of insoluble organic matter at the air-sea interface (Hoffman and Duce, 1976; Gershey et al., 1983b). This natural enrichment of the ocean surface layer by organics has been shown to be a function of both meteorological factors, such as surface wind speed, and oceanic parameters including the concentration of dissolved organic carbon ([DOC]), particulate organic carbon ([POC]), chlorophyll-*a* ([Chl-*a*]), and type of organic carbon (OC) in the air-sea interface and underlying water (Hoffman and Duce, 1976; Gershey, 1983a, b; O'Dowd et al., 2004, 2008; Russell et al., 2010).

The air-sea interface can roughly be thought of as being in three regimes where (1) very high organic enrichment leads to visible biogenic surface slicks thicker than 50 μm , (2) moderate organic enrichment leads to a non-slick microlayer $\sim 50 \mu\text{m}$ thick, and (3) no enrichment is present. Figure 1 shows the conceptual relationship between the organic enrichment of the air-sea interface (given as an OC/Na ratio based on

values summarized by Russell et al., 2010) and surface wind speed based on a review of previous works. This figure shows that for given chemical composition of seawater, the highest enrichments are expected during calm winds. An increase in wind speed above 3–4 m s⁻¹ will cause a rapid decrease in the enrichment (the Langmuir circulations starts breaking up the slicks), and its effective removal for wind speeds in exceed 8 m s⁻¹ (when the wave breaking thoroughly mixes the microlayer with the underlying water). Although there is significant spread in observational data, most of the studies agree that slicks in the open ocean are only observed for surface wind speed <5 m s⁻¹ (Romano, 1996). When winds increase from 2 to 5 m s⁻¹, there is an increase in the formation of gaps and a decrease in the satellite-derived areal extent of ocean slicks (Marmorino et al., 2008). This increase of gaps and subsequent decrease in slick coverage is consistent with the Dysthe (2006) model describing the tearing of a surface film in a region of positive surface straining from Langmuir circulations (Langmuir, 1938; Leibovich, 1983). Many studies also report a decrease in microlayer (non-slick organic layer of moderate enrichment) concentration relative to the underlying seawater for surface wind speeds in excess of ~4 m s⁻¹ (Obernosterer et al., 2008), except for Wurl et al. (2010) who report constant microlayer surfactant enrichment for winds up to 5.5 m s⁻¹, with enrichment persisting for winds up to 10 m s⁻¹. When surface winds exceed 8 m s⁻¹, the initiation of wave breaking is expected to cause the destruction of the microlayer by mixing it into the underlying seawater (Carlson, 1983). Extrapolation of the linear decrease in microlayer thickness with wind speed observed by Liu and Dickhut (1998) predicts a microlayer thickness of 0 μm at a wind speed of ~8.5 m s⁻¹, consistent with this picture. At wind speeds in excess of ~11 m s⁻¹, the mechanism for sea spray generation is via mechanical disruption of wave crests (e.g., Andreas, 1998 and references therein). The spume drops torn from the wave crests consequently have composition of bulk seawater with little enrichment. Overall, review of published studies indicates that surface wind speed can play a crucial role in determining the physical and chemical characteristics of the air-sea interface and thus the organic enrichment of sea spray.

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In addition to the potential impact from wind speed, the organic enrichment of sea spray has been shown to be affected by the chemical composition of seawater. Past studies have consistently found that the marine aerosol OC concentration is higher over regions of high biological activity (O'Dowd et al., 2004; Sciare et al., 2009; Miyazaki et al., 2010). The results of multiple ambient and laboratory studies indicated that the upwind concentrations of [Chl-*a*], [POC], and [DOC] can be used as a proxy for the organic enrichment of sea spray (Gershay, 1983a, b; Hoffman and Duce, 1976; O'Dowd et al., 2004, 2008; Russell et al., 2010).

Several attempts to quantify and characterize size-dependent chemical composition of sea spray aerosols have recently been carried out. These measurements consistently have shown that the organic mass fraction of submicron sea spray increases with decreasing particle size (Oppo et al., 1999; Keene et al., 2007; Facchini et al., 2008), with a small and relatively constant organic fraction for supermicron particles with aerodynamic diameter $<10\ \mu\text{m}$ (Oppo et al., 1999; Keene et al., 2007; Facchini et al., 2008). Keene et al. (2007) reported that the water soluble organic carbon of artificially generated aerosols within a chamber was highly enriched in all aerosol size fractions, with greatest enrichments in size fraction with geometric mean diameter of $0.13\ \mu\text{m}$. In this smallest size fraction sampled, organic carbon comprised up to 80% of aerosol mass and dominated the mass of most individual particles (Keene et al., 2007). Facchini et al. (2008) similarly reported a high contribution of organic matter in submicron sea spray particles (up to $77 \pm 5\%$ in the $0.125\text{--}0.25\ \mu\text{m}$ size range). However, their analysis discriminated between water soluble and insoluble organic carbon (WSOC and WIOC, respectively), finding a dominant contribution (up to $94 \pm 4\%$) from the water insoluble fraction.

Despite the likely impact of both meteorological factors and chemical composition of seawater on sea spray chemistry, most of the recent parameterizations of marine POA emissions use only [Chl-*a*] as a proxy for deriving the organic mass fraction of sea spray aerosols (O'Dowd et al., 2008) or for determining the total magnitude of the emissions (Spracklen et al., 2008). In this paper, we develop a new parameterization

for organic fraction of sea spray aerosol based on the conceptual picture of Fig. 1, and constrain it using ambient data from two different oceanic regions.

2 Data and methods

2.1 Aerosol chemistry

Two coastal stations with long-term measurements of aerosol chemistry were selected for this study; the Mace Head Atmospheric Research Station (53.33° N, 9.90° W) on the Atlantic coast of Ireland and a site at the Point Reyes National Seashore (38.12° N, 122.91° W) on the Pacific coast of California. Measurements of the ~50–100 h average chemical composition of aerosols with an aerodynamic diameter <1.5 μm from Mace Head (Cavalli et al., 2004; Yoon et al., 2007) have been supplemented with measurements of the chemical composition of aerosols with diameters <2.5 μm at Point Reyes near San Francisco, CA taken as part of the United States' Interagency Monitoring of Protected Visual Environments (IMPROVE) network (<http://vista.cira.colostate.edu/improve/>). At Point Reyes, aerosols are captured on filters for 24 hours (midnight to midnight local time) every 3 days and analyzed for chemical composition. Point Reyes was selected from the large (170 stations) IMPROVE monitoring network because it is located very close to the coast (~4 km) and has the highest frequency of aerosols exhibiting marine characteristics (White, 2008).

Several compounds are included in the analysis of aerosol chemical composition, including elemental carbon (EC), OC, and sodium (Na). To avoid potential problems with Na measurements (White, 2008), the data at Point Reyes were analyzed for the years 2000–2001 and 2005–2007. For Mace Head, the aerosol concentrations measurements conducted only during periods of “clean sector” conditions (wind direction from 180° to 300°, particle concentration <700 cm⁻³, EC < 0.05 μg m⁻³) for the years 2002 to 2006 are included in the analysis. The OC measurements from Mace Head are further separated into WSOC and WIOC following Cavalli et al. (2004). Unlike

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Mace Head, OC measurements obtained from the IMPROVE network do not segregate WSOC and WIOC. To be consistent with the Mace Head data, it was assumed that 70% of the OC measured at Point Reyes is insoluble. This WIOC/OC fraction was similar to that observed at Mace Head (Cavalli et al., 2004; O'Dowd et al., 2004) and Amsterdam Island (37.80° S, 77.57° E) (Sciare et al. 2009) during the summer when OC concentrations were highest. The organic mass fraction of sea spray (OM_{ss}) can then be determined by converting the OC measurements to organic matter (OM) using a 1.4 OM/OC ratio (Descari et al., 2007; Facchini et al., 2008) and using the equation $OM_{ss} = WIOM/(WIOM+sea-salt)$ first employed in O'Dowd et al. (2008), where sea-salt is estimated as $Na/0.3061$ corresponding to the mass fraction of sodium in dry sea-salt (Seinfeld and Pandis, 2006). OM_{ss} is used here as the primary variable of the analysis because it is independent of the sea spray concentration (considering that POA of marine origin and sea-salt have similar production mechanisms and sinks) and can be easily applied to published sea spray emission parameterizations.

2.2 Meteorology

Wind speed and direction data used in the analysis is different for the two stations. At Mace Head, 10 m wind speed and direction was measured at the same location as the aerosol measurements, with the speed recorded only during periods when aerosols were being measured. At Point Reyes, hourly wind speed and direction at 5 m was measured at the Bodega Bay buoy #46013 (38.24° N 123.30° W) as part of the United States' National Data Buoy Center network (<http://www.ndbc.noaa.gov/>). This buoy is located ~37 km west of Point Reyes in the Pacific Ocean. The 5 m wind speed was converted to 10 m using the power-law wind-profile as follows: $U_{10} = U_5(\frac{10}{5})^{0.11}$ (Hsu et al., 1994). In order to make the aerosol measurements between the two sites more comparable, a wind direction filter was used on the Point Reyes data including only days with all 24 h having onshore wind directions (between 180° and 315°). Such rigorous filtering reduced the number of qualifying days from 365 to 36. Further filtering of the Point Reyes aerosol measurements included only days with $EC < 0.05 \mu g m^{-3}$ in

order to avoid potential influence of OM from anthropogenic sources (Clarke, 1989) and to make them consistent with the Mace Head clean sector data.

2.3 Ocean chemistry

The oceanic parameters derived from satellite data include 8-day average Level 3 surface [Chl-*a*], [POC], and [DOC] from the Sea Wide Field-of-view Sensor (SeaWiFS) for the years 2000–2007. [Chl-*a*] is determined using the OC4v4 algorithm (O'Reilly et al., 1998, 2000), and [POC] was calculated from normalized water-leaving radiances at 443, 490, and 555 nm using the method of Stramska et al. (2009). In order to estimate the ocean [DOC] distribution, a sea surface temperature (SST)-based approach (Siegel et al., 2002) and a colored dissolved organic matter (CDOM)-based approach (Mannino et al., 2008) were combined to generate the estimates of surface layer [DOC] for the years 2000–2007. The 8-day average SST data was obtained from Moderate Resolution Imaging Spectroradiometer (MODIS), and the 8-day average normalized water leaving radiance data at 490 and 555 nm (required for the CDOM-based approach) were retrieved from SeaWiFS. As each method is designed to estimate the [DOC] in different parts of the ocean, the SST-based approach was used for the open ocean and the CDOM-based approach was used for coastal waters (Siegel et al., 2002; Mannino et al., 2008). All of the satellite-derived data was regridded to $1^\circ \times 1^\circ$ spatial resolution. The reported global root mean squared error of the [Chl-*a*], [POC], and [DOC] were $\sim 0.2 \text{ mg m}^{-3}$, 20 mg m^{-3} , and $2.6\text{--}8.6 \mu\text{mol L}^{-1}$, respectively (Siegel et al., 2005; Stramska et al., 2009; Mannino et al., 2008; Siegel et al., 2002). For each aerosol measurement, a non-weighted upwind average of the temporally-collocated [Chl-*a*], [DOC] and [POC] was calculated using 24-h back-trajectories based on the observed wind speed and direction (grouped into 45° vectors).

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3 Results and discussion

3.1 Factors affecting organics in sea spray

3.1.1 Wind speed

A plot of organic carbon fraction against surface wind speed in Fig. 2a shows that for both the Mace Head and Point Reyes measurement stations, the OM_{ss} decreases with increasing 10 m wind speed in a manner consistent with our conceptual picture given on Fig. 1. For wind speeds above 10 m s^{-1} , sea spray aerosols are largely composed of sea-salt with very little contribution from organics. As wind speeds decrease to 5 m s^{-1} , a strong increase in the organic fraction of sea spray was observed. This trend can be explained by the presence of a surface microlayer described by our conceptual wind speed-organic enrichment relationship. No data exists for surface winds of less than 4 m s^{-1} due to longer averaging time at the two stations (Mace Head data was typically averaged over 50–100 h and Point Reyes over 24 h). However, considering that wind speed of $\geq 4 \text{ m s}^{-1}$ is typically associated with the onset of bubble formation (Monahan and O'Muircheartaigh, 1986), enrichment at wind speed values lower than that may not be environmentally relevant. Nevertheless, our conceptual picture on Fig. 1 predicts that OM_{ss} should plateau at a wind speed value of $\sim 3 \text{ m s}^{-1}$. Using the MATLAB curve fitting tool and the proposed conceptual relationship, we have developed a logistic function that is a good fit ($R^2 = 0.38\text{--}0.47$) for the existing ambient measurements and requires no artificial cutoff. The general relationship between sea spray organic enrichment and wind speed shown on Fig. 2a suggests that winds (through their effect on air-sea interface) may play a considerable role in the enrichment of sea spray aerosol.

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3.1.2 Ocean chemistry

Analysis of ambient marine aerosol chemical composition suggests that there is a maximum enrichment of sea spray aerosols by organics that, although usually occurring at low wind speed, can be vastly different for different measurement locations. Figures 2b–d show that each ocean chemistry parameter examined ([POC], [DOC], and [Chl-*a*]) had a positive relationship with OM_{ss} . It is worth noting that the relationship between OM_{ss} and [POC] (Fig. 2c) was stronger than [DOC] (Fig. 2d) for Mace Head, consistent with the hypothesis of Russell et al. (2010) that [POC] influences organic enrichment of Northern Atlantic sea spray aerosols. Overall, out of the three ocean parameters examined, the strongest and most consistent relationship was found between OM_{ss} and [Chl-*a*] (Fig. 2b). Therefore, in our new parameterization of the organic carbon fraction of sea spray, [Chl-*a*] was chosen as a proxy for the ocean chemistry. Using the MATLAB surface fitting tool and a general multi-variable logistic equation, the combined relationships of OM_{ss} vs. U_{10} and OM_{ss} vs. [Chl-*a*] were found to be very similar for both stations. The corresponding coefficients in each equation were averaged to yield:

$$OM_{ss}(\text{Chl-}a, U_{10}) = \frac{OM_{ss}^{\max}}{1 + \exp(-2.63[\text{Chl-}a] + 0.18 U_{10})} \quad (1)$$

where [Chl-*a*] is in units of mg m^{-3} and U_{10} is in units of m s^{-1} . The magnitude-determining numerator of the equation, OM_{ss}^{\max} , is the maximum OM_{ss} observed at the two sites during the entire measurement period (0.24 for Point Reyes and 0.78 for Mace Head). When compared to measurement data, the R^2 value for Eq. (1) is ~ 0.52 and 0.56 at Point Reyes and Mace Head, respectively (see Fig S1 for scatterplot). A 3-D visualization of Eq. (2) in Fig. 3 shows the interdependence of [Chl-*a*] and U_{10} in determining OM_{ss} , where the steepest slope corresponding to highest sensitivity occurs in the intermediate [Chl-*a*] and U_{10} values and the shallowest slopes (constant OM_{ss}) occur at the extreme values where [Chl-*a*] $> 1 \text{ mg m}^{-3}$ and $U_{10} > 1.5 \text{ m s}^{-1}$. Despite the

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relationship between [Chl-*a*] and wind speed that exists because of seasonal effects (high [Chl-*a*] and low winds in summer, low [Chl-*a*] and high winds in winter), the inverse relationship between OM_{ss} and U_{10} remains even when the measurements are binned into “low”, “moderate” and “high” [Chl-*a*] regimes (see Fig. S2). Comparison of Eq. (1) with the high organic aerosol concentration episode at Mace Head described in Ovadnevaite et al. (2011) reveals remarkably good agreement, with an observed and predicted OM_{ss} of 0.57 and 0.54, respectively (assuming $U_{10} = 10 \text{ m s}^{-1}$ and $[Chl-a] = 1 \text{ mg m}^{-3}$ based on Table 1 and Fig. 1 from Ovadnevaite et al., 2011).

The differences between the relationship in Eq. (1) and those of previous parameterizations are greatest at high and low wind speeds. For a given [Chl-*a*], our equivalent submicron OM_{ss} will typically be lower than that of O’Dowd et al. (2008) and Viganti et al. (2010) for strong winds and higher for low wind conditions.

3.1.3 Aerosol size

The different aerosol sizes ($<1.5 \mu\text{m}$ from Mace Head and $<2.5 \mu\text{m}$ at Point Reyes) measured at each site were likely to contribute to the differences in OM_{ss}^{max} . Since neither the Mace Head nor Point Reyes datasets we used had size-resolved aerosol composition, to better constrain OM_{ss} as a function of aerosol size we used published measurements of the size-resolved organic mass fraction of aerosols generated from bubble-bursting of seawater over the biologically-active Northern Atlantic (Facchini et al., 2008). Figure 4 shows the OM_{ss} measurements from Facchini et al. (2008) fitted by the following equation:

$$OM_{ss}(D_p) = \frac{OM_{ss}^{\text{max}}(D_p)}{1 + 0.03 \exp(6.81D_p)} + OM_{ss}^{\text{min}}(D_p) \quad (2)$$

where D_p is the ambient ($RH = 80 \pm 8\%$) aerosol aerodynamic diameter in micrometers, and $OM_{ss}^{\text{max}}(D_p)$ and $OM_{ss}^{\text{min}}(D_p)$ are size-dependent maximum and minimum organic carbon fraction of sea spray with values of 0.82 and 0.03, respectively.

Our size-dependent organic enrichment is consistent with other measurements/parameterizations. Size-resolved measurements from Keene et al. (2007) give similar results for the organic fraction of sea spray, but the reported OC as only water soluble is potentially incompatible with the data from Mace Head. Both Oppo et al. (1999) and Long et al. (2011) describe similar size-dependent organic enrichment of sea spray through adsorption models.

3.1.4 Combination of wind speed, [Chl-*a*], and aerosol size

To create an OM_{ss} equation as a function of [Chl-*a*], U_{10} , and sea spray size, we have combined Eqs. (1) and (2) in a way that retains the size dependence of OM_{ss} from Eq. (2) but scales with [Chl-*a*] and U_{10} as described in Eq. (1). Specifically, the $OM_{ss}^{max}(D_p)$ and $OM_{ss}^{min}(D_p)$ values from Eq. (2) were scaled with the OM_{ss} from Eq. (1) after setting $OM_{ss}^{max}(D_p)$ to 1 reflecting the highest potential enrichment in the organic fraction (Bigg and Leck, 2008). This allows the size dependence from biologically-active Northern Atlantic (Facchini et al., 2008) region to be globally applicable to areas with vastly different winds and [Chl-*a*]. The resulting final OM_{ss} parameterization is:

$$OM_{ss}(Chl-a, U_{10}, D_p) = \frac{1}{1 + \exp(-2.63[Chl-a] + 0.18 U_{10})} + \frac{0.03}{1 + \exp(-2.63[Chl-a] + 0.18 U_{10})} \quad (3)$$

There are two main advantages of Eq. (3): (i) it does not require any artificial cutoffs of wind speed or [Chl-*a*] and (ii) it can give the size-resolved organic carbon fraction of sea spray aerosols from globally-available U_{10} and [Chl-*a*] data. Figure 5 examines the dependence of OM_{ss} on three parameters: sea spray size (D_p), chemical composition of ocean seawater ([Chl-*a*]) and physical mechanism of aerosol emission (U_{10}) described by Eq. (3). To make the dependence of OM_{ss} on all three parameters easily visible, we have selected three different aerosol diameters roughly corresponding to typical marine aerosol Aitken, accumulation, and coarse mode diameters (Yoon et al., 2007). According to Fig. 5, OM_{ss} increases for higher ocean biological productivity and

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lower wind speed and shows very strong dependence on aerosol size. These general trends are non-linear; changes in OM_{ss} occur rapidly at moderate wind speed and [Chl-*a*] and more slowly at the extremes due to the plateaus found in the logistic curve. Our calculations suggest that there is little difference between the maximum organic fraction of the Aitken and accumulation mode aerosols (~ 0.8 – 0.9), while the coarse mode maximum organic fraction is substantially lower (< 0.02) due to the increasing dominance of sea-salt in these particles.

3.2 Estimated emission rate

To estimate marine POA fluxes in different parts of the oceans, we have used Mårtensson et al. (2003) and Gong (2003) sea spray functions with hourly averaged U_{10} calculated by the Community Atmosphere Model (CAM5) (Collins et al., 2006) and monthly-averaged climatological [Chl-*a*] retrieved by SeaWiFS. The Mårtensson et al. (2003) function was used for dry particle diameters ranging from ~ 0.02 to $2.8 \mu\text{m}$ and the Gong (2003) function for diameters from ~ 2.8 to $20 \mu\text{m}$. Both the Mårtensson et al. (2003) and Gong et al. (2003) functions are assumed to represent sea spray aerosols whose effective densities are determined by the fractional contribution of sea-salt and organics. Since the magnitude of OM_{ss} can potentially influence the hygroscopicity of the aerosol, a conversion between dry and ambient aerosol diameters was achieved by changing the OM_{ss} from Eq. (3) to an organic volume fraction ($\rho_{\text{organic}} = 1 \text{ g cm}^{-3}$ and $\rho_{\text{NaCl}} = 2.165 \text{ g cm}^{-3}$) and using it to estimate the resulting hygroscopicity ($\kappa - \kappa$) values. The growth factor (GF) was derived at ambient $\text{RH} = 80\%$ for the combined organic-inorganic particle by assuming $\kappa_{\text{organic}} = 0$ and $\kappa_{\text{NaCl}} = 1.12$ (Petters and Kreidenweis, 2007). Figure 6 shows the global annual submicron (dry aerosol diameter $< 1 \mu\text{m}$) marine primary organic aerosol emission rate, with the highest emissions occurring over the mid latitude waters (30 – 60°N and 30 – 60°S) where strong winds and high [Chl-*a*] are common throughout the year. Despite having a low OM_{ss} , areas with strong winds and low [Chl-*a*] still have moderate marine POA emissions due to the exponential wind speed dependence of sea spray emissions.

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The predicted emission rates in tropical oceanic regions, however, are larger than those from previous studies (Vignati et al., 2010; Langmann et al., 2008; Gantt et al., 2009). As there have been relatively few aerosol measurements from tropical oceanic regions, the predicted organic emission rates are difficult to verify. The global magnitude of submicron marine POA emissions in this study is 2.8 Tg C yr^{-1} , with emissions up to 5.6 Tg C yr^{-1} when using a growth factor of aerosols composed of 100% organic (GF = 1). In this later formation, a GF of 1 effectively assumes that the aerosol diameter measurements of Facchini et al. (2008) thus the D_p in Eq. (2) represents the dry aerosol diameter. This assumption is not unreasonable due to some drying from the pressure drop going down the stages of the Berner impactor used in Facchini et al. (2008). The measurements of Facchini et al. (2008) did not extend as far into the coarse mode as the model, so our estimated total global marine POA emission rate of $15.9 \text{ Tg C yr}^{-1}$ (up to $18.7 \text{ Tg C yr}^{-1}$ when using a growth factor of (1) is highly uncertain. The considerable organic enrichment in the largest aerosol size fraction (up to 40% of aerosol mass) reported in size-dependent measurements of Keene et al. (2007) suggests that this value could be a low estimate.

4 Caveats and uncertainty

One of the main caveats in our study arises from the use of observed or estimated WIOC at a few coastal sites as a proxy for global marine POA. To derive chemical composition of the organic fraction of sea spray, we have used values reported by Facchini et al. (2008) which reported a high contribution (up to $77 \pm 5\%$) of organic matter in small diameter sea spray particles. Although this large organic contribution was also observed by Keene et al. (2007), such high fractions of organics have not been found in all locations (Modini et al., 2010). Additionally, the WIOC/OC ratio used for the Point Reyes data is also uncertain as not all sites have observed this ratio (Miyazaki et al., 2010). At Mace Head, long term measurements have revealed that this ratio is not consistently observed (Rinaldi et al., 2010).

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Coastal sites also suffer from uncertainties related to the influence of the surf zone and continental aerosols. Even though sea spray aerosol production by surf-zone wave breaking were shown to be 1–2 orders of magnitude higher than that of the open ocean (de Leeuw et al., 2000; Vignati et al., 2001), previous studies have suggested that aerosol chemical composition and flux parameterizations derived from coastal measurements can provide a suitable proxy for open ocean conditions and therefore can be successfully used for global emissions assessments (Clarke, 2006; Rinaldi et al., 2009). The use of wind speed measurements near the aerosol sampling location for both the Point Reyes and Mace Head sites also leads to some uncertainty due to the size of the concentration footprint from which the sea spray is emitted. Ceburnis et al. (2008) describes that while the flux footprint of the Mace Head station is typically within 5 km, the concentration footprint can be 10–100 times further upwind. We feel as though this uncertainty does not result in large errors in the magnitude and direction of our wind speed dependence because of the similarity between Mace Head nearshore and offshore wind speeds derived from NASA's Quick Scatterometer (QuikSCAT) (see Fig. S3) when averaged over the 50–100 h sampling period. Continental sources can also potentially contribute to OM_{ss} measurements at marine sites (Turekian et al., 2003), though it is expected such sources to be minor in this study due to the very long (thousands of kilometers) upwind fetch of open ocean at both sites and evidence from a recent study that 80% of the onshore flow organic aerosols at Mace Head had a marine source (Ceburnis et al., 2011). These inconsistencies and uncertainties highlight the difficulty in confidently applying our organic sea spray parameterization (derived using limited spatiotemporal measurements) to the global scale.

Moreover, when we compared our conceptual model to measurement data from Mace Head and Point Reyes, we assumed consistency between measured or estimated WIOM and POA. Such an assumption may lead to additional uncertainty in marine POA emissions, as it disregards the fraction marine SOA that may be water insoluble or POA that is water soluble (even though these contributions are expected

to be minimal). The well recognized SOA component of marine aerosol derived from ocean-emitted BVOCs is methanesulfonate (MS^-) from dimethyl sulfide (Saltzman et al., 1983), but other SOA precursor gases such as biogenic isoprene and monoterpenes emitted from phytoplankton have also been postulated to affect marine organic aerosols (Meskhidze and Nenes, 2006; Gantt et al., 2009; Luo and Yu, 2010). Additionally, oxidation of marine primary OM has been suggested to lead to the formation of WSOM which has typically been considered to have secondary sources (Ceburnis et al., 2008; Claeys et al., 2009; Rinaldi et al., 2010; Ovadnevaite et al., 2011). Such uncertainty regarding the origin of marine-source primary and secondary organic aerosols and inadequate understanding of marine organic aerosol formation processes adds to the difficulty in estimating marine POA emissions.

5 Conclusions

A conceptual relationship between wind speed and sea spray organic mass fraction (OM_{ss}) has been developed and used to calculate marine primary organic aerosol (POA) emissions in different parts of the global oceans. Our analysis predicts the highest enrichments of marine aerosol with organics during calm winds, when large amounts of organic material can accumulate at the sea-surface often forming visible slicks. An increase in wind speed (above $3-4 \text{ m s}^{-1}$) will initiate Langmuir circulations causing a breakup of the slicks and a rapid decrease in OM_{ss} . Wave breaking (caused by wind speeds in excess of 8 m s^{-1}) will thoroughly mix the microlayer with the underlying water, effectively reducing organic enrichment at the air-sea interface and thus in sea spray aerosols. Derived relationships between observed 10 m wind speed (U_{10}) and remotely-sensed upwind oceanic concentrations of chlorophyll-*a* ([Chl-*a*]), particulate organic carbon ([POC]), and dissolved organic carbon ([DOC]) were compared to aerosol chemical composition from the Mace Head (Atlantic) and Point Reyes (Pacific) coastal sites. Our results indicate that wind speed, [Chl-*a*], and aerosol size are likely to be the three most important parameters regulating OM_{ss} .

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The new empirical parameterization using a logistic fit for the relationship between OM_{ss} and $[Chl-a]$, U_{10} , and aerosol diameter was then used to calculate size-resolved global marine POA emissions using established sea spray functions. Although our model-predicted submicron marine organic carbon emission rate ranging from 2.8 to 5.6 Tg C yr⁻¹ is comparable to several recent studies (Spracklen et al., 2008; Vignati et al., 2010), the wind speed dependence of our parameterization gives a distinct spatial distribution. The exponential wind speed dependence of the sea spray function leads to the highest marine POA emissions in areas with strong winds; however, we also predict moderate emissions widespread throughout the global oceans due to the inverse relationship between wind speed and OM_{ss} . Our parameterization suggests that oceanic regions with high biological productivity are more likely to have higher marine POA emissions than areas with low productivity. Additional comparative modeling analysis of different marine POA emission schemes, combined with long-term measurements of marine aerosol concentration in previously under-sampled areas (i.e., tropical oceans), is needed to determine the accuracy of this marine organic aerosol emission parameterization.

Supplementary material related to this article is available online at:
**[http://www.atmos-chem-phys-discuss.net/11/10525/2011/
acpd-11-10525-2011-supplement.pdf](http://www.atmos-chem-phys-discuss.net/11/10525/2011/acpd-11-10525-2011-supplement.pdf)**

Acknowledgements. This research was supported by the Office of Science (BER), U.S. Department of Energy, Grant No. DE-FG02-08ER64508 and a NASA NESSF fellowship. The authors would like to thank Markus Petters and Jurgita Ovadnevaite for helpful discussion and suggestions. The experimental work performed at Mace Head was supported by the EU Project MAP (Marine Aerosol Production), the EPA Ireland project EASI-AQCIS (Exchange at the Air-Sea Interface: Air Quality & Climate ImpactS), and HEA PRTL14. ACCENT (Atmospheric Composition Change the European Network of Excellence) is also gratefully acknowledged.

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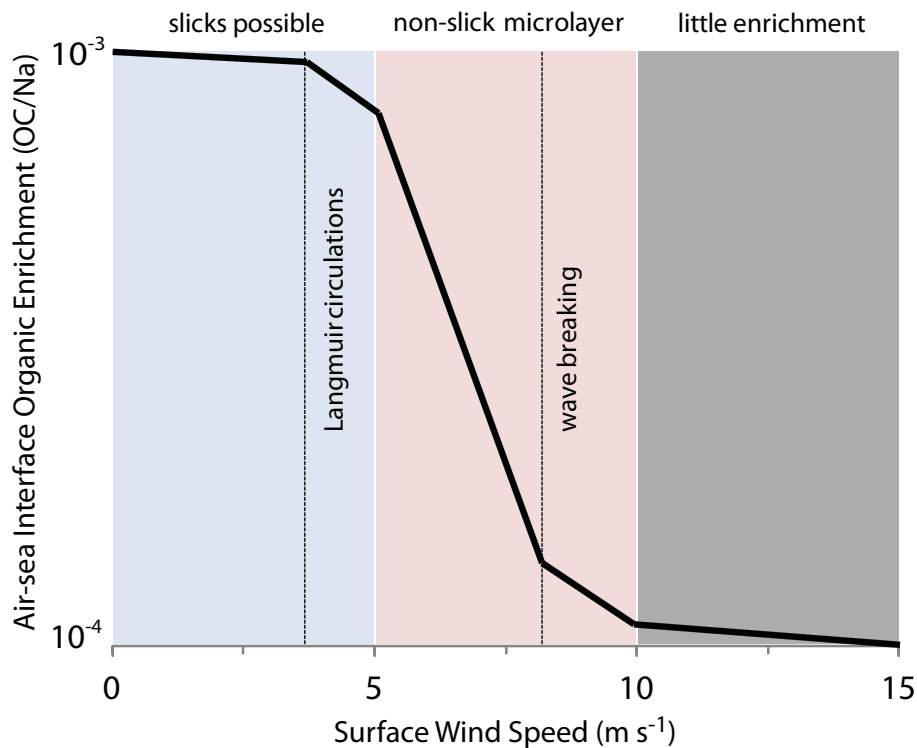


Fig. 1. Conceptual relationship between the organic enrichment of the air-sea interface and surface wind speed. Different colors depict potential regimes of the physical and chemical characteristics of the air-sea interface. These values represent global averages and may not be applicable for any given location.

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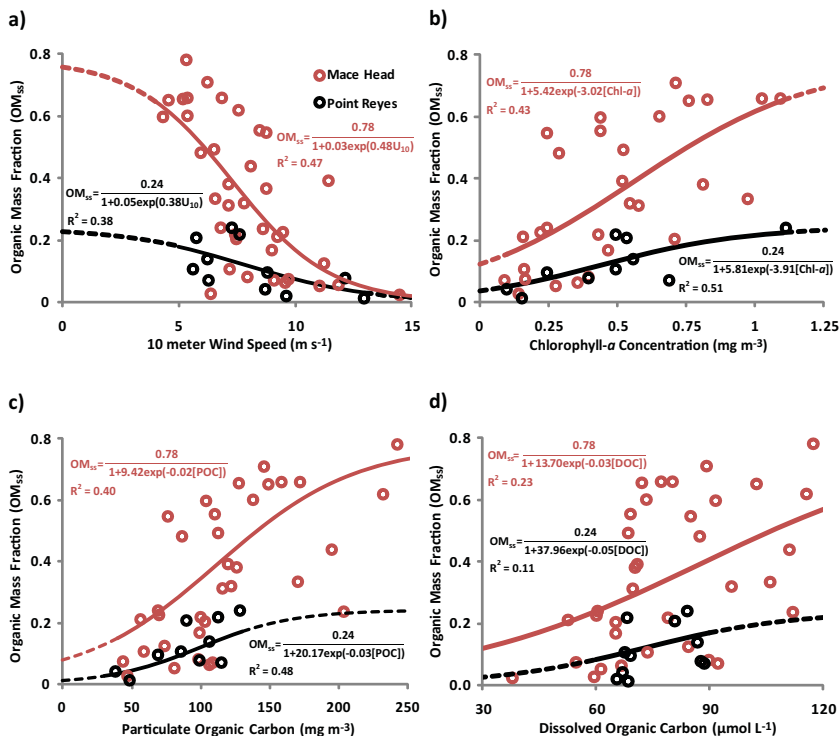


Fig. 2. Organic mass fraction of sea spray as a function of (a) observed 10 meter wind speed (U_{10}) and upwind averaged ocean concentrations of 8-day averaged (b) [Chl- a], (c) [POC], and (d) [DOC] for Mace Head and Point Reyes. In Fig. 2b, four outlier data points for Mace Head [Chl- a] $>1.25 mg m^{-3}$ have been excluded due to their occurrence during anomalously high chlorophyll- a concentrations in the region. The aerosol sizes measured at Mace Head and Point Reyes were $PM_{1.5}$ and $PM_{2.5}$, respectively, the solid lines show regression relationships for each station, and the dotted lines show an extrapolation of the regression relationship.

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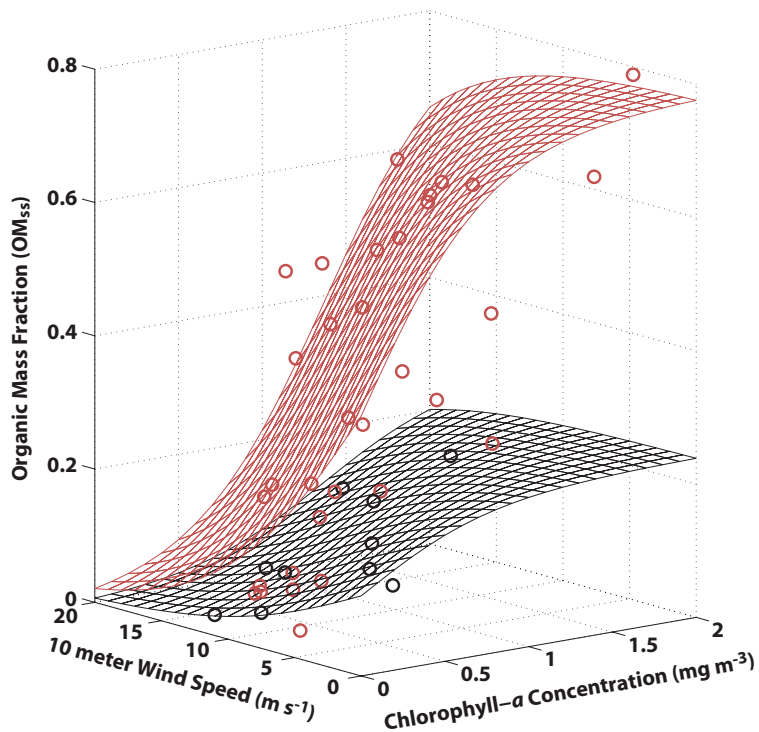


Fig. 3. Organic mass fraction of sea spray as a function of both 10 m wind speed and [Chl-*a*] for Mace Head (red) and Point Reyes (black) with the surface regression based on Eq. (1) in the same color scheme for each site.

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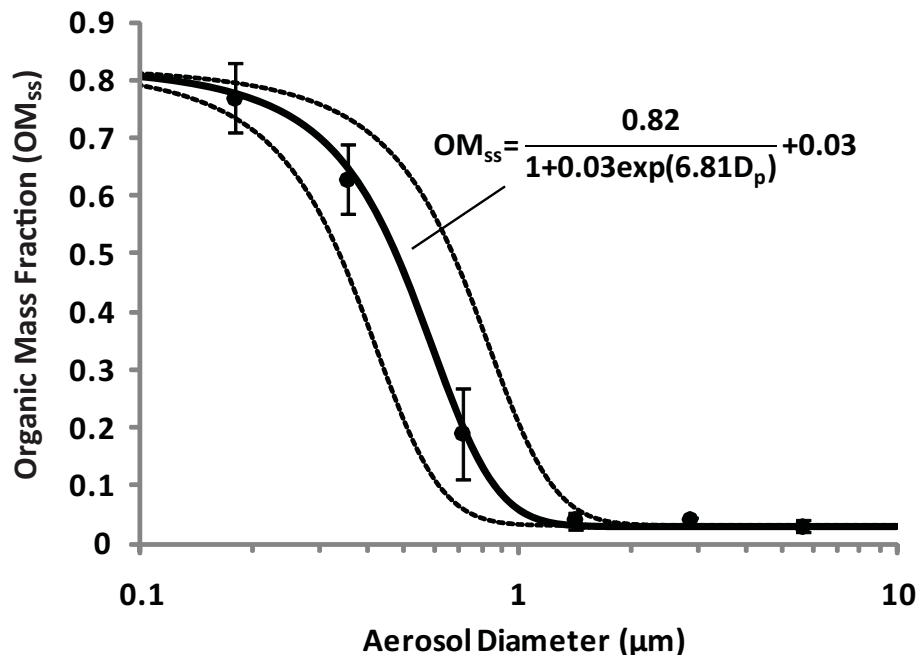


Fig. 4. Observed organic mass fraction of sea spray (solid circles) as a function of ambient aerosol aerodynamic diameter from Facchini et al. (2008). The solid curve shows a logistic fit to the geometric mean of the diameter range, with the dashed curves corresponding to the logistic fit of the high and low end of the measured aerodynamic size ranges. Vertical bars show one standard deviation from the mean as described in Facchini et al. (2008).

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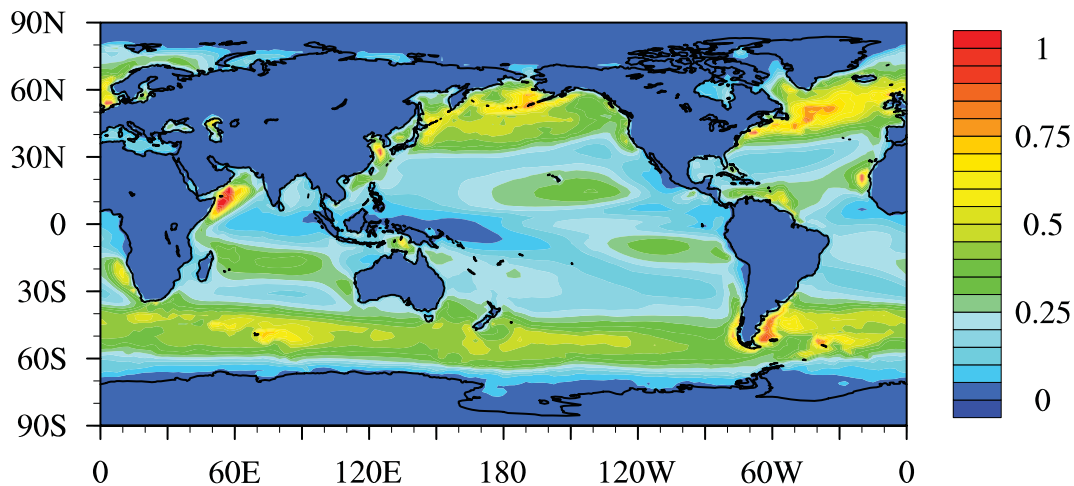


Fig. 6. Annual average submicron marine POA emission rate in units of $\text{ng C m}^{-2} \text{s}^{-1}$ using the Mårtensson et al. (2003) sea spray function, monthly average climatological [Chl-*a*] from SeaWiFS, and modeled U_{10} when global submicron marine POA emissions are 2.8 Tg C yr^{-1} .

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