

# Ocean Flux of Salt, Sulfate, and Organic Components to Atmospheric Aerosol

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## Abstract

The oceans contribute to aerosol particles in the atmosphere through two different physical mechanisms: first by the production of sea spray aerosol (SSA), and second by emitting gases that condense to produce secondary marine aerosol (SMA). These aerosol emissions include three types of chemical compounds: salt particles account for >90% of the mass, most of which is >1  $\mu\text{m}$  dry diameter; sulfate particles are <0.5  $\mu\text{m}$ , typically constituting most of the number and the largest impacts on clouds; organic components include the greatest variety of compounds and the most uncertain effects on clouds. Most SSA particles are expected to form from bubbles as film drops that are <1  $\mu\text{m}$  dry diameter and form from flapping bursting bubbles, although >1  $\mu\text{m}$  film drops can form by ligament fragmentation. SMA particles include contributions from marine biogenic gas emissions, including dimethylsulfide (DMS), isoprene, amines, and monoterpenes. The role of particles from the ocean in the atmosphere varies by region and by season, but since atmospheric concentrations of ocean-derived <1  $\mu\text{m}$  particles are typically much smaller than the concentrations of their continental counterparts, they have the largest impacts on climate in regions where continental sources are limited. Most efforts to quantify global SSA and SMA emissions rely on global models, where representations of marine aerosol sources are constrained by a small number of field measurements. Satellite-based retrievals of coarse and marine aerosol optical depth (AOD<sub>c</sub> and MAOD, respectively) provide near global coverage that has been linked to coincident wind speed, whitecaps, and biological productivity for >1  $\mu\text{m}$  particles. The current best estimate of SSA flux of 5000 Tg/yr can be used to calculate SSA-related carbon flux as 35 TgC/yr, by approximating <1  $\mu\text{m}$  SSA particles as 10% of SSA flux with 7% organic carbon and >1  $\mu\text{m}$  particles (as 90% of SSA flux with no organic carbon). SMA is estimated to contribute 0.6 TgC/yr as DMS, 0.6 TgC/yr as amines, and an additional trace amount from isoprene and monoterpenes for a total of <2 TgC/yr. Because of the limited availability of observations to constrain SSA and SMA global estimates, oceanic fluxes to aerosol and aerosol precursors could vary by over two orders of magnitude. Key open questions that require additional observational constraints include the variability in >1  $\mu\text{m}$  SSA mass size distributions, the relative contributions of SSA and SMA to large number concentrations of particles <0.5  $\mu\text{m}$ , and the regional and seasonal factors that may control these <0.5  $\mu\text{m}$  particle concentrations.

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## 1. Introduction: Ocean Sources of Atmospheric Aerosol Particles

Oceans cover two-thirds of the Earth, and they produce two sources of aerosol particles – one directly as particles (primary) and the other as gases that later form particles (secondary). These naturally occurring sources are produced from most of the area covered by the world oceans, but quantifying the contributions of these sources is a complex challenge limited by instrument and simulation capabilities, as well as by logistical obstacles for open-ocean measurements. The direct or primary source of particles to the atmosphere is that of sea spray aerosol (SSA), the particles formed from the bursting of bubbles generated by wind-driven breaking waves (Gong, 2003; Monahan, 1968). The indirect source of particles is the emission of gases from the ocean that can condense to particles after they are in the atmosphere, which are considered “secondary” marine aerosol (SMA) (Gantt et al., 2010; Meskhidze et al., 2010; Shaw, 1987; S. L. Shaw et al., 2010). SSA consists largely of sea salts from the ocean but also includes a contribution of organic components that are produced by the metabolism of biological organisms in the ocean. These ocean ecosystems also produce and release organic gases that can form SMA.

This review provides an update on the understanding of SSA and SMA, building from prior reviews (de Leeuw et al., 2011; Gantt & Meskhidze, 2013; Lewis & Schwartz, 2004; O'Dowd & De Leeuw, 2007; Quinn et al., 2015; S. L. Shaw et al., 2010). The objective is to summarize the findings most relevant to the physical drivers of marine aerosol sources and impacts, focusing on the fluxes of SSA and SMA. We consider the regional and seasonal differences that may influence SSA and SMA, including the role of ocean biota, and compile estimates of their global fluxes. Finally, we discuss the most important open questions for which answers are needed to improve our understanding of the role of ocean fluxes of aerosol in contributing to Earth's climate.

## **2. SSA Fluxes**

Aerosol particle fluxes from the ocean known as “sea spray aerosol” (SSA) are composed of inorganic sea salts, including mostly NaCl and trace amounts of other salts, and organic components (Gantt & Meskhidze, 2013; Russell et al., 2010). The flux of these particles has been characterized indirectly by measurements of particle concentrations in marine areas with minimal continental contributions, and have been shown to increase with wind speed (Monahan, 1968; Monahan et al., 1983). Laboratory measurements have quantified the links between bubbling of trapped air and particle production, showing that the flux may depend not only on the wind-speed-driven entrainment of air but also on the conditions of the sea surface and its composition (Blanchard, 1989; Martensson et al., 2003; K. Sellegri, C. D. O'Dowd, et al., 2006). The understanding of the SSA number size distribution is well developed for particles  $>1 \mu\text{m}$  but quite limited for particles  $<0.5 \mu\text{m}$ , which has substantial implications for SSA contributions to CCN (O'Dowd, Lowe, & Smith, 1999; O'Dowd, Lowe, Smith, et al., 1999).

### **2.1 Modes of SSA Production**

The modes of SSA have been associated with different processes involved in bubble bursting, with jet drops ejected from bubbles often attributed as the producers of particles 1-10  $\mu\text{m}$  dry diameter, film drops from bubble cap fragmentation as the

producers of smaller particles  $<1 \mu\text{m}$  dry diameter (Woolf et al., 1987), and spume drops from wave tearing as producers of particles at  $100 \mu\text{m}$  diameter (Anguelova et al., 1999; Fairall et al., 2009; Monahan et al., 2017; Veron, 2015; Wu, 1993). The formation processes for film and jet drops are illustrated in Figure 1, and the size ranges of film, jet, and spume particles are illustrated in Figure 2. The bubbles formed by wave-breaking in the ocean span a range of sizes from  $<50 \mu\text{m}$  radius to  $>10 \text{mm}$  radius (Figure 3), but there are few measurements characterizing bubble production in the open ocean (Deane & Stokes, 2002). Most of the quantitative relationships between bubble production and particle size distributions are based largely on laboratory rather than ocean conditions (Lewis & Schwartz, 2004). The scarcity of these measurements means that extrapolations to global and annual SSA are highly uncertain. This uncertainty means that current models use parameterizations rather than bottom-up calculations of sea spray particle production.

For this review as for most of the literature, bubble sizes are reported as observed geometric radius. Airborne particle sizes, in particular those related to chemical composition, typically are referenced to dry diameter to provide a more standard and reproducible metric for quantification. Aerosol particle diameter may be measured by mobility, aerodynamic, optical, or geometric properties, and here the diameter given is most often calculated as equivalent to geometric for spherical particles. Exceptions, as in Figure 2, are noted.

Bubbles larger than  $1 \text{mm}$  radius have been observed to produce film drops with a maximum dry diameter given by the area and thickness of the bubble cap (Lhuissier & Villermaux, 2012) and apparent minimum diameters as small as  $0.01 \mu\text{m}$  (Martensson et al., 2003). Photographic evidence indicated that ligaments form after the inertial destabilization of the rim enclosing the initial rupture in the bubble, and the elongation of those ligaments leads to the formation of droplets  $<10 \mu\text{m}$  wet diameter (Lhuissier & Villermaux, 2012; Spiel, 1998). Ligament-mediated (also called centripetal) fragmentation of the bubble film is thought to be relatively independent of bubble size (Lhuissier & Villermaux, 2012), producing a characteristic gamma distribution of drops (Spiel, 1998). The wet drops produced from bursting bubbles of radius  $12 \text{mm}$  were measured from  $5$  to  $500 \mu\text{m}$  (Lhuissier & Villermaux, 2012), which is approximately equivalent to dry diameters ranging from  $1$  to  $100 \mu\text{m}$  (Figure 4). High speed photographic observations account for the formation of particles  $<0.1 \mu\text{m}$  by capturing the role of flapping bubble films for bubbles  $<2 \text{mm}$  (X. H. Jiang et al., 2022). The flapping of the film begins as the bubble film recedes from the initial hole at the foot of the bubble cap, resulting in film fragmentation and drop formation. The flapping motion of the bubble film results from the thinness of the films for small bubbles and has been proposed to control the size of particles produced, with particles as small as  $0.04 \mu\text{m}$  from saltwater bubbles of radius  $73 \mu\text{m}$  (Gañán-Calvo, 2022; X. Jiang et al., 2022; X. H. Jiang et al., 2022).

Film drops have been thought to represent the majority (60-80%) of the particles produced in wave breaking (Veron, 2015). Laboratory measurements with controlled bubbling show production of many  $<1 \mu\text{m}$  particles from bubble bursting events

(Martensson et al., 2003). Those laboratory experiments included a range of bubble sizes, precluding the use of these experiments to establish a quantitative relationship between particle production and bubble film fragmentation. Recent evidence shows that bubbles smaller than 10-20  $\mu\text{m}$  radius produce particles with dry diameters 0.5-1  $\mu\text{m}$ , suggesting that jet drops may also play a role in  $<1$   $\mu\text{m}$  particle production (Wang et al., 2017), even though there are not yet open-ocean photographic measurements of bubbles smaller than 50  $\mu\text{m}$  radius (Deane & Stokes, 2002). Acoustic measurements of more complete bubble sizes have been captured in the open ocean, providing evidence of large numbers of particles as small as 10-20  $\mu\text{m}$  radius (Medwin, 1977; Medwin & Breitz, 1989; Vagle & Farmer, 1998). Smaller bubbles have been measured in tanks with plunging water (Stokes et al., 2016; Stokes et al., 2013), with particles from jet drops with dry diameters 0.5-1  $\mu\text{m}$  accounting for 20-40% of  $<1$   $\mu\text{m}$  particle number concentrations and the remainder being film drops with dry diameters 0.04-0.4  $\mu\text{m}$  (Wang et al., 2017).

Unlike film drops, jet drops tend to form particles that are determined by the bubble size (Blanchard, 1989; Wang et al., 2017; Wu, 2002). The “10% rule” for jet drops predicts jet drop dry diameter to be 5% of the bubble radius, and observations typically find 1-3 jet drops are produced per bubble (Veron, 2015; Wang et al., 2017). Contamination from surfactants has been shown to change the distribution of surface bubbles, thereby changing the production of jet drops (Neel et al., 2022). Interestingly, the mean size for film drops has been shown to be a function of the bubble radius and the ratio of the density of the bubble film to the enclosed gas, making its size dependent on the surface water composition and temperature (X. H. Jiang et al., 2022). An upper limit on film drop size can be calculated from the bubble cap volume assuming one film drop per bubble, but larger production is expected and broad size distributions have been observed (Martensson et al., 2003). Consequently, there are a wide range of particle production functions (Figure 5) that may reflect the role of environmental conditions on bubble bursting. Several studies have reported a variety of dependences of SSA production on ocean and atmospheric parameters (Table 1). One notable similarity across laboratory-generated size distributions (Martensson et al., 2003), coastal observations of vertical differences in concentrations (Clarke et al., 2003; Clarke et al., 2006), and shipboard bubbling proxies (Keene et al., 2017) is the presence of a particle mode  $<0.1$   $\mu\text{m}$  dry diameter. There are not yet direct measurements of  $<0.1$   $\mu\text{m}$  SSA flux in open-ocean conditions because of the lack of instrumentation that are chemically specific for sea salt, quantitative for sea salt mass concentration, and sufficiently resolved for sizes  $<0.1$   $\mu\text{m}$  at time resolutions of hours or less. The impacts of ocean (or laboratory) conditions on the bubble bursting process likely explain the variability between proposed sea spray production functions, which have been derived from laboratory generation of SSA (Figure 5).

Spume drops are the largest and shortest-lived contributors to aerosol particles (Figure 2), with sizes extending from 30 to 4000  $\mu\text{m}$  diameter (Erinin et al., 2022). At  $>20$   $\mu\text{m}$  diameter, their fall velocity often exceeds  $0.1$   $\text{m s}^{-1}$  making it unlikely for them to persist in the atmosphere. In addition, most chemical and physical measurements of aerosol particles are limited to diameters  $<10$   $\mu\text{m}$  because of the high loss rates of larger

particles in instrumentation, as discussed in Section 2.2.1. For these reasons, this review will focus on film and jet drops.

## 2.2 Approaches to Quantifying SSA Size Distributions

Direct measurements of SSA fluxes are generally not available for the  $<1 \mu\text{m}$  and  $>1 \mu\text{m}$  size distributions over most of the world oceans (Markuszewski et al., 2018; Norris et al., 2008), likely because of the combined challenges of designing instruments to measure sea salt components quantitatively at high time resolution and the low and dispersed ambient net flux rates. Instead, three complementary approaches have been used to identify the mass and number concentrations of SSA particles to open-ocean and coastal aerosol. The first and most chemically specific is size-resolved measurements of Na and Cl composition of particles, which can then be scaled to estimate inorganic “sea salt” (Quinn et al., 2000; Russell & Singh, 2006). The second is to infer the sea salt concentration based on its characteristic contribution to the size distribution and correlation to wind speed (Dedrick et al., 2022; R. L. Modini et al., 2015; P. K. Quinn et al., 2017; Georges Saliba et al., 2019). The third is to associate measured physical properties (such as volatility or hygroscopicity) of the size distribution with sea salt composition (Clarke et al., 2003; Clarke et al., 2006; Xu et al., 2022; Xu et al., 2021). The three methods vary in their uncertainty for different size ranges, their accuracy for number or mass, their time resolution, their labor intensiveness and consistency across groups, their reliance on assumptions that may be region-specific, and their ability to resolve non-salt contributions.

### 2.2.1 Chemical mass measurements

The most comprehensive and consistent set of size-resolved chemically-specific open-ocean sea salt measurements are available from a series of open-ocean cruises (<https://saga.pmel.noaa.gov/data/>). Size-resolved aerosol was collected and analyzed for  $\text{Na}^+$  and  $\text{Cl}^-$  with analysis by ion chromatography on more than a dozen open-ocean cruises by researchers at NOAA Pacific Marine Environmental Laboratory (PMEL) (Bates, 2009; Quinn & Bates, 2011; Quinn et al., 2019; Quinn et al., 2000). Compiling those concentrations from six different ocean cruises that had measurements quantifying the continental influence shows clear similarities with peaks in mass concentrations between 2 and 6  $\mu\text{m}$ , at  $\sim 3 \mu\text{m}$  aerodynamic dry diameter (Figure 6a). SSA mass concentrations have been shown to be correlated to wind speed in prior work (Lewis & Schwartz, 2004; Georges Saliba et al., 2019) but the mass distributions illustrate the consistency in the peak of the  $>1 \mu\text{m}$  mass distribution and reflect a strong degree of similarity of SSA modes across seasons and regions.

These mass size distributions are collected from six open-ocean cruises that also measured *in situ* tracers to exclude continental influences, namely  $<1 \mu\text{m}$  absorption (at 530 nm from PSAP) less than  $0.8 \text{ Mm}^{-1}$  and radon concentration less than  $540 \text{ mBq m}^{-3}$ . In this simple summary of measurements, organic contributions are excluded because they were not available for the seven stage impactor bins, and sea salt is scaled from measured  $\text{Na}^+$  and  $\text{Cl}^-$  and ammonium bisulfate from measured non sea salt sulfate ( $\text{SO}_4^{2-}$ ). The accuracy of the mass distributions is limited by the number and width of the impactor size bins, as well as by other sampling constraints, with an effective

compromise afforded by sampling from a 7-stage Berner impactor (Quinn et al., 2000). The mass concentration distributions illustrate the large mass concentrations  $>1 \mu\text{m}$ , with  $<1 \mu\text{m}$  bins contributing 10% or less (Kleefeld et al., 2002; Murphy et al., 1998; Quinn et al., 1998; Zheng et al., 2018).

The open-ocean sea salt mass concentrations show a weak correlation ( $0.12 < R^2 < 0.2$ ) of the  $>1 \mu\text{m}$  mass concentrations (and the summed mass from all bins) to the wind speed, but the correlations for the  $<1 \mu\text{m}$  bins have  $R^2 < 0.15$ . To quantify the size dependence on wind speed, the mass concentrations were normalized to wind speed and then the remaining variability was quantified as the ratio of the standard deviation to the mean. For  $>1 \mu\text{m}$  size bins, the standard deviations of the mass concentrations were near or below the means, making the ratio of standard deviation to mean approximately 1 or smaller (Figure 6d). For the  $<0.5 \mu\text{m}$  bins, the ratio of the standard deviations to the means of the mass concentration normalized to wind speed are all larger than 1. This high standard deviation relative to the mean of the wind speed normalized mass concentrations reflects an apparent weakness of the dependence of the  $<0.5 \mu\text{m}$  bin mass concentrations on wind speed. The low wind speed dependence for  $<0.5 \mu\text{m}$  particles is analogous to the higher correlations of sea spray particles to wind speed when  $<0.5 \mu\text{m}$  particles were excluded (Figure S17 of Saliba et al. 2019). The  $<0.5 \mu\text{m}$  sea salt mass concentration may still be produced from breaking waves, but the processes that produce the smaller particles may have a greater dependence on other environmental conditions (such as surface composition, whitecaps, sea surface temperature (SST), and coastal effects) than on wind speed. Smaller particles could also experience more influence from confounding sources and longer lifetimes. Such variability in conditions could explain the wide range of reported parameterizations in this size range (Figure 5).

The measured mass concentrations for each size bin can also be used to estimate the number concentrations, assuming as a starting point that the two different types of particles are externally mixed, with one consisting of sea salt and the other of ammonium bisulfate (Quinn et al., 2000), and using an effective density of 2.0 for sea salt (Saliba et al., 2020) and 1.78 for ammonium bisulfate. The largest uncertainty in this calculation of number concentrations arises from the width of the measured bins, which bounds the possible mean diameter of the particles within each bin. This sensitivity to the bin mean diameter is quantified here with three different estimates to bound the mean diameter: (1) an upper bound at the 50% cutoff diameter for the bin ( $D_{\text{high}} = D_{50}$ ), (2) a lower bound at approximately the 50% cutoff diameter of the next smaller bin ( $D_{\text{low}} = D_{50} - d \log D$ ), and (3) a best estimate at the geometric mean of these two values ( $D_{\text{mid}} = D_{50} - 0.5 d \log D$ ). This range of mean diameter means that the mass per particle varies by a factor of 5-9 for the bins with  $D_{50}$  of 0.31, 0.55, 1.06, 2.02, 4.13, and by factors of 12 for the smallest bin ( $D_{50}$  of 0.18) and 16 for the largest bin ( $D_{50}$  of 10.3). The uncertainty in the smallest bin has the largest effect on the number concentration, making the uncertainty in the number concentration a factor of 12 for the  $<0.18 \mu\text{m}$  bin. Since the number concentration in the  $<0.18 \mu\text{m}$  bin represents more than half of the summed 7-bin number concentration, the uncertainty in the sum is more than a factor of 12. This uncertainty is too large to use mass distributions to evaluate the absolute

number flux, but since the error in bin mean diameter is likely to affect the sea salt and ammonium bisulfate number concentrations in the same way, the relative fractions of sea salt and ammonium bisulfate can be compared with less uncertainty. By normalizing by the number per bin by the particles summed from all bins, for both sea salt and ammonium bisulfate particle types, the sensitivity to the mean diameter uncertainty is reduced (Figure 6c). The resulting number distributions show that ammonium bisulfate accounts for the majority of  $<0.5 \mu\text{m}$  particles in most projects (Figure 6c). In particular, the mean number concentration for the  $<0.18 \mu\text{m}$  bin range shows that sea salt accounts for 28% to 34% compared to 40% to 49% ammonium bisulfate particles (Figure 6c). If the assumption of two separate particle types is relaxed as is likely more realistic, then the  $<0.18 \mu\text{m}$  bin has particles with 44% sea salt and 56% ammonium bisulfate (neglecting organic and trace constituents). Such mixed particles could be classified either as SMA based on the majority of mass being ammonium bisulfate or as SSA based on the sea salt being the primary particles to which secondary mass was added.

### 2.2.2 Size distribution correlations

The size distribution correlation approach for estimating sea salt concentrations was developed to attribute modes fitted to merged submicron and supermicron size distributions to sea spray (R. L. Modini et al., 2015). The interpretation of particles as sea spray was supported by showing that the summed mass concentration increased with wind speed (Figure 7), from which a flux dependence on wind speed could be inferred (Georges Saliba et al., 2019). By incorporating the high size resolution from differential mobility measurements along with  $>1 \mu\text{m}$  optical or aerodynamic size distributions, and using simultaneous  $<1 \mu\text{m}$  chemical composition measurements for corroboration, the contribution of SSA to smaller sizes has also been evaluated. This approach showed that a single SSA mode with median diameter ranging from 0.05 to  $1.1 \mu\text{m}$  was correlated with  $<1.1 \mu\text{m}$   $\text{Na}^+$  and  $\text{Cl}^-$  concentrations as well as wind speed for a number of open-ocean campaigns (R. L. Modini et al., 2015; P. K. Quinn et al., 2017; Georges Saliba et al., 2019). Smaller modes were also retrieved simultaneously from each distribution, but comparisons to chemical measurements and wind speed indicated their source was largely sulfate and organic components (Figure 8) without a wind speed dependence (R. L. Modini et al., 2015; Georges Saliba et al., 2019).

Recent work has studied the accuracy of this approach in the absence of  $>1 \mu\text{m}$  size distribution measurements (Dedrick et al., 2022; Sanchez et al., 2020), showing that reasonable SSA number concentrations are retrieved with only  $<1 \mu\text{m}$  size distributions but that SSA mass concentrations are improved by constraining to coarse-mode scattering measurements (Dedrick et al., 2022). The range of retrieved sea spray modes does vary in not only number but also size and width (Table 2). The range of the reported parameters from each study does not appear to depend on the relative humidity of the measurements, suggesting the variability is greater than the dependence on hygroscopicity. The advantage of the size distribution-based approach is that it measures particle number concentrations directly with 10 bins or more per decade of diameter at high time resolution ( $<10 \text{ min}$ ), even though chemical composition is inferred. The correlation of retrieved SSA mode mass to measured  $<1.1$

$\mu\text{m}$   $\text{Na}^+$  and  $\text{Cl}^-$  composition provides indirect support for the chemical identification, although this approach is not expected to be sufficiently sensitive to identify particles  $<0.1 \mu\text{m}$ . The small and variable mass concentrations for particles  $<0.1 \mu\text{m}$  noted above may obscure any relationship between chemical composition and modes in this size range, which makes it difficult to rule out SSA  $<0.1 \mu\text{m}$  but also provides no support for the contribution of SSA in that size range.

### 2.2.3 Physical property proxies

The physical property proxy approach has used the low volatility or the high hygroscopicity of sea salt particles to identify particles with those properties as sea salt. The volatility approach separates the particles measured by optical or mobility size distributions by heating sufficient to evaporate sulfate and some organic components and identifying the remaining particles as nonvolatile SSA (Blot et al., 2013; Clarke et al., 1987; Clarke & Kapustin, 2003; Odowd & Smith, 1993). Measurements at three levels of a coastal tower were used to identify the SSA contribution by the amount the  $>0.04 \mu\text{m}$  concentrations at  $360^\circ\text{C}$  sampled at 5m above sea level (ASL) exceeded the 20m ASL concentrations, where the near-surface concentration exceedances at 5 m were interpreted as recent emissions of SSA in the coastal zone that had not yet mixed up to the 20m level (Clarke et al., 2003; Clarke & Kapustin, 2003; Clarke et al., 2006). The measurements showed a heated peak in the number size distribution at  $0.03 \mu\text{m}$ , which could result from evaporation of semi-volatile material from  $\sim 0.1 \mu\text{m}$ . These particles could be consistent with a mixture of the sea salt and sulfate mass concentration in the  $<0.18 \mu\text{m}$  bin measured by Quinn and colleagues (Figure 6c). To rule out the confounding effects of nonvolatile organic components that may be associated with quantifying the contribution from SSA, chemical measurements were used to show that SSA represented a small contribution to exclude measurements with significant organic or black carbon for open-ocean cruises (Blot et al., 2013). A disadvantage is that the heated size distributions measure particles at diameters reduced from their size at ambient temperature, making it necessary to back-calculate the ambient particle size distribution from the heated distribution.

Hygroscopicity has also been used as an indirect way to identify sea salt particles at a coastal site in the northeastern North Atlantic, showing that 2% of the measurements collected during a 5 year period have a large contribution to particles  $<0.2 \mu\text{m}$  dry diameter (Xu et al., 2022; Xu et al., 2021). This method relies on a cutoff value of  $\kappa=0.67$  for the size-selected hygroscopic tandem differential mobility analyzer measurements, which is interpreted as a mixture of organic components contributing less than half of the volume (assuming  $\kappa<0.1$  for organic components) and sea salt contributing the majority (with a 20% uncertainty from the uncertainty in its hygroscopicity for  $\kappa=1.0-1.2$ ) (Zieger et al., 2017). The interpretation of particles with  $\kappa>0.67$  as SSA assumes no contribution from ammonium bisulfate ( $\kappa=0.8$  for ammonium bisulfate), even though the ammonium bisulfate is typically measured in particles  $<0.18 \mu\text{m}$  in marine environments (Quinn et al., 2000). (Note that the hygroscopicity of ammonium bisulfate was estimated from Köhler theory, with the molality-dependent osmotic coefficient using an ion-interaction approach with tabulated



parameters (Pilinis & Seinfeld, 1987; Pitzer & Mayorga, 1973)). This interpretation of particles with  $\kappa > 0.67$  as sea salt results in a much higher SSA number concentration and contradicts the open-ocean results for the measured mass distributions (Figure 6a) and for the size distribution correlation approach (Figure 8). The high SSA concentration implies that there are no sulfate or organic particles contributing to CCN during the 2% of measurements selected. Certainly, this is possible for the 940 hr of measurements during a 5-yr period (with unspecified selection criteria), but the absence of supporting chemical correlations for the hygroscopicity identification suggest the results should be considered with caution. The sparse sampling of the extended data set suggests also that the apparent contradiction between this finding for a coastal location and those for the open-ocean studies, and the lack of correlation to wind speed, may indicate that regional and seasonal variability are important factors.

### **2.3 Composition of SSA (inorganic salts and organic components)**

Seawater includes a mixture of dissolved inorganic salts, with  $\text{Na}^+$  and  $\text{Cl}^-$  typically accounting for 86% of the inorganic mass and the remainder constituted primarily by  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{K}^+$ ,  $\text{SO}_4^{2-}$  (sulfate),  $\text{HCO}_3^-$ ,  $\text{Br}^-$ ,  $\text{B}^{3+}$ , and  $\text{F}^-$  (Holland, 1978). Together the mass contribution of these sea salts can be estimated as from the relation  $\text{Na}^+ + 1.47 \cdot \text{Cl}^-$  (Quinn et al., 2000). The contribution of trace salts to seawater means that the particle hygroscopicity is slightly lower than that of pure NaCl (de Leeuw et al., 2011; Ming & Russell, 2001; Tang et al., 1997; Zieger et al., 2017). The inorganic components of sea spray are expected to be generally refractory, which is the term used in aerosol mass spectrometry to indicate that they are largely nonvolatile when heated to 600°C. The presence of  $\text{SO}_4^{2-}$  as nearly 8% of the dissolved inorganic salts means that measurements of sulfate in marine aerosol will include both sulfate from seawater and sulfate from SMA and other secondary sources. To distinguish between these types of sulfate, the amount of sulfate associated with sea salt is calculated from the seawater composition and the remainder is designated as “non sea salt” sulfate. Analogous “non sea salt” quantities can also be defined for  $\text{Br}^-$  and  $\text{K}^+$ , in order to calculate the amount of these constituents that may be associated with wildfires, and for  $\text{Ca}^{2+}$ , in order to identify the amount that may be associated with dust (Bottenus et al., 2018; Gilardoni et al., 2009; Gilardoni et al., 2016; Song et al., 2021).

The presence of  $\text{Na}^+$  and  $\text{Cl}^-$  in the  $< 0.18 \mu\text{m}$  bin of the open-ocean ion chromatography measurements (Figure 6) is important because the presence of nonvolatile components can only be explained by the direct production of droplets during bubble bursting, namely there is no secondary source of  $\text{Na}^+$  and  $\text{Cl}^-$ . Reports of SSA particles  $< 0.03 \mu\text{m}$  dry diameter in coastal and lab measurements are consistent with a tail of larger modes, although separate modes of particles  $< 0.1 \mu\text{m}$  dry diameter have been reported (Clarke et al., 2003; Martensson et al., 2003; Xu et al., 2022). There are no direct chemical measurements in this size range, but hygroscopicity values less than that of salt and mass loss during volatility measurements indicate the measured modes are partially sea salt with the remainder including less hygroscopic and more volatile sulfate and organic components.

The ocean contains both dissolved (DOC) and particulate (POC) forms of organic carbon, which are usually defined operationally as the quantities that do (dissolved) and do not (particulate) pass through a submicron-sized pore filter when suspended in seawater (Carlson & Hansell, 2015). Several techniques have been used to identify organic components of SSA (Table 3). Both DOC and POC mix with sea salts when film and jet drops are formed (Gong, 2003; Walls & Bird, 2017), although the contributions of 20% on average (but up to 60%) organic components for 0.1-1  $\mu\text{m}$  fraction are likely associated primarily with film drops (Bates et al., 2020; Keene et al., 2017). In contrast, the  $>1 \mu\text{m}$  sizes have negligible organic contributions, with 100% of mass produced by bubbling proxies having a negligible organic volume fraction according to open-ocean bubbling experiments (Keene et al., 2017) and open-ocean ambient measurements (Quinn et al., 1998). The qualitative explanation for the size dependence of the organic contribution is that the thinner the bubble film (and the smaller the bubble), the larger the contribution of the organic surface constituents to the film composition (Figure 9)(Frossard, Russell, Burrows, et al., 2014; X. H. Jiang et al., 2022).

Many experiments using realistic physical simulations of the sea spray aerosol production process have shown that organic components can be measured in primary submicron SSA immediately or very shortly after emission, suggesting that at least some organic components enter the aerosol together with salt rather than through secondary formation from condensation of volatile gases (Alpert et al., 2017; Ault et al., 2013b; T. S. Bates, P. K. Quinn, A. A. Frossard, L. M. Russell, J. Hakala, T. Petaja, et al., 2012; Maria Cristina Facchini, Stefano Decesari, et al., 2008; Frossard, Russell, Burrows, et al., 2014; Gao et al., 2012; W. C. Keene et al., 2007; Kieber et al., 2016; Long et al., 2014; Quinn et al., 2014; Schmitt-Kopplin, Liger-Belair, Koch, Flerus, Kattner, Harir, Kanawati, Lucio, Tziotis, Hertkorn, & Gebefugi, 2012). The primary sea spray origin of marine organic components is also supported by laboratory studies and field experiments using a variety of analytical methods (including H-NMR, FTIR, FT-ICR, GC-MS, HF-ToF-AMS, EGA, IC, HPLC, TOC liquid analysis, Raman microspectroscopy, and AFM) (Ault et al., 2013b; Bondy et al., 2017; Frossard, Russell, Massoli, et al., 2014). Because inorganic sea salts are generally refractory, measurements of organic contributions to sea salt by non-refractory methods have been shown to miss the organic components on particles with sea salt (Frossard, Russell, Massoli, et al., 2014).

The organic components of seawater are expected to be produced as the metabolic byproducts of phytoplankton and other ocean biota, which also serve as nutrients for a variety of ocean organisms (Aluwihare et al., 1997; Burrows et al., 2014; Gantt et al., 2011). Some of the organic components are expected to track with the populations of these biota for which chlorophyll has been used as a proxy, however the variety of organisms and their differing (or nonexistent) amounts of chlorophyll make it problematic as a general tracer for all organic-producing organisms. The composition of the organic components of seawater is likely to include thousands of molecules that range from carbohydrates to amino sugars to lipids, many of which are macromolecules that are difficult to isolate or to characterize fully as molecular structures (Aluwihare et al., 1997; Aluwihare et al., 2005; Repeta et al., 2002). Individual molecules have been

speciated by Gas Chromatography Mass Spectrometry (GCMS) and other techniques, accounting for a few percent of the seawater composition (Maria Cristina Facchini, Stefano Decesari, et al., 2008; Maria Cristina Facchini, Matteo Rinaldi, et al., 2008; Gagosian, 1983; Lawler et al., 2020). Indirect characterization of the organic carbon in  $<1 \mu\text{m}$  marine aerosols has accounted for a larger proportion of organic components using functional group characterization (Russell et al., 2010). Subsequent work has shown consistently a majority of quantified organic mass can be characterized by alcohol groups with minor contributions from amine and alkane groups (Figure 10) (Frossard, Russell, Burrows, et al., 2014). This composition is consistent with amino sugars and more generally carbohydrates, although the uncertainties do not preclude contributions from lipids.

Primary contributions of amine groups are evident in  $<1 \mu\text{m}$  samples collected from proxies of bubble bursting and ambient particles (Berta et al., 2022 in preparation; Frossard, Russell, Burrows, et al., 2014). Measurements of the organic functional groups of marine aerosol particles  $<0.5 \mu\text{m}$  show some similarities to the  $<1 \mu\text{m}$  composition (Lewis et al., 2021). The higher variability and stronger contribution from secondary amine and acid groups  $<0.5 \mu\text{m}$  likely indicates a combination of primary and secondary marine sources to the aerosol (Berta et al., 2022 in preparation).

#### **2.4 SSA Contribution to Scattering, CCN, and INP**

SSA particles contribute to light scattering in the atmosphere, cloud condensation nuclei (CCN), and ice nucleating particles (INP). To quantify SSA impacts, a combination of laboratory measurements and clean marine observations have been incorporated in global transport and climate models using parameterizations that predict particle mass as functions of wind speed for a specified size distribution (Anguelova & Webster, 2006; Brumer et al., 2017; Gong, 2003). More sophisticated parameterizations have also incorporated dependence on white caps and SST. Verification of these parameterizations with observations has been promising but limited in scope due to the mismatch of time scales between available observations and global models (Figure 11) (Jaegle et al., 2011). Recent advances in satellite retrievals may improve these comparisons with new products for coarse and fine marine-related particle mass concentrations (Dasarathy et al., 2021; Dror et al., 2018), although chemical composition cannot be retrieved directly by satellite.

SSA is important in the atmosphere because of its direct contributions to albedo, as well as its indirect impacts on clouds and precipitation. The direct contribution is from the scattering of light, and this effect is controlled by the mass concentration of salt particles and their humidity-dependent hydration. Almost all of the particle mass ( $>90\%$ ) comes from particles  $>1 \mu\text{m}$  dry diameter (Figure 6a). The indirect effect on albedo is the result of the provision of CCN to clouds, which often scales with the number concentration of particles and provides the nucleation sites on which droplets form. Clouds respond to increases in CCN number immediately by an increase in cloud droplet number, resulting in brighter clouds (Twomey, 1977). A variety of “knock-on” effects and more subtle aerosol-cloud impacts also occur that can change the extent, depth, and optical thickness of clouds (Fan et al., 2016). These so-called “cloud adjustments” typically

enhance the sensitivity of Earth's climate to aerosol emissions (Sherwood et al., 2015). Supermicron salt particles can also act as "giant" CCN and affect precipitation (Dziekan et al., 2021; Feingold et al., 1999; Jensen & Lee, 2008; Jung et al., 2015; Reiche & Lasher-Trapp, 2010).

In addition, the glaciation of supercooled cloud droplets can be affected by particles that act as ice nucleating particles (INP), potentially impacting cloud evolution, precipitation, Earth's radiative budget, and cloud feedback, especially when emissions occur in remote regions such as the Arctic and Southern Ocean, where cloud phase feedbacks are important (Burrows, McCluskey, et al., 2022; Gettelman & Sherwood, 2016; Tan et al., 2022; Tan & Storelvmo, 2016; Tan et al., 2016; Vergara-Temprado et al., 2018).

The impacts of SSA on CCN, INP, and scattering depend strongly on SSA size distributions. CCN are primarily sensitive to number concentrations; the majority of sea spray particle number concentration is  $<1 \mu\text{m}$  dry diameter (Figure 6c). In contrast, marine INP are more sensitive to larger SSA; supermicron SSA particles contribute more effectively to INP populations but quantifying the impact of these larger SSA in the atmosphere remains challenging (Mitts et al., 2021; Steinke et al., 2022). Supermicron SSA also play an important role in scattering, although this effect is driven by mass rather than number concentration (Figure 6a). The different size dependences of these effects make it important to identify the size distribution of sea spray particles and their relative contribution to the atmosphere in open-ocean regions.

#### **2.4.1 SSA Contribution to Light Scattering**

Sea salt particles  $>1 \mu\text{m}$  have been shown to have a large role in scattering light in clean marine conditions, indicating a large role for wind-driven particles in scattering light over the ocean (Chamaillard et al., 2006; Covert et al., 1972; Kleefeld et al., 2000; O'Dowd et al., 2010; Quinn et al., 1998; Quinn et al., 2004). Satellite and surface-based aerosol optical depth (AOD) measurements in clear sky conditions have shown a dependence on local wind speed that is consistent with parameterizations based on *in situ* observations (Mulcahy et al., 2008; O'Dowd et al., 2010; Smirnov et al., 2012). Early retrievals based on Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 5 may have overestimated AOD at wind speeds  $>6 \text{ m/s}$ , due to wind-speed dependent changes in open-ocean surface roughness and white cap foam fraction (Kleidman et al., 2012). MODIS Collection 6 resolved this systematic bias by employing a wind speed look-up table (Kleidman et al., 2012; Levy et al., 2013). This revision produced a more modest dependence of AOD on wind speed over the open ocean (Merkulova et al., 2018). The remaining wind speed dependence, when combined with the observations of  $>1 \mu\text{m}$  particles accounting for 90% or more of sea spray mass (Section 3.2.1, Figure 6), indicate that satellite clear-sky retrievals of aerosol optical depth include signatures for wind-driven sea spray aerosol in marine regions.

Column retrievals of coarse-mode aerosol optical depth (AOD<sub>c</sub>) from MODIS are limited by cloud coverage and lack of vertical resolution (Ichoku et al., 2004), but recent approaches to marine aerosol optical depth (MAOD) have used the NASA Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) spaceborne lidar

to retrieve vertical profiles of layer-specific AOD (including the marine boundary layer) (Dasarathy et al., 2021; Kim et al., 2018). The lidar technique can measure under conditions that would challenge passive remote sensors, such as partial cloud cover and low sun angles. Recent work based on AODc and MAOD retrievals has investigated a seasonal biological contribution in the tropics and in the Antarctic, which show that the dependence on wind speed may be reduced or enhanced by biologically-produced surfactants in the summer (Dasarathy et al., 2022 in review; Dror et al., 2018). Comparisons with surface-based column optical depth measurements from AERONET have shown good correspondence between with satellite and *in situ* measurements (Asmi et al., 2018; Heslin-Rees et al., 2020; Holben et al., 2001; Kaufman et al., 2001; Porter et al., 2003; Smirnov et al., 2011; Virkkula et al., 2022).

#### **2.4.2 SSA as CCN**

The most direct measurements of SSA are chemical mass distributions, but CCN concentrations require the accurate size resolution afforded by number distributions. Calculating the contribution of SSA to CCN based on the chemical mass approach is uncertain, since the largest number of particles resides in the smallest bin. The estimated mean diameter of 0.12  $\mu\text{m}$  for the  $<0.18 \mu\text{m}$  bin is below the critical diameter of 0.13  $\mu\text{m}$  for sea salt particles for a supersaturation of 0.1% (Figure 12) (Sanchez et al., 2018), meaning that the high uncertainty in the  $<0.18 \mu\text{m}$  bin may not be relevant for low marine stratocumulus. However, for higher supersaturations the uncertainty in estimating number from mass reduces the utility of using size-resolved mass concentrations to estimate chemically specific number concentrations.

Using the size distribution correlation approach to evaluate the contribution of SSA to CCN has shown that SSA contributes a third or less of CCN in open-ocean clean marine conditions (Figure 13) (P. K. Quinn et al., 2017). This contribution is similar to the results suggested by the mass distribution-based calculation of number distribution in Figure 6c, which indicates  $<1 \mu\text{m}$  particles have an SSA contribution of 38% compared to 62% from SMA (ammonium bisulfate) for the Dmid mean diameters including the smallest bin with  $D_{\text{mid}}=0.12 \mu\text{m}$ . SMA is discussed in detail in Section 3, and the role of SMA in contributing to CCN is summarized in Section 3.3. If only particles larger than the SSA critical diameter of 0.13  $\mu\text{m}$  at 0.1% supersaturation are included, that would give contributions of 30% SSA and 70% SMA. The similarity of the open-ocean results from size-distribution correlations approach (Figure 13) and chemical mass measurements approach (based on the number distributions in Figure 6c) shows broad agreement across the first two approaches for open-ocean measurements from six cruises. The physical property approach with volatility found that SSA accounted for 20%-40% of CCN over the southeastern Pacific (Blot et al., 2013), consistent with the first two approaches described above. More larger particles, implying more CCN, were also attributed to entrainment of free tropospheric aerosol than to  $<1 \mu\text{m}$  SSA at Christmas Island (Clarke et al., 1996). A model-based estimate of CCN for the Southern Ocean was also consistent with this result, concluding that the seasonal variations in sulfate aerosol were primarily attributable to SMA rather than SSA (McCoy et al., 2015).

The physical property approach with hygroscopicity has identified a much higher contribution of SSA to CCN, estimated as 2 or 3 times more than the size distribution correlation approach (Xu et al., 2022). There is occasional evidence for this higher CCN contribution in estimated number concentrations from the open-ocean mass mode  $<0.18 \mu\text{m}$  (Figure 6c) for a few times in each project, possibly representing 2% or more of the sampling time. Such similar conditions could explain the Mace Head observations, although the high number contributions they reported do not seem sufficiently representative to use as the basis for a global mean parameterization for marine areas since the majority of time the number concentrations are generally lower for sea salt than for ammonium bisulfate.

While satellite-derived AOD has retrieved reasonable estimates of the SSA mass concentration, the large contribution of relatively few  $>1 \mu\text{m}$  SSA to the light scattering limits its ability to quantify the more numerous and CCN-relevant  $<1 \mu\text{m}$  SMA and SSA particles. Recent advances in relating AOD to fine particle mass concentration in urban environments using high-spectral resolution lidar (Sawamura et al., 2017) are not effective in the presence of the coarse mode aerosol found in marine environments. However, combining satellite retrievals of cloud drop number concentrations (Painemal et al., 2020; Painemal et al., 2021) with AOD retrievals may provide an approach to quantifying CCN number concentrations.

#### **2.4.3 SSA as Ice Nuclei**

Although SSA is largely comprised of inorganic salts that tend to suppress ice formation, SSA can provide a source of ice-nucleating particles (INP). In particular, the inclusion of organic and biological components appears to play a critical role in enabling SSA to act as INP, thereby enhancing the immersion-mode nucleation of supercooled droplets to ice (DeMott et al., 2016). The ice-nucleating entities that contribute to the INP activity of SSA may be associated with the sea surface microlayer (Wilson et al., 2015), and the presence of marine INP in the ambient atmosphere may correlate to ocean biological indicators (Trueblood et al., 2021).

Marine INP are typically less effective and more scarce than dust or terrestrial biological particles (DeMott et al., 2016; Ickes et al., 2020; Irish et al., 2017; Irish et al., 2019; McCluskey, Hill, et al., 2018; Twohy et al., 2021; Wex et al., 2019). Specifically, results from a wave tank experiment showed that SSA particles have  $<1\%$  of the ice nucleation effectiveness (nucleating sites per particle area) compared to continental particles (DeMott et al., 2016). In addition, INP concentrations in clean marine regions such as the Southern Ocean are two or more orders of magnitude smaller than INP concentrations in continental air (McCluskey, Hill, et al., 2018). However, both models and observations have shown that SSA particles can serve as INP in the absence of better nuclei such as dust (Chen et al., 2021; DeMott et al., 2016; McCluskey, Hill, et al., 2018; Vergara-Temprado et al., 2017; T. W. Wilson et al., 2015). In addition, observations have identified episodes in which marine INP provide the main INP source along the coastlines of the North Atlantic and Pacific Oceans (Cornwell et al., 2019; McCluskey, Ovadnevaite, et al., 2018). Given the limited role of SSA as INP when

better INP such as dust are present, it is not yet possible to quantify the overall role of SSA as INP globally.

In addition to acting as INP when immersed in supercooled cloud droplets, some constituents of SSA have been shown to be capable of contributing to ice formation by serving as nuclei for the deposition of water vapor, a mode of ice nucleation that can be important in the upper troposphere (Knopf et al., 2011; Wolf et al., 2019). However, observations of this ice nucleation mode for ambient SSA are limited, and it is unclear how to extrapolate from the observed ice nucleation behavior of individual constituents, to complex ambient SSA particles. Few direct observations are available of particle chemical composition and INP in the upper troposphere. Consequently, constraining the impacts of SSA on deposition-mode ice nucleation remains difficult.

Global models have been used as a tool to estimate climate impacts of INP, but such models are not yet capable of representing all of the processes important in controlling the climate response to INP at the necessary scales. More research is needed to better quantify SSA contributions to INP in order to assess which areas of the Earth would be most sensitive to SSA contributions to INP (Burrows, McCluskey, et al., 2022).

### **3. SMA Fluxes**

The contributions of gases to the aerosol are considered “secondary” because they are emitted in the vapor phase and only form liquid or solid particles after atmospheric processing, which typically includes oxidation to convert the emissions into less volatile compounds. Several approaches have been used to distinguish secondary contributions of SMA from the primary contributions of SSA (Table 4). Chemically-specific contributors are methanesulfonate (MSA) and non-sea-salt sulfate, which are known to form as secondary aerosol from DMS (Quinn & Bates, 2011; Sanchez et al., 2018). A variety of organic compounds also contribute to SMA.

#### **3.1 Marine Gas Emissions**

DMS, which is produced from dimethylsulfoniopropionate (DMSP) (Hatton & Wilson, 2007; Stefels et al., 2007), is the sole source of MSA and the marine source of sulfate in the atmosphere. DMS emissions are poorly quantified at the global scale, and climatologies of their emissions are heavily dependent on sparse observations (Bock et al., 2021; Lana et al., 2011). A recently proposed satellite-based parameterization uses Chlorophyll-a, ocean mixed-layer depth, photosynthetically available radiation, and SST as inputs to an algorithm that predicts ocean DMS concentrations (Gali et al., 2018). DMS emissions have been increasing at high latitudes as Arctic sea ice recedes, which could mean that additional sulfate aerosol serving as CCN would have a disproportionate impact on global climate by slowing the loss of Arctic ice (Gali et al., 2019; Lelievre, 2013). Prediction of future DMS is complicated by shifts in phytoplankton community composition (Wang et al., 2018).

Other ocean-emitted gases that may contribute to SMA include isoprene, monoterpenes, iodine, and amines (Maria Cristina Facchini, Stefano Decesari, et al., 2008; Gantt et al., 2010; O'Dowd et al., 2002; Shaw, 1987), but there are very limited

observations of the magnitude of their contributions. The limited observations that are available suggest that the contributions of these gases to SMA production are minor in comparison to the contributions from DMS. For example, Kim et al. (2017) conducted ship-board eddy covariance measurements of the vertical flux of isoprene, monoterpenes, and DMS over the North Atlantic during fall, and calculated the SMA production rates from DMS to be ten times greater than production from either isoprene or monoterpenes. Other ocean-emitted gases, such as acetone, acetaldehyde, and alkyl nitrates, do not contribute significantly to SMA, but have important impacts on OH, HO<sub>x</sub>, and NO<sub>x</sub> chemical reactions (Novak & Bertram, 2020). More details on the ocean production of volatile organic compounds (VOCs) are provided by Halsey (this issue). After production, gases in the ocean are transferred to the atmosphere by wind-driven, bubble-mediated sea-to-air transfer processes, which have been reviewed elsewhere (Blomquist et al., 2017; Blomquist et al., 2014; Brumer et al., 2017; Garbe et al., 2014; Liss & Slater, 1974; Liss et al., 1993; Yu, 2019; Zavorsky & Marandino, 2019).

### **3.2 Secondary Marine Aerosol Composition**

The organic components of SMA are typically formed from the oxidation and condensation of oceanic volatile precursor gases emitted from the ocean that contribute to observed sulfate and organic mass in marine aerosol (Ceburnis et al., 2008a; Maria Cristina Facchini, Stefano Decesari, et al., 2008; Facchini, Decesari, et al., 2010; Maria Cristina Facchini, Matteo Rinaldi, et al., 2008; P. Fu et al., 2013; Meskhidze & Nenes, 2006; Colin D. O'Dowd et al., 2004; M. Rinaldi et al., 2010; S. L. Shaw et al., 2010; Turekian et al., 2003). The Quinn and colleagues measurements support these observations over a broad range of clean marine conditions (Figure 6b) (Quinn et al., 2000; P. K. Quinn et al., 2017), with non-sea salt sulfate providing the largest contribution to <1 μm inorganic mass concentration, consistent with studies in a variety of ocean conditions (Sanchez et al., 2018; Sanchez et al., 2016; Twohy et al., 2021). Some organic mass contributions for <1 μm particles have also been identified as SMA based on their carboxylic acid group composition, their non-refractory molecular fragments, and their increases with shortwave radiation (Frossard, Russell, Burrows, et al., 2014; Lewis et al., 2021; Saliba et al., 2020). Isoprene oxidation products include molecules with carboxylic acid groups (Clafin et al., 2021; Ziemann & Atkinson, 2012) and may be consistent with an ocean source, although field observations may also have contributions from continental sources. Amines have been shown to have both primary and secondary contributions to marine aerosol (Maria Cristina Facchini, Stefano Decesari, et al., 2008).

There is recent evidence suggesting that secondary products have larger relative contributions to particle fractions <0.5 μm than to <1 μm (Sanchez et al., 2018), which is consistent with expected contributions for condensational growth (Maria et al., 2004; Seinfeld & Pandis, 2016). Volatile gases can also be produced by photosensitized heterogeneous reactions at the air-water interface, potentially leading to formation of SMA (Bernard et al., 2016; Tinel et al., 2016). Detailed chemical analyses of marine aerosol at coastal sites have shown organic and sulfate as SMA contributions, including a study at Amsterdam Island that showed that water-soluble organic matter (WSOM) accounted for 2.8% of fine aerosol mass and was predominantly from methanesulfonate



(MSA). Additional WSOM was attributed to oxidation of fatty acid residues from SSA (Claeys et al., 2010).

Open-ocean measurements in the North Atlantic in four seasons showed SMA in remote marine aerosol during late spring and summer months (Lewis et al., 2021; Saliba et al., 2020; Sanchez et al., 2018), despite relatively low measured rates of local production of dimethyl sulfide and isoprene (among other VOCs) (Davie-Martin et al., 2020). The apparent inconsistency of low DMS with substantial submicron sulfate may be attributed to lofting to the free troposphere, where colder conditions with lower particle concentrations enhance gas-to-particle transfer (Clarke et al., 1999; Clarke et al., 2013; Raes, 1995; Sanchez et al., 2018). The SMA signature of carboxylic acid groups by FTIR was clearly present in May-June and September, but almost entirely absent in November and March (Lewis et al., 2021). This seasonal dependence is generally consistent with the chlorophyll and sunlight-based emission parameterization used in global models (Myriokefalitakis et al., 2010). Observed carboxylic acid group mass of 10% of marine organic mass implies a larger vapor-phase precursor concentration than was explained by measured DMS or isoprene (Saliba et al., 2020). Carboxylic acid groups are a common component of SMA from isoprene and monoterpenes (Claflin et al., 2021; Lewis et al., 2021), but the lack of simultaneous precursor measurements precludes a specific source attribution.

### **3.3 Secondary Marine Aerosol as CCN**

SMA may provide crucial increases in particle size that allow SSA and other particles emitted at sizes  $<0.1 \mu\text{m}$  dry diameter to grow to sizes large enough to serve as CCN at water vapor supersaturations of  $<0.2\%$  (Robert J. Charlson et al., 1987; Sanchez et al., 2018). SMA can either add new particles by homogeneous nucleation or can grow particles from sizes too small to be CCN (generally  $<0.1 \mu\text{m}$  for  $0.1\%$  supersaturation) to sizes large enough to be CCN. While there is little evidence for new particle formation occurring in the marine boundary layer given the competing sink of aerosol surface area (Quinn & Bates, 2011), exchange between the free troposphere and the boundary layer may provide transported or nucleated particles that can be grown into CCN by SMA (Zheng et al., 2018; Zheng et al., 2021). The magnitude of this contribution to CCN varies with the condensable gases, ambient temperature and relative humidity, and the availability of other sinks. For example, in the North Atlantic, the seasonal difference in the role of SMA as CCN varies from sulfate contributing 34% of CCN at  $0.1\%$  supersaturation in windy fall compared to 64% in the sunny late spring, both to wind-driven differences in the SSA produced and differences in the DMS emitted and oxidized (Sanchez et al., 2018). For cloud supersaturations  $>0.1\%$ , the contribution of SMA to CCN number concentration would be expected to increase given the typically high concentrations of particles  $<1 \mu\text{m}$ .

## **4. Regional Ocean Differences and other Controlling Factors**

Parameterizations of SSA rely on empirical measurements rather than a theoretical formulation, since even idealized bubble bursting process simulations at micrometer scales have not quantified particle production (Bird et al., 2010; Lhuissier & Villermaux, 2012; Veron et al., 2012). Ocean conditions include diverse processes at many scales

that interact with small scale processes. In addition, ocean ecosystems differ across ocean regions (Burrows et al., 2018; Burrows, Easter, et al., 2022; Burrows et al., 2014), with different organic components of sea spray contributing compounds from different ecosystem populations (Figure 14). There is considerable evidence that SSA and SMA have some similar properties globally, including the general similarity of the mass size distributions of sea salt and non-sea salt sulfate in regions as diverse as the North Atlantic and the Southern Ocean (Figure 6a). However, in the absence of *a priori* (bottom up) predictions of either SSA or SMA, it is reasonable to consider that differences in regions, seasons, and other environmental conditions influence both the concentrations of aerosol sources and their sizes.

Future climate may also bring significant changes in SSA associated with changes in surface wind speeds. SSA emissions will change as surface wind speeds are either increased or decreased by a warming climate and a recovering ozone layer (Gettelman et al., 2016; Korhonen et al., 2010). In addition, the retreat of sea ice and ice shelves leaves more ocean surface exposed and increases the emissions of SSA, although the net climate effects of such an increase in emissions are uncertain (Browse et al., 2014; Struthers et al., 2011).

#### **4.1 Open-Ocean, Coastal, and Polar Regions of Marine Aerosol**

Marine regions include three distinct types of ocean areas, namely open-ocean, coastal, and polar regions. Open-ocean areas are expected to be characterized by large and relatively homogeneous expanses of wave breaking, with consistent near-surface wind speeds and temperatures. Coastal regions are generally expected to be influenced by open-ocean conditions upwind, with only recent production of SSA and SMA affected by shore-influenced wave breaking (Clarke et al., 2003). Polar regions may have a mix of breaking waves as well as processes associated with sea ice and open-lead formation, below-ice ecosystems, and resuspension of deposited SSA and SMA as blowing snow or frost flowers (Abbatt et al., 2019; Chang et al., 2011; Frey et al., 2020).

Open-ocean measurements are very limited given the deployment costs, but the largest fraction of the area of global oceans is expected to be represented by such measurements. No direct flux measurements are available for the open ocean, although proxies from bubbled seawater have been collected underway on the open ocean to determine fluxes (Keene et al., 2017). The chemical composition measurements of Quinn and colleagues (Figure 6) provide a consistent  $>1 \mu\text{m}$  SSA size distribution signature across many regions, with a mass concentration peak at  $\sim 3 \mu\text{m}$  dry diameter. Their measurements have also characterized the role of SMA in the open ocean, typically showing an  $\text{NH}_4^+/\text{SO}_4^{2-}$  molar ratio of 1, consistent with ammonium bisulfate.

Substantial differences between open-ocean SSA (interpreted as aerosol measured at 20 m during onshore winds) and near-shore SSA (measured as the difference between 5 m and 20 m ASL) were observed at Hawaii. Their observations showed a majority of particles at 5 m ASL were smaller than  $0.1 \mu\text{m}$  dry diameter (Clarke et al., 2003), similar to the inferred fluxes at a North Atlantic coastal site at Mace Head (Xu et al., 2022). There is some support for a small particle mode similar to that observed in coastal

studies in open-ocean observations (<0.18  $\mu\text{m}$  bin, Figure 6a) and laboratory studies, but this SSA mode could also be generated by coastal dynamics since it is not evident in open-ocean studies.

SSA in polar regions has a strong seasonal variation with sea ice coverage, because sea ice reduces the area of breaking waves in winter when the wind speeds are expected to be highest (Liu et al., 2018; P. M. Shaw et al., 2010). Polar regions are very different between the north and the south, with Antarctic measurements lacking the springtime haze events that confound measurements of SSA and SMA in the Arctic (Lubin et al., 2020). The unique biogeochemistry associated with seasonal sea ice could also result in differences in SSA and SMA to a greater extent than at the milder lower latitudes.

#### **4.2 Temperature Dependence**

Wave breaking and the subsequent bursting of bubbles is controlled by the fluid motion of seawater at the ocean surface (Veron, 2015). The sea surface temperature changes the density, viscosity, and surface tension of the fluid, of which the viscosity is likely the most sensitive parameter for the film rupture process (Lhuissier & Villermaux, 2012). Laboratory experiments demonstrated the role that temperature played in changing the size distribution of bubble-generated particles, showing a significant increase in flux with temperature (Martensson et al., 2003). Incorporating this effect in a global model showed that this increase improved the model correspondence to open-ocean SSA measurements (Jaegle et al., 2011). Further evidence of the effect of SST on SSA is provided by four seasons of open-ocean measurements in the North Atlantic, where the mean size of the SSA mode varied with SST (Liu et al., 2021; Georges Saliba et al., 2019).

#### **4.3 Whitecap Dependence**

Persistent bubbles on the ocean surface appear as “whitecaps” that can affect the persistence and bursting of bubbles (Andreas & Monahan, 2000; Monahan et al., 1983). Whitecaps are difficult to replicate in laboratory measurements, although foam persistence has been noted to change SSA production (Keene et al., 2017; Stokes et al., 2016; Stokes et al., 2013). Open-ocean observations of whitecaps have also been carried out (Callaghan et al., 2008), but correlations to SSA are difficult because of the mismatch in the spatial scale of the observed whitecaps and the fetch of the SSA produced (Hoppel et al., 2002). Retrievals of whitecaps from satellite may improve parameterizations for SSA (Albert et al., 2016; Angelova & Webster, 2006; Salisbury et al., 2013).

#### **4.4 Sea Surface Microlayer**

Rudimentary separation of a lighter, more surface-active layer in the top millimeter of the ocean from the seawater below has been accomplished with rotating drums and simple mesh screens (Miller et al., 2015). These methods are not able to quantify the thickness of the layer collected, but the composition and properties of the material have been shown to be substantially different from the seawater below (Aller et al., 2017; Crocker et al., 2022; Ickes et al., 2020; Irish et al., 2017; Lewis et al., 2022; Mungall et

al., 2017; Theodore W. Wilson et al., 2015). The location of this layer at the sea surface means that it is expected to affect the bubble formation and bursting processes, unless sufficiently disturbed by wave-breaking processes. Modeling of bubble drainage shows an important role for contamination of the seawater surface (Poulain et al., 2018). Laboratory experiments have included proxies for this layer, from simple surfactants to complex biological mixtures (Modini et al., 2013; K. A. Prather et al., 2013).

Comparison of the organic functional group signature in the FTIR alcohol group absorbance of the sea surface microlayer reveals a heterogeneity from day to day that is consistent with other microlayer measurements and with the atmospheric aerosol composition measured in clean conditions (Lewis et al., 2022). Similarities in composition of  $<1 \mu\text{m}$  marine aerosol and the sea surface microlayer have included the presence of TEP and similar moieties from biological sources (Maria Cristina Facchini, Stefano Decesari, et al., 2008). The variability in the shape and peak locations of the alcohol group absorbance in the microlayer and aerosol particles supports a role for the microlayer in SSA formation, although the quantitative ratios of alcohol to alkane and amine group mass concentrations are also generally consistent with the subsurface water composition (Lewis et al., 2022). Combined these results indicate that both subsurface seawater and sea surface microlayers contribute to SSA formation, consistent with general expectations.

#### **4.5 Biological Diversity**

Since ocean ecosystems produce organic compounds that contribute to subsurface and sea surface composition, SSA and SMA will be affected by regional and seasonal differences in those ecosystems. Ocean ecosystems in the “polar”, “westerlies” (mid-latitude), “trades” (equatorial) and “coastal” biomes are controlled by different processes and exhibit different seasonality (Longhurst, 1995). The dominant phytoplankton functional groups and their seasonal cycles also vary between ocean biomes (Figure 15)(Alvain et al., 2008). In particular, the Phaeocystis group, which is a dominant group only at high latitudes, has long been recognized as an important contributor to atmospheric DMS emissions (Belviso et al., 2004; Liss et al., 1994).

Several studies have supported the connection between ecosystems and marine aerosol by showing correlations between SSA and Chlorophyll (Colin D. O'Dowd et al., 2004), but limited observations from open-ocean studies have shown no dependence on daily time scales and only a weak dependence on seasonal time scales (Bates et al., 2020; Saliba et al., 2020). A metric more specific to overall biological productivity is net primary productivity (NPP), which has also been shown to explain some of the variability in SSA (Lewis et al., 2022; Georges Saliba et al., 2019). Observation-based efforts to link SMA to either Chl or NPP have not been successful, perhaps due to the time lag and complexity of those processes (K. J. Sanchez et al., 2021). However, bottom-up modeling of ocean ecosystems has shown that the seasonal cycle in the North Atlantic for ocean biological effects on sea spray may have a narrow maximum that occurs in July (Figure 16) rather than during the peak in chlorophyll intensity that was sampled in May (Burrows, Easter, et al., 2022) (Burrows et al., 2022). More complete sampling of the seasonal dependence, as well as the latitude dependence, of open-ocean

measurements of SSA would be required to observe the seasonal cycles predicted by models and observed at coastal sites (Rinaldi et al., 2013; Sciare et al., 2009).

## 5. SSA and SMA Carbon Budget

The best model-based estimates for the global emissions of primary marine organic mass (OM) span the range of 6.9 – 76 Tg/yr for  $<1 \mu\text{m}$  emissions and 7.5–58 Tg/yr for  $>1 \mu\text{m}$  emissions (using an organic mass to organic carbon ratio of 3 to convert where appropriate) (Gantt et al., 2011; Ito & Kawamiya, 2010; Long et al., 2011; Myriokefalitakis et al., 2010; Spracklen et al., 2008; Vignati et al., 2010; Westervelt et al., 2012), with earlier estimates reviewed previously (Tsigaridis et al., 2013). These studies used both different emissions parameterizations and different model systems, which may impact the simulated atmospheric residence time of SSA and SMA. As a simple comparison, we can use the open-ocean measurements from Quinn and colleagues (Figure 6) to see that  $>90\%$  of  $<10 \mu\text{m}$  sea salt mass concentration is coarse ( $>1 \mu\text{m}$ ), with the remaining  $<1 \mu\text{m}$  contribution ranging from 3% to 10%. The average ambient organic fraction of SSA  $<1 \mu\text{m}$  is 20% OM or 7% OC. Using the global SSA estimate of 5000 Tg/yr, organic components account for 7% of  $<1 \mu\text{m}$  SSA at 105 Tg/yr (35 TgC/yr), which is within the range from the models noted above for the combined  $<1 \mu\text{m}$  and  $>1 \mu\text{m}$  emissions of 134 Tg/yr. If  $>1 \mu\text{m}$  organic carbon measurements have shown negligible contributions to SSA mass as has been typically reported (Keene et al., 2017), then the  $<1 \mu\text{m}$  organic mass flux estimated here is within the model-estimated range.

Secondary aerosol formed from DMS oxidation products is an important aerosol source in remote marine regions (Ayers & Gras, 1991; Charlson et al., 1992). The recent multi-model estimate of contemporary global ocean DMS emissions is 16–24 TgS/yr, even though the observation-derived range of 16 to 28 TgS/yr. Global sources of SOA from oceanic precursor gases are thought to be substantially smaller than primary organic. This is especially true of sources other than DMS and its oxidation products, which are already included in models. According to one set of estimates, marine volatile organic carbon emissions contribute less than 2 TgC/yr to organic SMA globally, with DMS oxidation contributing about 0.6 TgC/yr (Quinn et al., 2015). Combined amine compounds contribute approximately 0.6 TgC/yr and isoprene and monoterpenes up to 1 TgC/yr (Quinn et al., 2015).

Oxidation products of marine isoprene may contribute a substantial fraction (over 30%) of organic aerosol mass over oceans in the tropics, where the SSA source is small due to low wind speeds (Gantt et al., 2009). The SMA contribution to marine organic carbon at mid- and high latitudes, and globally, was minor,  $<0.2\%$  of global marine organic aerosol mass (Gantt et al., 2009). SMA from marine hydrocarbon precursors was found to be negligible compared to the estimated marine  $<1 \mu\text{m}$  organic SMA source of about 5 Tg/yr, but this flux consisted primarily of DMS oxidation products (78%), with most of the remaining mass originating from dialkyl amine salts (21%) (Myriokefalitakis et al., 2010).

These fluxes of 35 TgC/yr as SSA and 1.5 TgC/yr as SMA to the atmosphere are expected to have a lifetime of 5-7 days in the marine boundary layer, after which time they will generally be deposited to the surface with a small fraction transported upwards to the free troposphere. A general estimate based on the ocean-covered area of the Earth would be that more than 70% would be deposited back to the ocean and the remaining fraction would be deposited to land. The resulting flux from ocean to land may be about 30% of 37 TgC/yr, or 11 TgC/yr.

## 6. Open Questions

SSA plays an important role in light scattering in Earth's atmosphere, contributing approximately 5000 Tg/yr of mostly  $>1 \mu\text{m}$  particle mass. *In situ* measurements and satellite retrievals show a clear wind speed dependence of these particles, providing reasonable constraints for global models. The uncertainties in satellite retrievals of SSA-specific AOD prevents global inventories from including accurate size distributions, so *in situ* measurements of SSA provide important constraints. Consequently whether the SSA coarse mode is correctly represented by satellite AOD retrievals and wind speed parameterizations merits additional evaluation (Schutgens et al., 2021), and observationally-constrained global modeling simulations could reduce the uncertainty associated with this question. Additional measurements of  $>1 \mu\text{m}$  mass size distributions would be essential for improving constraints on SSA radiative effects.

Open-ocean measurements of SSA and SMA are limited in availability and in specificity of either number concentration or chemical composition, especially for particles  $<0.5 \mu\text{m}$  and  $<0.1 \mu\text{m}$  dry diameter. These open-ocean reports include sea salt particle modes  $<0.5 \mu\text{m}$  (Figure 6b) similar to coastal and laboratory reports. However, the open-ocean observations of SSA  $<0.5 \mu\text{m}$  do not show a clear dependence on wind speed and vary substantially with region and time (Figure 6d). These limitations and the contrasting observations from open-ocean studies mean that there remain several open questions about the magnitude and contribution of  $<0.5 \mu\text{m}$  and  $<0.1 \mu\text{m}$  particles, as well as very little information on the extent to which wind speed or other factors control their production. Specifically, there are no direct measurements of how many CCN are SSA or SMA, so it is an open question whether they contribute 10%-30% of CCN or much higher numbers globally. Open-ocean observations with simultaneous measurements of size-resolved composition and hygroscopic and volatility properties could help to close the gap on these questions. More complete sampling of the open-ocean seasonal cycle could also address the extent to which ocean biology impacts SSA composition. Combining satellite retrievals of cloud drop number concentrations (Painemal et al., 2020; Painemal et al., 2021) with AOD-based fine particle mass concentrations (Sawamura et al., 2017) may also make it possible to continuously retrieve marine CCN, or possibly even SMA and SSA, number concentrations from remote regions with continued global coverage.

While the mechanism and size dependence for producing most SSA  $>1 \mu\text{m}$  particles is expected to be jet drops at 10% of the bubble radius, particles smaller than  $0.5 \mu\text{m}$  are produced from film drops that extend over a wide range of sizes for a given bubble size. This process is also likely affected by regional and seasonal conditions, with an

important open question relating to the mechanistic role of the sea surface microlayer. Further work on theoretical approaches and numerical simulations is needed to understand these processes, with supporting observations from atmospheric and ocean conditions.

### **7. Acknowledgements**

This work was funded by NASA grant NNX15AE66G and by the U.S. Department of Energy under Contract DE-SC0021045. SB was supported by the U.S. Department of Energy (DOE), Office of Science, Office of Biological and Environmental Research through the Early Career Research Program and the Energy Exascale Earth System Model (E3SM) project. This review is PMEL contribution 5403.

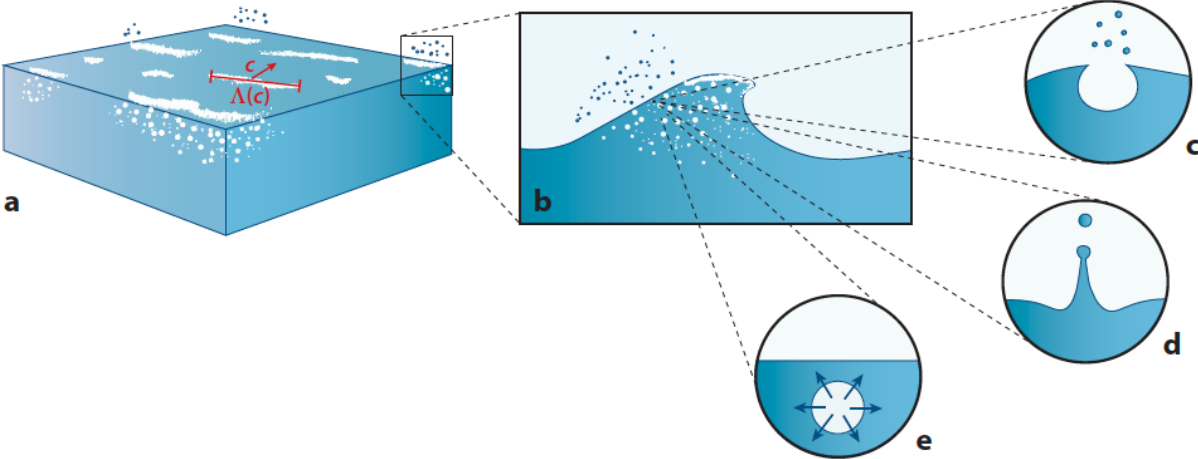
### **8. Data Availability**

Data shown in Figure 6 are available at <https://saga.pmel.noaa.gov/data/>.

**FIGURE CAPTIONS**

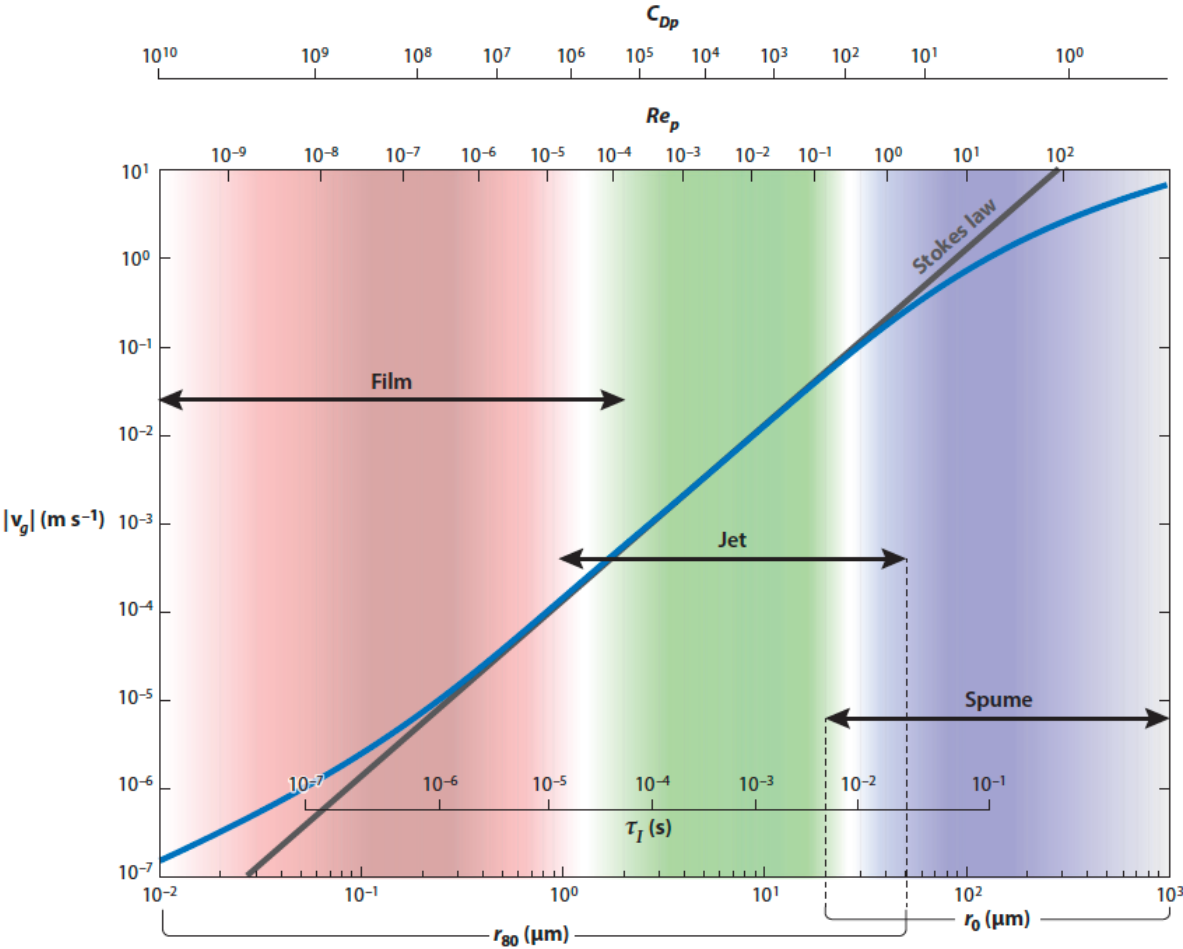
**Figure 1.** The multiscale approach discussed in this review to model mass exchange due to breaking waves, drops, and bubbles. At moderate to high wind speeds, breaking waves form whitecaps on the ocean surface. (a) The breaking statistics can be described by the length distribution  $\Lambda(c)$ . (c) of breaking crests moving at speed  $c$ , typically from 1 to 10 m s<sup>-1</sup>. (b) Each breaker’s dynamics is assumed to be self-similar and is described by its speed  $c$ , leading to scaling models for the associated energy dissipation, air entrainment, and bubble statistics. At the smallest scales, bubbles with sizes ranging from ~1  $\mu$ m to ~10 mm (c,d) burst at the surface to produce liquid sea sprays via (c) film and (d) jet drops, with sizes ranging from ~0.1  $\mu$ m to ~1 mm, and (e) exchange gas in the turbulent upper ocean. Reproduced from (Deike, 2022).

FIGURE 1

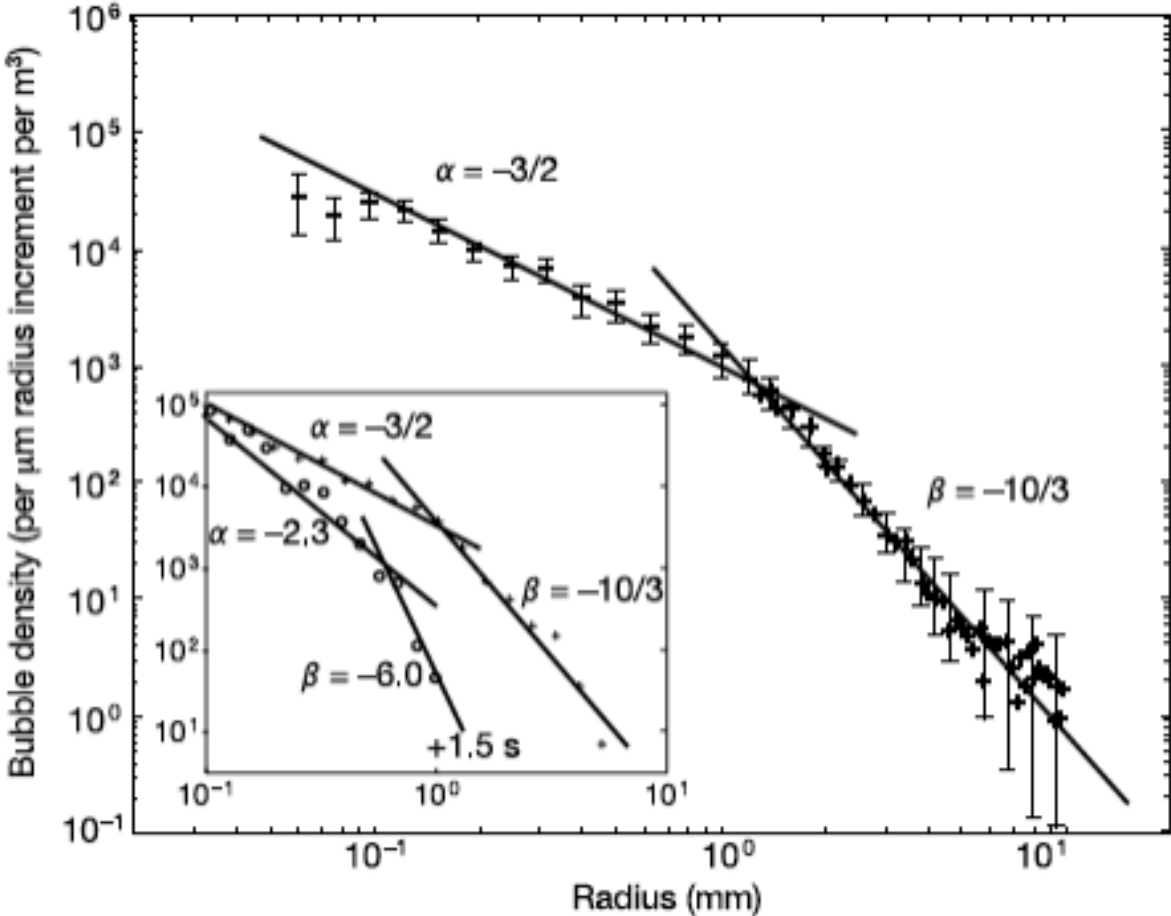




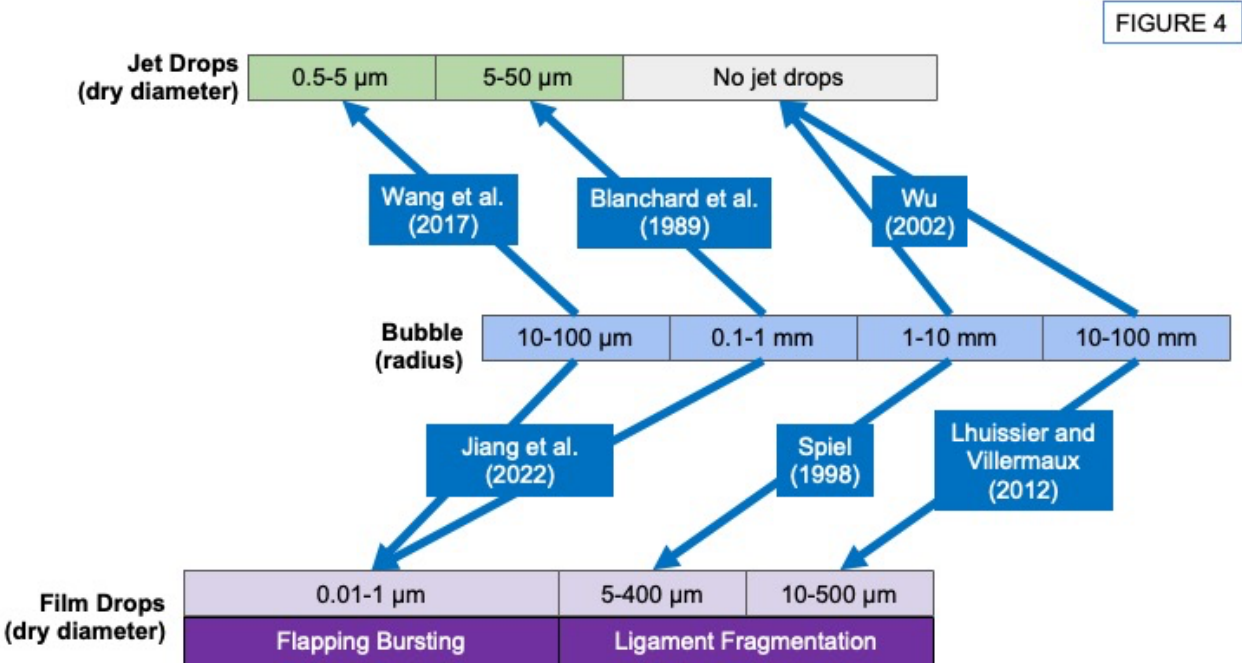
**Figure 2.** Terminal fall velocity of a water droplet in quiescent air at ambient temperature and pressure. The gray line shows the Stokes solution. Additional axes shown are the particle Reynolds number,  $Re_p$ , and drag coefficient,  $C_{Dp}$ , as well as the particle inertial relaxation time.  $R_0$  is the radius at formation and  $R_{80}$  is the radius at 80% relative humidity. Also shown is the size range for the film, jet, and spume drops. Reproduced from (Veron, 2015).



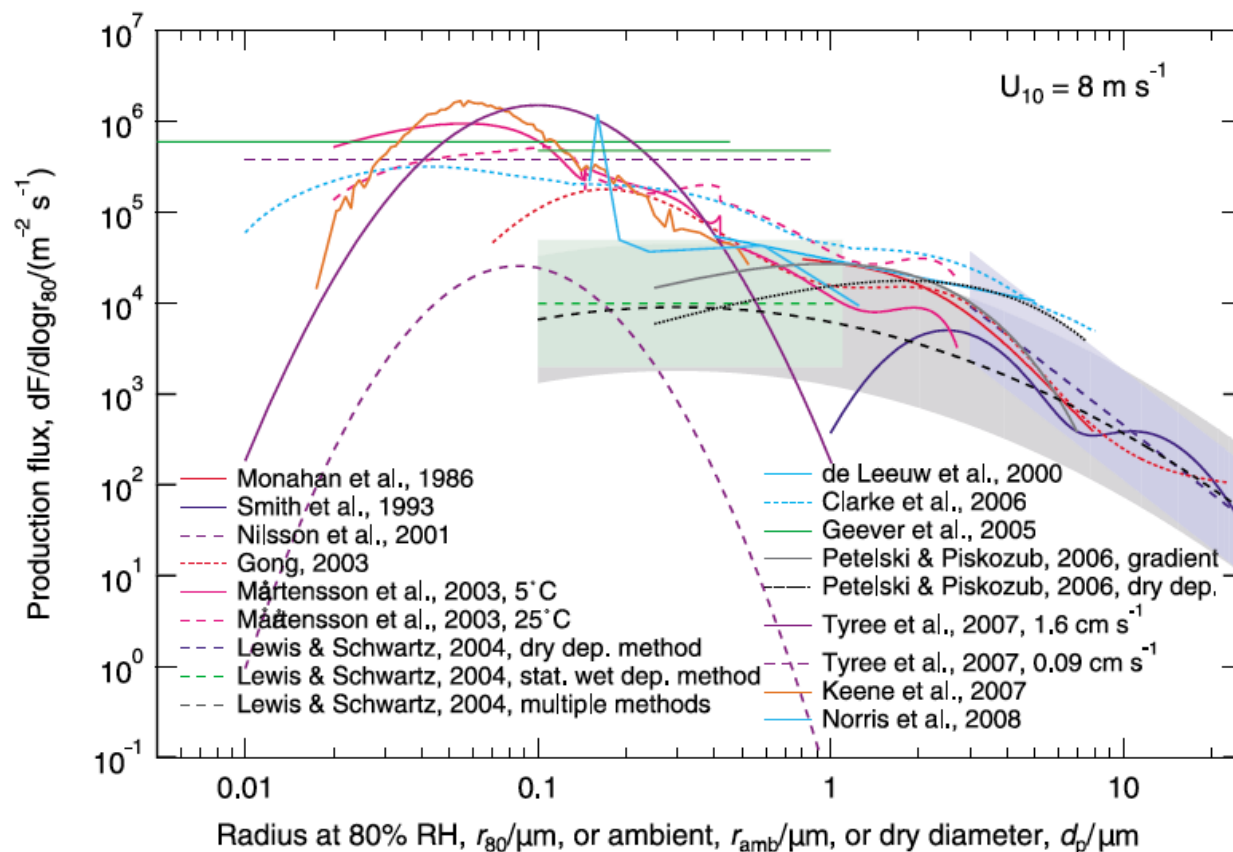
**Figure 3.** The average bubble size spectrum estimated from 14 breaking events during their acoustic phase. Two camera magnifications were used and the results superimposed to obtain the slightly greater than two decades of bubble radii observed. The vertical scale is number of bubbles per  $m^3$  in a bin radius  $1 \mu m$  wide. Vertical bars show  $\pm$  one standard deviation. The size distribution shows a marked change in slope at a radius that we are identifying as the Hinze scale. Bubbles larger and smaller than this scale respectively vary as  $(radius)^{-10/3}$  and  $(radius)^{-3/2}$  denoted by  $\beta$  and  $\alpha$ . Inset, the bubble size distribution at the beginning of the quiescent phase (crosses) and 1.5 s into the quiescent phase (open circles). Both slopes of the bubble spectrum have increased noticeably during this time interval. This rapid evolution becomes important when interpreting size distributions collected during the plume quiescent phase. Reproduced from (Deane & Stokes, 2002).



**Figure 4.** Summary of relationships identified between bubble radius, jet drop dry diameter, and film drop dry diameter, with selected references included (Blanchard, 1989; X. H. Jiang et al., 2022; Lhuissier & Villermaux, 2012; Spiel, 1998; Veron, 2015; Wang et al., 2017; Wu, 2002). Connecting lines are approximate representations of the size ranges from the references cited. For film drops, the different size regimes for flapping bursting and ligament fragmentation (centripetal) processes are noted (X. H. Jiang et al., 2022). Spume drop production is not included. The size ranges indicated are on different scales to show relevant ranges.



**Figure 5.** Parameterizations of size-dependent SSA production flux, evaluated for wind speed  $U_{10} = 8 \text{ m s}^{-1}$  (or  $U_{22} = 8 \text{ m s}^{-1}$  for Geever et al. (Geever et al., 2005)). Also shown are central values (curves) and associated uncertainty ranges (bands) from review of Lewis and Schwartz (Lewis & Schwartz, 2004), which denote subjective estimates by those investigators based on the statistical wet deposition method (green), the steady state deposition method (blue), and taking into account all available methods (gray); no estimate was provided for  $r_{80} < 0.1 \mu\text{m}$ . Lower axis denotes radius at 80% relative humidity,  $r_{80}$ , except for formulations of Nilsson et al. (Nilsson et al., 2001), Mårtensson et al. (Martensson et al., 2003), and Clarke et al. (Clarke et al., 2006), which are in terms of dry particle diameter,  $d_p$ , approximately equal to  $r_{80}$  and those of Geever et al. (Geever et al., 2005), Petelski and Piskozub (Petelski & Piskozub, 2006) (dry deposition method), and Norris et al. (Norris et al., 2008) which are in terms of ambient radius,  $r_{\text{amb}}$ . Formulation of Petelski and Piskozub (Petelski & Piskozub, 2006) by the dry deposition method. Formulations of Tyree et al. (Tyree et al., 2007) are for artificial seawater of salinity 33 at the two specified bubble volume fluxes. Formulations of Nilsson et al. (Nilsson et al., 2001) and Geever et al. (Geever et al., 2005) of particle number production flux without size resolution are plotted arbitrarily as if the flux is independent of  $r_{\text{amb}}$  over the size ranges indicated to yield the measured number flux as an integral over that range. Note that the different diameters indicated on the horizontal axis represent different measurement conditions that limit direct comparisons of some measurements. Reproduced from (de Leeuw et al., 2011).



**Figure 6.** Mass size distributions (a,b) collected from six open-ocean cruises that also measured *in situ* tracers to exclude continental influences, namely  $<1 \mu\text{m}$  absorption (at 530 nm from PSAP) less than  $0.8 \text{ Mm}^{-1}$  and radon concentration less than  $540 \text{ mBq m}^{-3}$  (Quinn et al., 2000). Panel b has the same information as panel a, but with the axes zoomed in to show the submicron concentrations. The six cruises with appropriate measurements were the First Aerosol Characterization Experiment (ACE1) in 1995, the International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) in 2008, the Western Atlantic Climate Study (WACS) in 2014, and the North Atlantic Aerosol and Marine Ecosystem Study (NAAMES) campaigns in Winter 2015 (NAAMES1), Late Spring 2016 (NAAMES2), and Autumn 2017 (NAAMES3). Organic contributions are excluded because they were not available. Sea salt is scaled from measured  $\text{Na}^+$  and  $\text{Cl}^-$ ; ammonium bisulfate is based on measured non sea salt sulfate ( $\text{SO}_4^{2-}$ ). Relative number size distributions (c) averaged over all six cruises in (a) are calculated using effective sea salt density of 2.0 (Saliba et al., 2020) and 1.78 for ammonium bisulfate. This sensitivity to the mean diameter is quantified here with three different estimates to bound the mean diameter: (1) an upper bound at the 50% cutoff diameter for the bin ( $D_{\text{high}}=D_{50}$ ), (2) a lower bound at approximately the 50% cutoff diameter of the next smaller bin ( $D_{\text{low}}=D_{50}-d\log D$ ), and (3) a best estimate at the geometric mean of these two values ( $D_{\text{mid}}=D_{50}-0.5d\log D$ ). The ratio of the standard deviation to the mean of the mass distributions Dry Diameter normalized to the wind speed (d) are also shown for the six cruises.

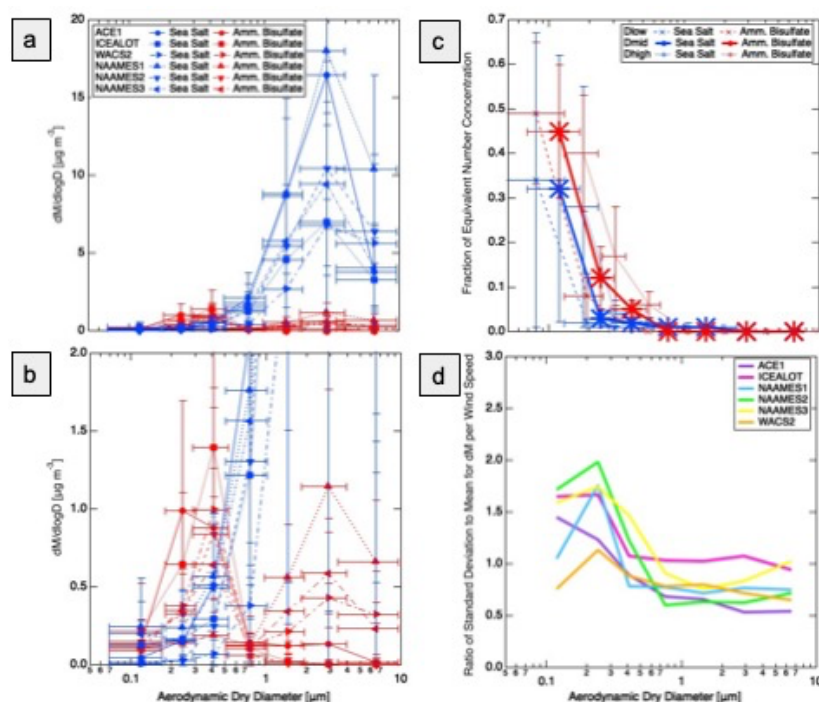
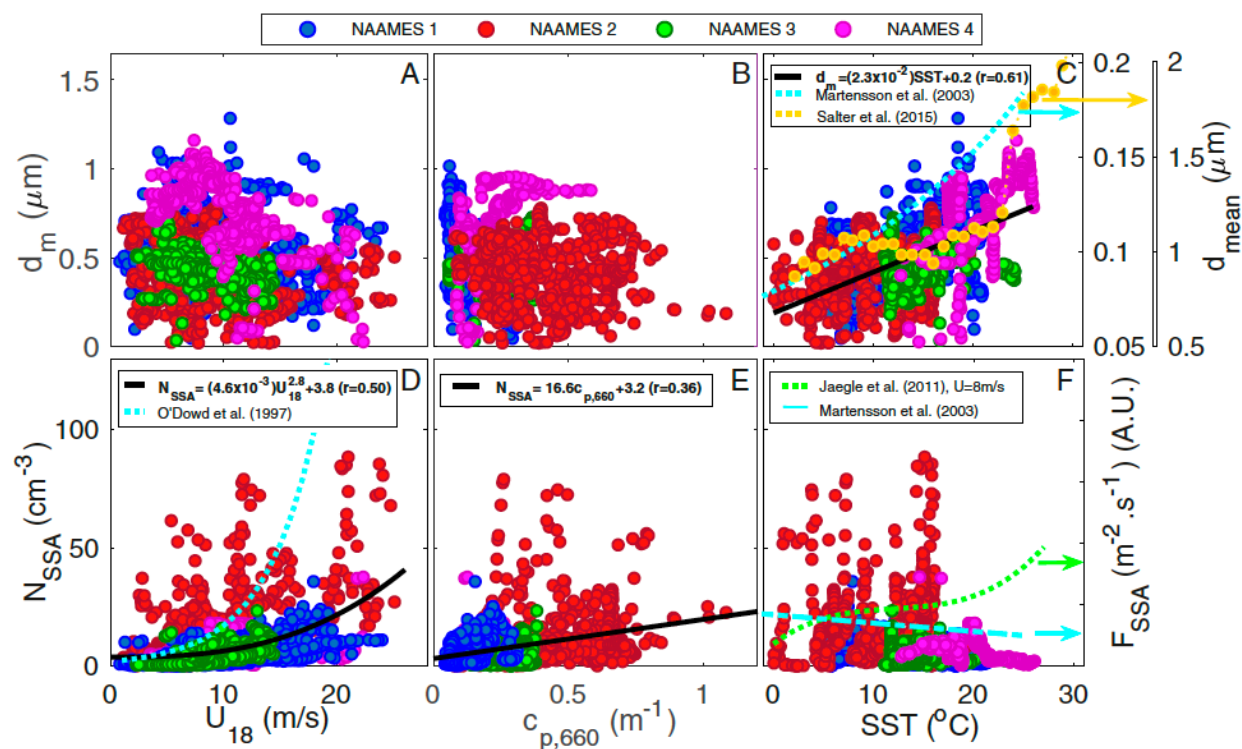
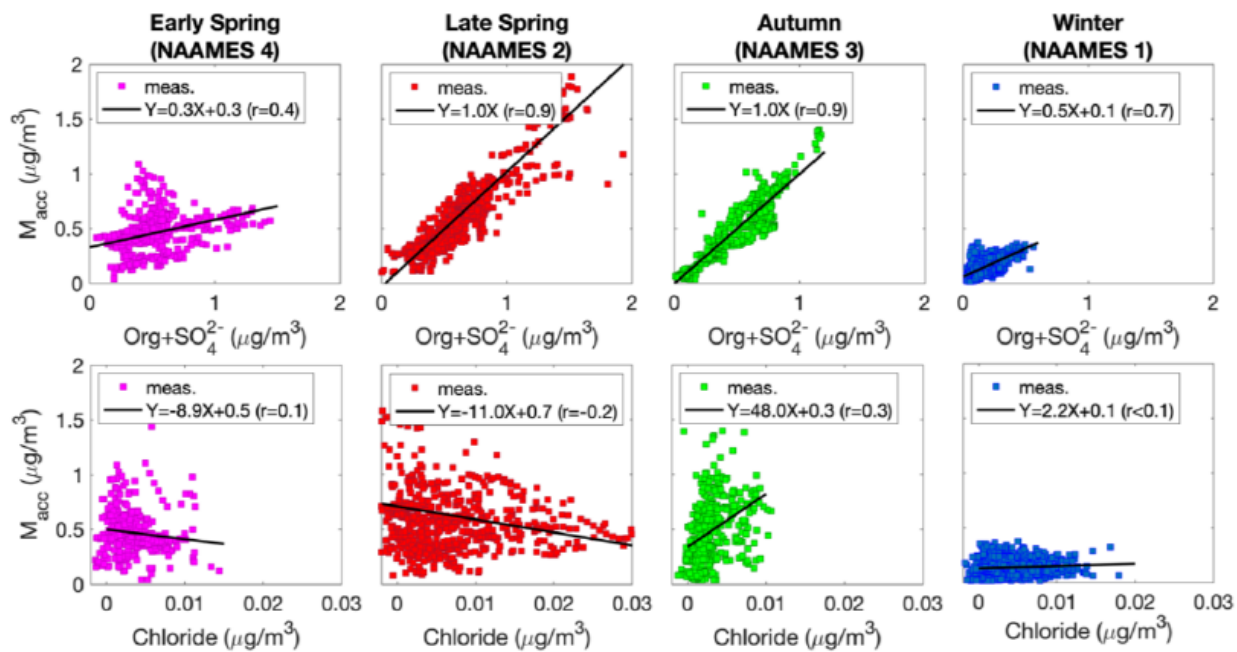


FIGURE 6

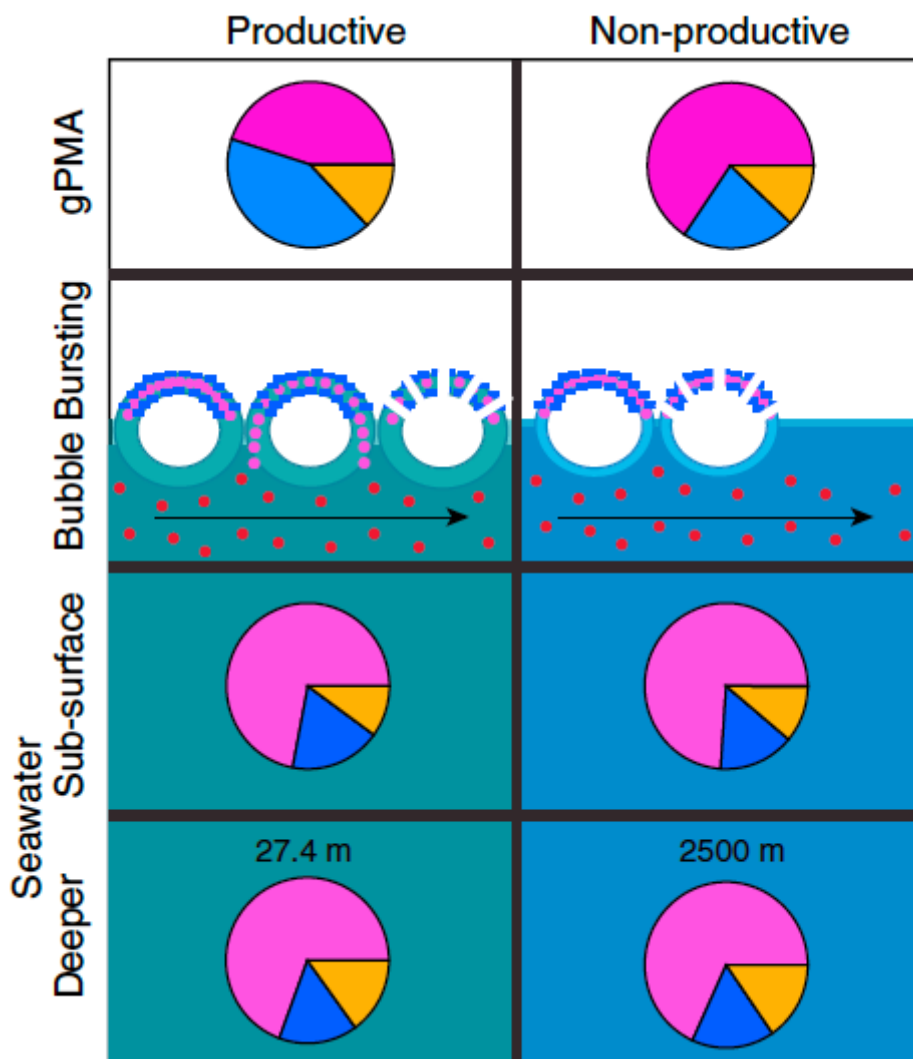
**Figure 7.** SSA mode diameter ( $d_m$ ) versus (A) wind speed ( $U_{18}$ ), (B) seawater particle attenuation at 660 nm ( $c_{p,660}$ ), (C) and SST. SSA number concentrations ( $N_{SSA}$ ) versus (D) wind speed, (E)  $c_{p,660}$ , and (F) SST. The measurements are colored by campaign for Winter 2015 (NAAMES1), Late Spring 2016 (NAAMES2), Autumn 2017 (NAAMES3), and Early Spring 2018 (NAAMES4). Lines of best fit for all four NAAMES campaigns are plotted as solid black lines if  $|R| > 0.3$ . Published parameterizations are plotted as dotted colored lines. The dotted blue curve in C is the mean diameter of the flux size distribution from Mårtensson et al. (Martensson et al., 2003) (first right y axis) and the dotted yellow curve in C is the mean diameter of the particle size distribution from Salter et al. (Salter et al., 2015) (second right y axis). The O’Dowd et al. (Odowd et al., 1997) SSA parameterization versus wind speed is shown in (D) as a dotted blue line. SSA flux production versus SST from Jaeglé et al. (Jaegle et al., 2011) at a wind speed of 8 m s<sup>-1</sup> (green dotted line right y axis) and SSA flux production from Mårtensson et al. (Martensson et al., 2003) (blue dotted line, right y axis) versus SST are shown in F. Parameterization for  $d_m$  versus SST is shown in bold in C. The correlation between  $d_m$  and SST ( $R = 0.61$ ) decreased when excluding NAAMES 4 measurements sampled at latitudes south of 30°N ( $R = 0.46$ ). Reproduced from (Georges Saliba et al., 2019).



**Figure 8.** (Top row) AMS-based organic + sulfate (Org + SO<sub>4</sub><sup>2-</sup>) submicron mass concentrations and accumulation mode mass concentration (M<sub>acc</sub>, calculated by integrating the marine number size distribution between 0.08 μm and 0.32 μm and assuming spherical particles and a density of 1.0 g/cm<sup>3</sup>) for NAAMES. (Bottom row) AMS submicron chloride mass concentrations versus M<sub>acc</sub>. Solid black lines are lines of best fit obtained. AMS-based concentrations are calculated assuming a collection efficiency of unity, so the numerical values the slopes shown are not quantitative. Reproduced from (Georges Saliba et al., 2019)

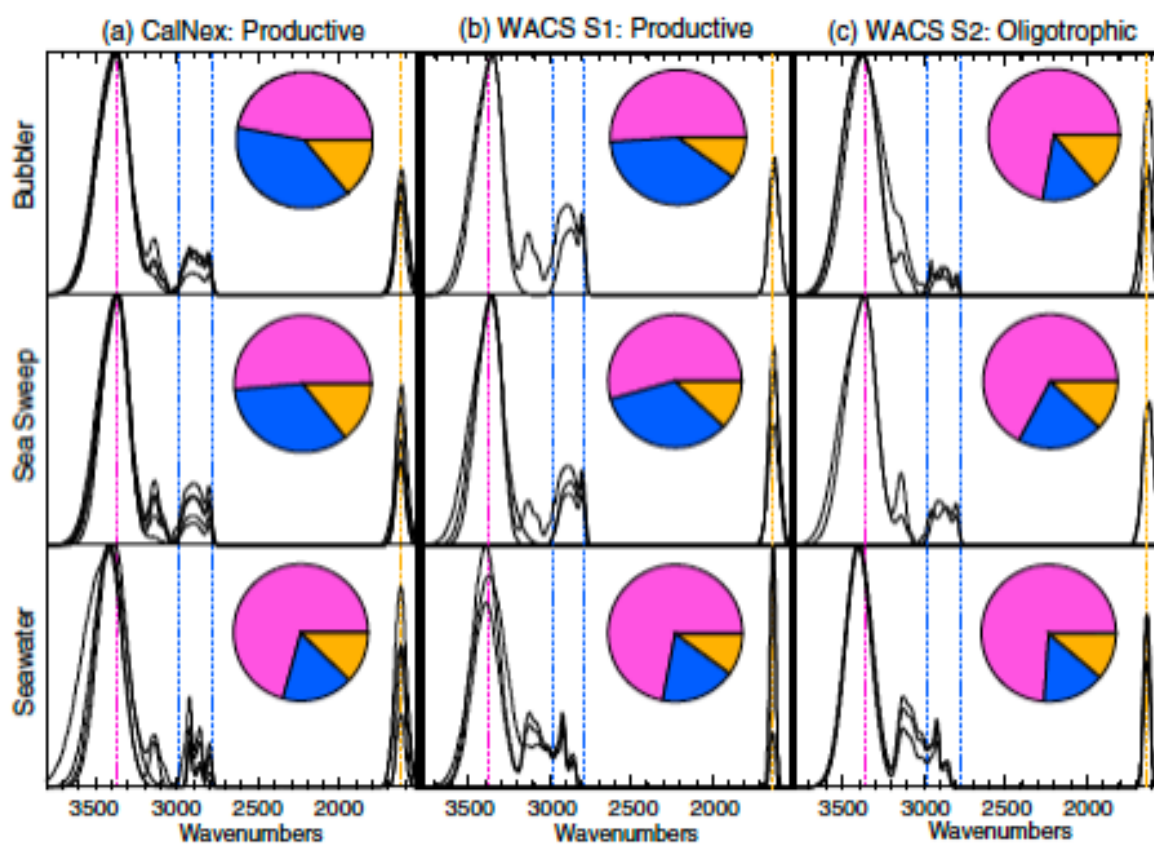


**Figure 9.** Average functional group composition of OM in (top) particles generated with Sea Sweep (a floating bubble generator) and Bubbler (a continuous-feed, on-board bubble generator), (middle) surface seawater, and (bottom) deep seawater at 27.4 m and 2500 m measured in (left column) productive and (right column) nonproductive seawater during WACS. The term “productive” is used to mean seawater with high chlorophyll relative to the campaign average. The colors in the pies represent the organic functional group fractions for hydroxyl (pink), alkane (blue), and amine (orange). The bubbles show the bubble draining process in both seawater types, with more surfactant in the productive seawater. The OM is shown as hydrophobic (blue squares), hydrophilic (pink circles), and polysaccharides (red circles). Reproduced from (Frossard, Russell, Burrows, et al., 2014).

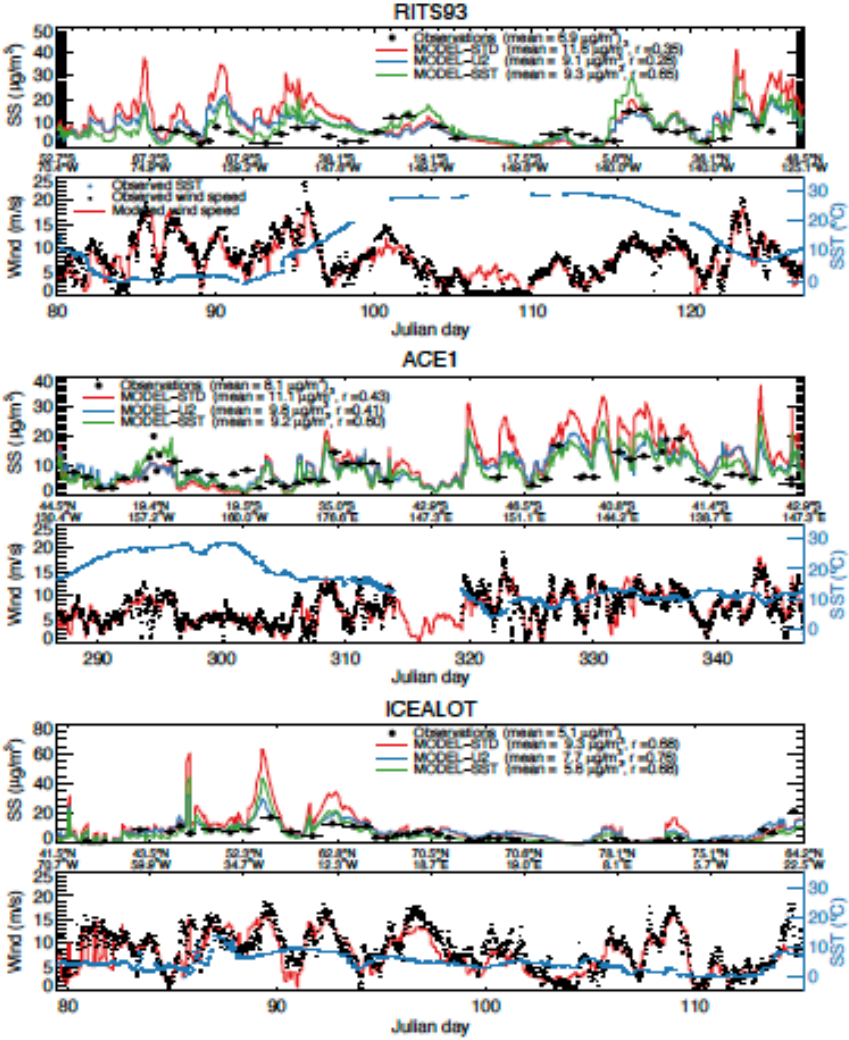




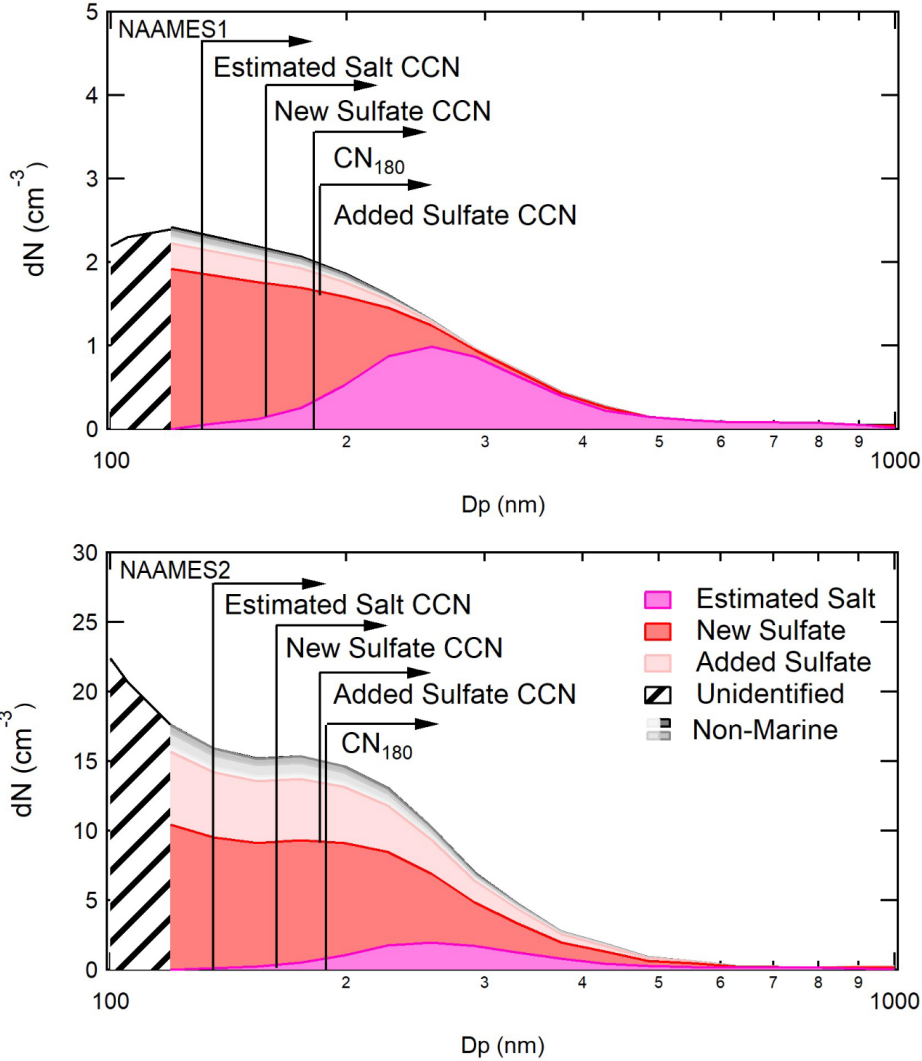
**Figure 10.** Comparison of the selected normalized organic FTIR spectra and average functional group composition measured at (a) the California Nexus study (CalNex), (b) WACS Station 1, and (c) WACS Station 2 in the gPMA generated with (top) the Bubbler, (middle) the Sea Sweep, and (bottom) the corresponding composition of OM in surface seawater. Pies represent the organic functional group composition as hydroxyl (pink), alkane (blue), and amine (orange). The dashed vertical lines indicate hydroxyl functional group peak absorption at  $3369\text{ cm}^{-1}$  (pink) and amine functional group peak absorption at  $1630\text{ cm}^{-1}$  (orange). The range of alkane functional group absorption from  $2980$  to  $2780\text{ cm}^{-1}$  (blue dashed lines) is also shown. The higher wave number peak absorption of the hydroxyl functional groups is evident in the seawater panel. The functional group compositions and spectra are from the subset of collocated samples. Reproduced from (Frossard, Russell, Burrows, et al., 2014).



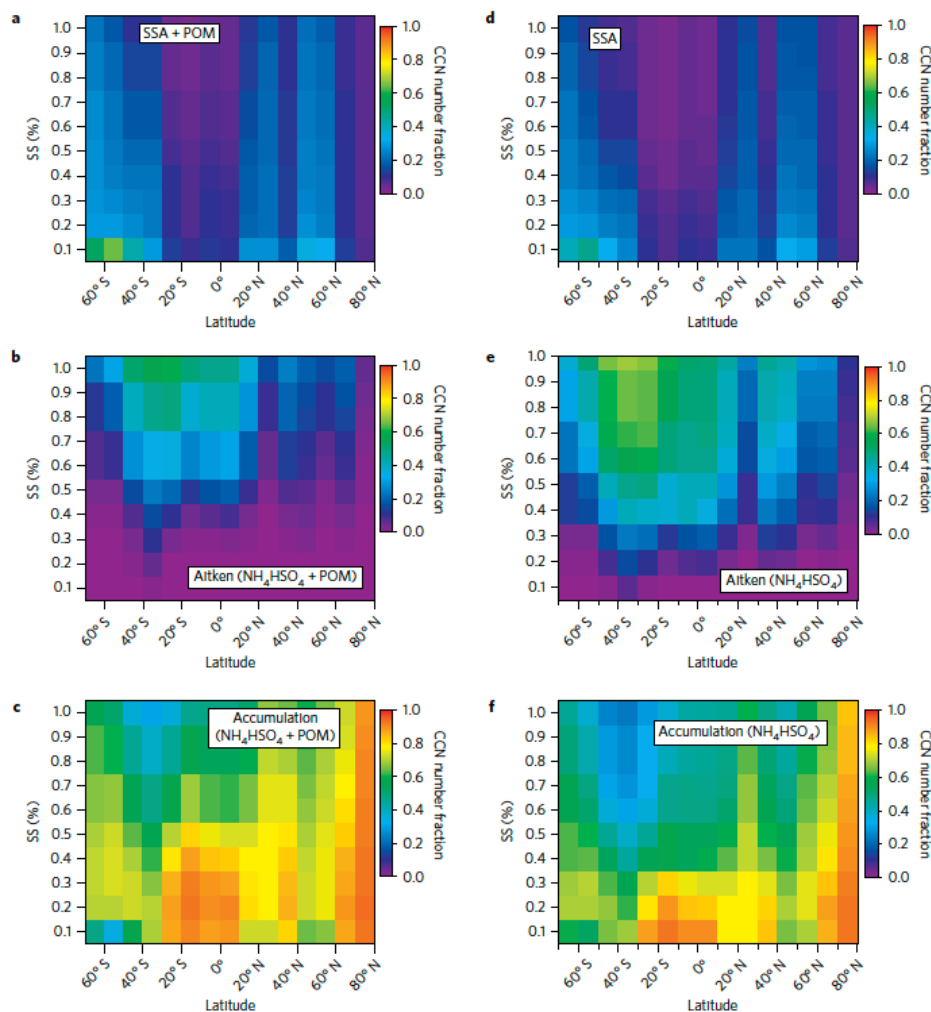
**Figure 11.** Time series of coarse mode sea spray mass concentration during the Radiatively-Important Trace Species 1993 (RITS-93), ACE1, and ICEALOT PMEL cruises. For each cruise, observations of sea salt concentrations are shown with black circles. The horizontal bar corresponds to the instrumental averaging period. The three lines are the three different models: standard model (MODEL-STD, red), model using Eq. (3) (MODEL-U2, blue), model using Eq. (4) (MODEL-SST, green). The bottom panel shows the timeseries of observed 10m wind speed (black dots) compared to the modeled windspeed (red line) as well as the observed SST (blue). Reproduced from (Jaegle et al., 2011).



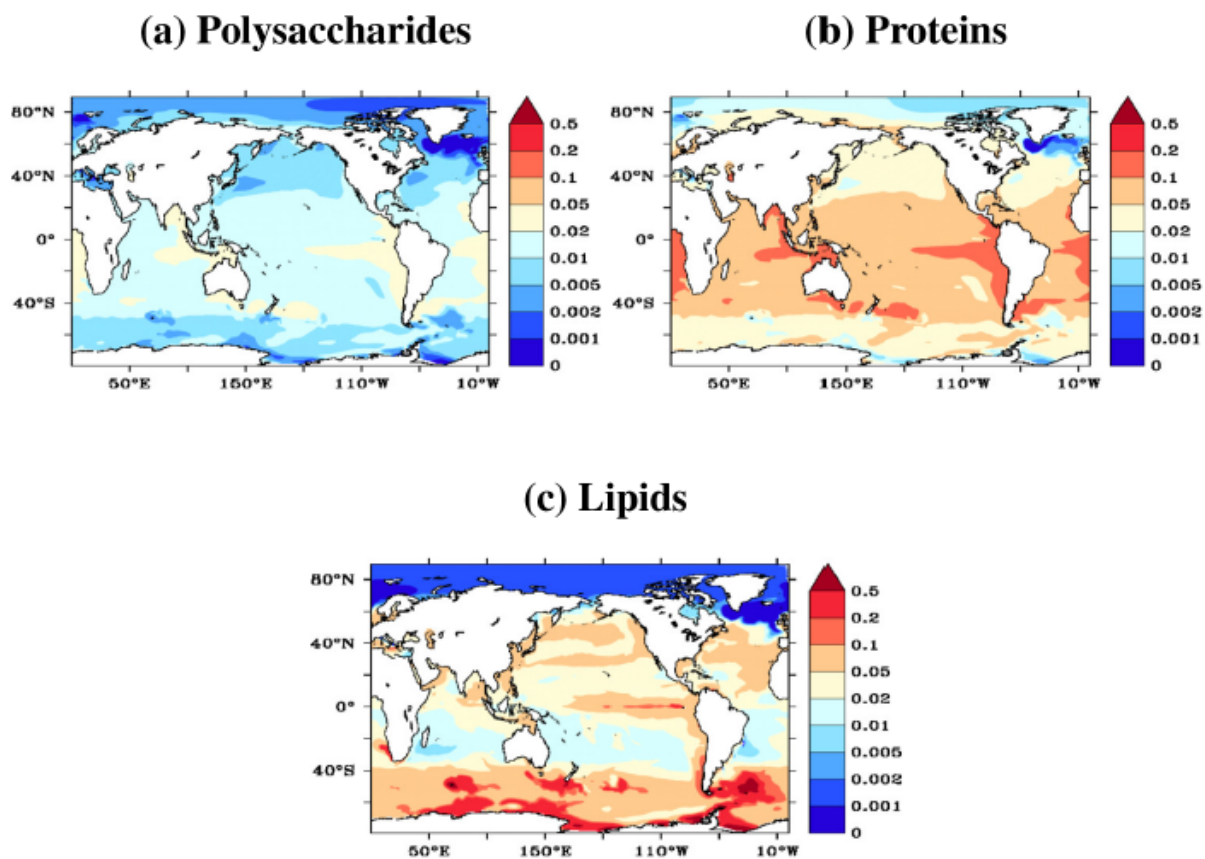
**Figure 12.** Particle type size distributions for examples from NAAMES1 and NAAMES2. Black arrows identify the 0.1% supersaturation activation diameters for the Estimated Salt (at 0.13  $\mu\text{m}$ ), New Sulfate, and Added Sulfate types. Reproduced from (Sanchez et al., 2018).



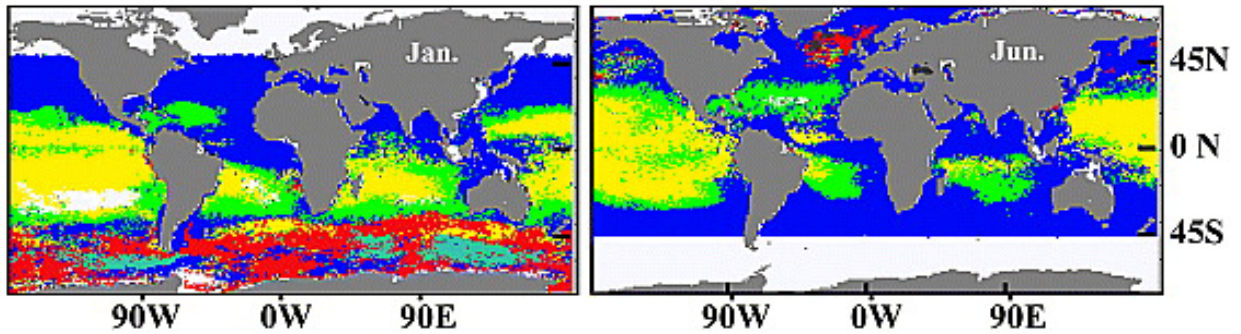
**Figure 13.** Calculated CCN modal number fraction as a function of supersaturation and latitude. a–f, Data are based on combined, latitudinally binned data from RITS-93, RITS-94, ACE-1 Legs 1 and 2, ICEALOT, the second Western Atlantic Climate Study (WACS-2), and NAAMES-1 for the SSA mode with a composition of sea salt and OM (a), Aitken mode for a composition of nss  $\text{SO}_4^{2-}$  (as  $\text{NH}_4\text{HSO}_4$ ) and OM (b), accumulation mode for a composition of nss  $\text{SO}_4^{2-}$  (as  $\text{NH}_4\text{HSO}_4$ ) and OM (c), SSA mode as pure SSA (d), Aitken mode as pure nss  $\text{SO}_4^{2-}$  (as  $\text{NH}_4\text{HSO}_4$ ) (e) and accumulation mode as pure nss  $\text{SO}_4^{2-}$  (as  $\text{NH}_4\text{HSO}_4$ ) (f). Reproduced from (P. K. Quinn et al., 2017).



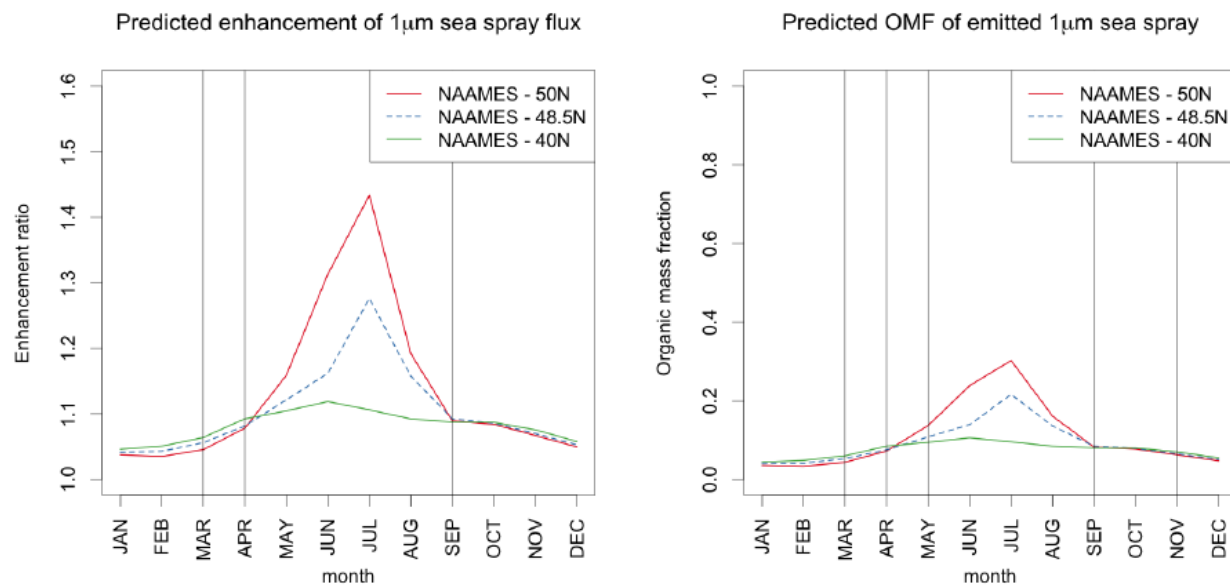
**Figure 14.** Submicron SSA dry mass fraction from each compound class for February from the Organic Compounds from Ecosystems to Aerosols: Natural Films and Interfaces via Langmuir Molecular Surfactants (OCEANFILMS) model BASE case. Fractions of processed and humic classes (not shown) are negligible. Reproduced from (Burrows et al., 2014).



**Figure 15.** Monthly climatology (January and June, 1998–2006) of the dominant phytoplankton group as retrieved by the spectral-based method for identifying phytoplankton from anomalies in satellite retrievals of ocean color known as PHYSAT. The colors indicate diatoms in red, nanoeucaryotes in blue, *Synechococcus* in yellow, *Prochlorococcus* in green, and phaeocystis-like in light blue. Reproduced from (Alvain et al., 2008).



**Figure 16.** OCEANFILMS-predicted seasonal cycle of biological enhancement in the emission flux (a; ratio of predicted marine organic aerosol plus sea salt flux to flux of pure sea salt) and the organic mass fraction (b) of sea spray particles with diameter  $<1\ \mu\text{m}$ , for the region of the NAAMES field campaign. The NAAMES campaign consisted of four cruises in different seasons, with most scientific sampling occurring close to  $40^\circ\text{W}$  and between approximately  $40^\circ\text{N}$  and  $50^\circ\text{N}$  (Behrenfeld et al., 2019). All model results are monthly mean output at  $40^\circ\text{W}$  and at the latitudes indicated in the legend. Vertical lines indicate the months during which each variable was measured by NAAMES. Reproduced from (Burrows, Easter, et al., 2022).



## TABLES

**Table 1.** Publications relevant to SSA production. Modified from Saliba et al. (2019).

| Reference   | Type                 | Findings   |
|---|----------------------|--|
| (Grythe et al., 2014)                               | Review               | Positive dependence of SSA production on SST   |
| (Lewis & Schwartz, 2004)                            | Review               | SSA flux dependence on wind speed based on multiple laboratory and field studies   |
| (Meskhidze et al., 2013)                            | Review               | CCN fluxes and optical properties are still poorly constrained   |
| (Forestieri & Moore, 2018; Forestieri et al., 2018) | Laboratory           | Positive dependence of SSA concentration on SST using artificial seawater, but no correlation when using filtered but not autoclaved seawater                      |
| (Fuentes et al., 2010)                              | Laboratory           | Complex response of SSA number and mean diameter to dissolved organic amount for water collected off the West African coast  |
| (Martensson et al., 2003)                           | Laboratory           | Complex dependence of SSA concentrations on SST using artificial seawater  |
| (Modini et al., 2013)                               | Laboratory           | Decreased SSA production efficiency and increased mean diameter of largest SSA mode (~0.2 $\mu$ m) with decreased surface tension                                  |
| (K. Sellegri, Colin D. O'Dowd, et al., 2006)        | Laboratory           | Enhancement of largest SSA mode (0.3 $\mu$ m - 0.5 $\mu$ m) with increased surfactants and wind stress (1m/s)  |
| (J. Zabori et al., 2012)                            | Laboratory           | SSA flux dependence on wind speed based on multiple laboratory and field studies   |
| Salter et al. (2015)                                | Laboratory and model | Increased SSA mean diameter with increased water temperature for laboratory experiments using artificial sea water and a temperature-controlled chamber            |
| (Jaegle et al., 2011)                               | Model                | Positive dependence of SSA production on SST using field measurements and satellite observations   |
| (Burrows et al., 2016)                              | Model                | Represented organic emissions in sea-spray using the OCEANFILMS partitioning model   |
| (Kasparian et al., 2017)                            | Observational        | Aerosol number concentrations in the North Atlantic for 1.0 - 2.5 $\mu$ m were consistent with Jaeglé et al. (2011)  |
| (Lehahn et al., 2014)                               | Observational        | Positive correlation with wind speed and negative correlation with SST and chlorophyll for SSA number concentrations with diameter larger than 0.5 $\mu$ m         |
| (Middlebrook et al., 1998)                          | Observational        | Most particles larger than 0.16 $\mu$ m contained organic compounds near Cape Grim in the Southern Ocean   |
| (R. L. Modini et al., 2015)                         | Observational        | Fitted the tail of the marine size distribution to a lognormal function and argued that this mode was the salt mode using measurements from the Eastern Pacific    |
| (Murphy et al., 1998)                               | Observational        | Large fraction of particles larger than 0.16 $\mu$ m sampled at Cape Grim in Southern Ocean contained sea salt   |
| (Odowd et al., 1997)                                | Observational        | Positive correlation of SSA concentrations on wind speed for the Northeast Atlantic  |
| (Colin D. O'Dowd et al., 2004)                      | Observational        | Organics components contributed a substantial part of the submicron aerosol mass, especially during plankton blooms  |
| (Ovadnevaite et al., 2012)                          | Observational        | Gong (2003), Mårtensson et al. (2003), and Fuentes et al. (2010) SSA parameterizations with wind speed were higher than measured NaCl mass in the Eastern Atlantic |



|                                   |                        |  |
|-----------------------------------|------------------------|--|
| <b>(Quinn et al., 2000)</b>       | Observational          | SSA concentrations and size depend on location, with submicron particles dominating SSA in the Northeastern Atlantic and Southern Ocean.   |
| <b>(P. K. Quinn et al., 2017)</b> | Observational          | Sea salt number concentrations contributed up to 30% of CCN in clean marine conditions and correlated positively with wind speed, using the Modini et al. (2015) method                                  |
| <b>(Russell et al., 2010)</b>     | Observational          | Organic compounds (similar to saccharides) contribute less than 50% of submicron particle mass and correlated positively with wind speed and Na <sup>+</sup> mass  |
| <b>(Gong, 2003)</b>               | Observational + model  | Modified the Monahan et al. (1986) parameterization of sea spray on wind speed to best fit concentrations from O'Dowd et al. (1997)  |
| <b>(Dror et al., 2018)</b>        | Satellite observations | Positive correlation between wind speed and AOD on daily and yearly timescales but not on seasonal timescales and negative correlation between AOD and chlorophyll concentrations on seasonal timescales |

**Table 2.** Literature reported values of sea spray modal parameters. Number mean diameters ( $D_{g,number}$ ) were converted to mass mean diameters ( $D_{g,mass}$ ) using Eq. (7), integrating over particle sizes 0.01 – 10  $\mu\text{m}$ , and averaging over a total particle concentration range of 1 – 100  $\text{cm}^{-3}$ . Values are averages unless noted as an upper or lower bound. Modified from Dedrick et al. (2022).

| Reference  | Experiment type   | Ocean basin                                   | Parameter                                  |  |  |
|--|---|---|--|--|--|
|  |   |   | $D_{g,number}$ ( $\mu\text{m}$ )           | $D_{g,mass}$ ( $\mu\text{m}$ )               | $\sigma_g$                                 |
| (Lewis & Schwartz, 2004),<br>(K Sellegri et al., 2006),  | field measurements<br>(RH: 80%)                                 |   | 0.3  | 1.3  | 3  |
| (W. Keene et al., 2007),<br>(Fuentes et al., 2010),<br>(Modini et al., 2010),<br>(T. Bates et al., 2012),<br>(J Zabori et al., 2012) | laboratory-based<br>bubble bursting<br>(RH: ambient)            |   | 0.05 (lower bound)<br>0.1 (upper bound)    | 0.25 (lower bound)<br>0.48 (upper bound)     | 2.8  |
| (K. Prather et al., 2013)  | laboratory-based<br>breaking wave flume<br>(RH: $10 \pm 15\%$ ) | N.E. Pacific                                  | 0.16                                       | 0.88   | 3  |
| (R. Modini et al., 2015)   | field measurements<br>(RH: < 40%)                               | N.E. Pacific                                  | 0.14 (lower bound)<br>0.26 (upper bound)   | 0.5 (lower bound)<br>1.3 (upper bound)       | 2.5 (lower bound)<br>3 (upper bound)       |
| (P. Quinn et al., 2017)  | field measurements<br>(RH: variable, mostly < 50%)              | Pacific,<br>Southern, Arctic,<br>and Atlantic | 0.3  | 1.08   | 2.5  |
| (G Saliba et al., 2019)  | field measurements<br>(RH: < 40%)                               | N. Atlantic                                   | 0.5  | 1.6  | 2.3  |
| (K. Sanchez et al., 2021)  | field measurements<br>(RH: ambient)                             | Southern Ocean                                | 0.6  | 0.71   | 1.4  |
| (Dedrick et al., 2022)   | field measurements<br>(RH = $55 \pm 10\%$ )                     | S. Atlantic                                   | 0.4<br>(UHSAS-only)<br>0.5<br>(UHSAS-NEPH) | 0.68<br>(UHSAS-only)<br>1.47<br>(UHSAS-NEPH) | 1.8<br>(UHSAS-only)<br>2.4<br>(UHSAS-NEPH) |

**Table 3.** Selected references investigating the organic composition and concentration of SSA, generated SSA, SMA, and general Marine Aerosol measured in different ocean regions (from Frossard et al., 2014).

| Reference  | Ocean Region <sup>a</sup>            | OM Composition   | Particle Size (OM, $\mu\text{g m}^{-3}$ ) |
|--|--------------------------------------|--|---|
| <b>SSA</b>   |                                      |  |   |
| (P. Q. Fu et al., 2013)  | Arctic                               | Saccharides  | Bulk                                      |
| (Leck et al., 2013)  | Arctic                               | Heteropolysaccharides                                    | 0.035 – 10 $\mu\text{m}$                  |
| (Russell et al., 2010)   | Arctic and N Atlantic                | Polysaccharides  | Submicron                                 |
| (Hawkins & Russell, 2010)  | Arctic and SE Pacific                | Polysaccharides, proteins, and phytoplankton fragments   | Submicron and Supermicron                 |
| (C. D. O'Dowd et al., 2004)  | NE Atlantic (Ireland)                | Enriched in WIOC with high molecular weight              | Submicron                                 |
| (Ceburnis et al., 2008b)   | NE Atlantic (Ireland)                | WIOC   | Submicron                                 |
| (M. C. Facchini et al., 2008)a   | NE Atlantic (Ireland)                | WIOM   | Submicron                                 |
| (Bigg & Leck, 2008)  | NE Atlantic (Ireland)                | Exopolymers  | Submicron, < 200 nm                       |
| (Ovadnevaite et al., 2011)   | NE Atlantic (Ireland)                | Hydrocarbon  | Submicron                                 |
| (Mochida et al., 2002)   | N Pacific                            | LMW saturated fatty acids                                | Bulk                                      |
| (Sciare et al., 2009)  | Austral Ocean                        | WIOC   | Bulk                                      |
| <b>Generated SSA</b>   |                                      |  |   |
| (Gao et al., 2012)   | Arctic                               | Polysaccharides  | Bulk                                      |
| (M. C. Facchini et al., 2008)a   | NE Atlantic (Ireland)                | WIOM: colloids   | Submicron                                 |
| (Facchini, Rinaldi, et al., 2010)  | NE Atlantic (Ireland)                | WIOM: lipo-polysaccharides                               | Submicron                                 |
| (W. C. Keene et al., 2007)   | NW Atlantic (Sargasso Sea)           | WSOC   | Submicron and Supermicron                 |
| (Schmitt-Kopplin, Liger-Belair, Koch, Flerus, Kattner, Harir, Kanawati, Lucio, Tziotis, Hertkorn, & Gebefuegi, 2012) | SE Atlantic                          | Biomolecules with high aliphaticity                      | < 10 $\mu\text{m}$                        |
| (T. S. Bates, P. K. Quinn, A. A. Frossard, L. M. Russell, J. Hakala, T. Petäjä, et al., 2012)                        | NE Pacific (Coastal)                 | Polysaccharide-like, Alkyl-like, pattern of CH-fragments | Submicron                                 |
| (Ault et al., 2013a)   | NE Pacific (Coastal)                 | Aliphatic hydrocarbons                                   | 0.15-10 $\mu\text{m}$                     |
| (Quinn et al., 2014)   | NE Pacific (Coastal) and NW Atlantic | Saccharide-like  | Submicron                                 |
| <b>SMA</b>   |                                      |  |   |
| (P. Q. Fu et al., 2013)  | Arctic                               | Isoprene product   | Bulk                                      |

|  |                            |   |   |
|--|----------------------------|---|---|
| (C. D. O'Dowd et al., 2004)  | NE Atlantic (Ireland)      | Enriched in WSOC (partly oxidized species with extended aliphatic moieties)                           | Submicron   |
| (Ceburnis et al., 2008b)   | NE Atlantic (Ireland)      | WSOC  | Submicron   |
| (M. C. Facchini et al., 2008)a   | NE Atlantic (Ireland)      | WSOM  | Submicron   |
|  | NE Atlantic                | WSOC – dimethyl and diethyl ammonium salts  | Submicron   |
| (Facchini, Rinaldi, et al., 2010)  | NE Atlantic (Ireland)      | Diethyl and dimethyl amine salts  | Submicron   |
| (Matteo Rinaldi et al., 2010)  | NE Atlantic (Ireland)      | WSOC: MSA, alkylammonium salts, dicarboxylic acids  | Submicron   |
| (Meskhidze & Nenes, 2006)  | Southern Ocean             | Isoprene product  | Bulk  |
| (Turekian et al., 2003)  | NW Atlantic (Sargasso Sea) | Oxalate   | Submicron and Supermicron   |
| <b>Marine Aerosol<sup>b</sup></b>  |                            |   |   |
|  |                            |   | Submicron OC (0.66); Supermicron OC (0.26)                                      |
| (Cavalli et al., 2004)   | NE Atlantic (Ireland)      | WIOC; aliphatic and partially oxidized humic-like substances  | Submicron OC (0.25)   |
| (Cavalli et al., 2004)   | NE Atlantic (Ireland)      | WSOC  | Total OC (0.07, LBA; 0.62, HBA)   |
| (C. D. O'Dowd et al., 2004)  | NE Atlantic (Ireland)      | WIOC and WSOC   | Total OC (1.2, spring; 0.1, winter)<br>Submicron OC (0.2, spring; 0.05, winter) |
| (Yoon et al., 2007)  | NE Atlantic (Ireland)      |   | Submicron (3.8)   |
| (Ovadnevaite et al., 2011)   | NE Atlantic (Ireland)      |   |   |
| (Decesari et al., 2011)  | NE Atlantic                | WIOC similar to lipids; WSOC containing fatty acids, alkanolic acids, aliphatic acids, sulfate esters | Submicron   |
| (Schmitt-Kopplin, Liger-Belair, Koch, Flerus, Kattner, Harir, Kanawati, Lucio, Tziotis, Hertkorn, & Gebefuegi, 2012) | SE Atlantic                | Biomolecules with high aliphaticity   | < 10 µm   |
| (Crahan et al., 2004)  | Tropical Mid-Pacific       | Dicarboxylic acids, carbohydrates   | < 3.5 µm  |
| (Kawamura & Gagosian, 1987)  | N Pacific                  | Oxo-, mono-, and di-carboxylic acids  | Bulk  |
| (Matsumoto & Uematsu, 2005)  | N Pacific                  | Free amino acids in WSOC  | < 2.5 µm  |
| (Bigg, 2007)   | SW Pacific (Tasmania)      | WIOC aggregates; exopolymeric gels  | Submicron, < 200 nm   |

|                           |  |  |                  |
|---------------------------|--|--|------------------|
| (Shank et al., 2012)      | SE Pacific                                 | N/A  | Submicron (0.01) |
| (Kuznetsova et al., 2005) | NW Mediterranean                           | Proteins, amino acids, and polysaccharides in gels | Bulk             |
| (Fu et al., 2011)         | N Pacific, N Atlantic, Indian, South China | LMW fatty acids, fatty alcohols, and sterols       | Bulk             |

<sup>a</sup>The specific stationary sampling locations are in parentheses.

<sup>b</sup>The OM in the studies in this category were not identified as SSA or SMA and are thus included as general atmospheric marine aerosol particles (aMA).

**Table 4.** Selected references on DMS as an SMA source (from Sanchez et al., 2018).

| Relevant Findings   | Ref.   | Location  | Observations or Model  |
|---|--|---|--|
| <p><b>Relationship between DMS and CCN</b><br/>DMS-derived sulfate aerosol account for most of the CCN in the remote marine boundary layer.</p> <p>MSA and CCN vary seasonally and have a non-linear relationship.<br/>DMS and CCN in boundary layer are strongly (non-linearly) correlated.<br/>CCN and DMS are correlated but relationship can be nonlinear because of SO<sub>2</sub> sinks.<br/>CN correlates strongly with atmospheric DMS and DMS flux but weakly with CCN.<br/>Modeled CN and CCN correlate with DMS flux; free tropospheric entrainment affects CN and CCN concentration in the marine boundary layer.</p>   | <p>(R. J. Charlson et al., 1987)</p> <p>(Ayers &amp; Gras, 1991)</p> <p>(Hegg et al., 1991)</p> <p>(Russell et al., 1994)</p> <p>(Andreae et al., 1995)</p> <p>(Raes, 1995)</p>    | <p>Global</p> <p>Cape Grim<br/>NE Pacific<br/>N/A</p> <p>S. Atlantic<br/>N/A</p>                                    | <p>Model plus Observations</p> <p>Observations</p> <p>Observations</p> <p>Model</p> <p>Observations</p> <p>Model</p> |
| <p><b>New Particle Formation from DMS Products</b><br/>The number of particles formed by homogeneous nucleation depends on the preexisting aerosol concentration.<br/>Particle number concentration increases rapidly after a decrease in particle surface area and increase in SO<sub>2</sub> concentration.<br/>After precipitation, marine boundary layer aerosol particles can be replenished from new particles formed by nucleation if DMS concentrations are high.</p>   | <p>(Warren &amp; Seinfeld, 1985)</p> <p>(Covert et al., 1992)</p> <p>(Pirjola et al., 2000)</p>  | <p>N/A</p> <p>NE Pacific</p> <p>N/A</p>   | <p>Model</p> <p>Observations</p> <p>Model</p>  |
| <p><b>Evidence of New Particle Formation in the Free Troposphere</b><br/>Vertical profiles of Aitken mode aerosol concentrations showed maximum values just above cloud tops.<br/>Aerosol nucleation is observed above cloud top and downwind of cloud outflows.<br/>CN and CCN were replenished on time scales of 2-4 days with transported nuclei from the free troposphere after precipitation scavenging.<br/>Variability in marine boundary layer aerosol concentration is closely linked to changes in vertical transport.<br/>Nucleation is observed in the free troposphere but not the marine boundary layer, and it is observed more frequently for particle surface area less than 5-10 μm<sup>2</sup> cm<sup>-3</sup>.<br/>CN concentration in the marine boundary layer is controlled by the rate of entrainment from the free troposphere in most conditions.</p> | <p>(Hegg et al., 1990)</p> <p>(Perry &amp; Hobbs, 1994)</p> <p>(Clarke et al., 1996)</p> <p>(Raes et al., 1997)</p> <p>(Clarke et al., 1998)</p> <p>(Katoshevski et al., 1999)</p> | <p>NW and NE Pacific<br/>N. Pacific</p> <p>Christmas Island</p> <p>NE Atlantic</p> <p>Southern Ocean</p> <p>N/A</p> | <p>Observations</p> <p>Observations</p> <p>Observations</p> <p>Observations</p> <p>Observations</p> <p>Model</p>     |

|  |                          |                  |                    |
|--|--------------------------|------------------|--------------------|
| Observed growth rates of new particles in the free troposphere cannot be explained by SO <sub>2</sub> products and water vapor so other components must contribute to condensation.  | (Reus et al., 2000)      | NE Atlantic      | Model/Observations |
| New sulfate particles do not form in the marine boundary layer but instead in the free troposphere and then are entrained downward.  | (Kazil et al., 2006)     | Global           | Model              |
| Entrainment of nucleated sulfate particles from the free troposphere account for 43-65% of CCN, but only 7-20% in the winter; long range transport of marine CCN results in a time lag between CCN and DMS concentrations. | (Korhonen et al., 2008)  | Southern Ocean   | Model              |
| 45% of marine boundary layer CCN (at 0.2%) are from nucleation that occurred in the free troposphere.  | (Merikanto et al., 2009) | Global           | Model              |
| Sulfate particles from DMS mixed up to the free troposphere are a source of marine boundary layer CCN.   | (Clarke et al., 2013)    | Tropical Pacific | Observations       |

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