Sea Spray Emissions

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Contents

Abstract Sea spray aerosol particles constitute one of the most important natural components of the atmosphere and are emitted from the Mediterranean Sea with characteristics linked to the high temperatures and oligotrophic properties of the seawater. Sea spray aerosol production results from the action of wind stress on the sea surface, creating waves and bubbles, and is traditionally assumed to be mainly composed of inorganic sea salt, an organic fraction, and water. While the inorganic matter present in sea spray is relatively well documented and parameterized in models, much more uncertainties are associated with the emission of marine organic matter. Biochemical processes in the seawater and surface microlayer are at play in determining the fraction of organic matter in sea spray, but also the number of sea

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spray particles emitted into the atmosphere. Under environmental conditions specific to the Mediterranean Sea, the relationship between seawater biology and sea spray properties has specificities that will be presented in the present chapter.

1 Introduction

Marine aerosol particles constitute one of the most important natural components of the atmosphere that significantly contribute to the Earth's radiative budget, but they also have local effects on regional air quality (O'Dowd & De Leeuw, 2007). Marine aerosols influence cooling or warming at the top of the atmosphere (Randles et al., 2004, Claeys et al., 2017; Mallet et al., 2022). These aerosols may also have indirect effects by becoming cloud droplets or ice crystals (Novakov & Corrigan, 1996; Novakov & Penner, 1993). Despite their importance, our ability to correctly describe and simulate marine aerosols is still limited by the poor understanding of the link between the properties of the seawater below and the formation of aerosol particles at the air-sea interface and their subsequent evolution in the atmosphere. Mesoscale models are unable to reproduce the number concentrations of aerosol particles in the marine boundary layer and their impact on cloud properties (Merikanto et al., 2009), and a large uncertainty in the projection of future climate can be attributed to a lack of knowledge on marine aerosols (Carslaw et al., 2013). In the Mediterranean area, the marine aerosol emission pathways may be different from those in other parts of the world, firstly due to the specific temperature range and oligotrophic properties of the seawater. Furthermore, high radiation and ozone levels in the atmosphere lead to an intense photochemical activity over a large period of the year. Marine aerosol particles may be produced via two different pathways. Primary marine aerosol production results from wind stress at the sea surface which gives rise to the mechanical production of sea spray aerosol (SSAer), traditionally assumed to be mainly composed of inorganic sea salt, an organic fraction, and water. Secondary marine aerosol production may occur via nucleation or the condensation of gas-phase species onto pre-existing particles. Primary marine aerosol sources (sea spray) are addressed in the present chapter, while nucleation processes from marine precursors are addressed in Sellegri & Rose (2022), as well as marine sources as precursors to SOA in Sartelet (2022).

2 Parameters Influencing the Sea Spray Emission Fluxes

At horizontal wind speeds greater than 4 m s^{-1}, breaking waves generate bubbles that rise in the water column and burst when protruding the surface. The disrupting film cap produces film drops; the rising jet produces jet drops. At wind speeds exceeding about 9 m s^{-1}, sea spray droplets, referred to as spume droplets, are directly blown off the wave tops. Sea spray aerosols make up a large portion of the natural aerosol emissions, with an estimated global contribution between 2000 and 10,000 Tg per year (Gantt & Meskhidze, 2013). Mulcahy et al. (2008) report a relationship between aerosol optical depth (AOD) in clean oceanic air and squared surface wind speed with AODs value of 0.3–0.4 under high wind speed conditions of 15–18 m s^{-1}. Sea spray particle emissions are mainly dependent on wind speed, but also on the sea surface temperature, the sea state (wave height, shape, etc.), and salinity (Grythe et al., 2014). Sea spray number fluxes may depend also on other seawater parameters such as organic matter of biological origin with surfactant properties (Fuentes et al., 2010). Laboratory measurements indicate that the number concentrations of sea spray aerosols produced by bubble bursting mainly peak at sizes of 100 nm and 40 nm for Atlantic Ocean waters (Mårtensson et al., 2003; Fuentes et al., 2010), which does not differ significantly in Mediterranean waters. Indeed, laboratory characterization of sea spray from Mediterranean waters shows that the SSAer size distribution can be best described by two main modes around 40–50 nm and 100 nm, an additional nucleation mode (around 20–25 nm), and a second accumulation mode at 260 nm (Schwier et al., 2015, 2017). The presence of surface-active components may influence the bubble bursting process and hence the size distribution of SSAer. A shift toward smaller particles was observed when artificial surfactants (Sellegri et al., 2006) or biogenic exudates (Fuentes et al., 2010) were added to artificial (inorganic) seawater. Also, several authors report an increase of the 200–300 nm mode number concentrations when organic matter is present (Sellegri et al., 2006; King et al., 2012). However, changes in the shape of SSAer size distribution due to the presence of organic matter in the seawater are not relevant for climate-relevant impact (see Section 4 hereafter).

However, changes in the relative contribution of the different modes to Mediterranean SSAer due to the presence of organic matter are relatively low. Figure 1 shows the mode decomposition of sea spray generated from Mediterranean surface waters from four different experiments. Modes are always found at about 40 nm and 100 nm with similar contributions that dominate the size distributions and at 250 nm with a minor contribution. The most variable mode presence is the nucleation mode at 20 nm. This mode is likely more dependent on the presence of surfactants, although there is still a lot of uncertainty in the factors influencing it and some further research is necessary.

Several parameterizations of the sea spray fluxes published in the literature and used in modelling exercises include a sea surface temperature (SST) dependence (Forestieri et al., 2018; Grythe et al., 2014; Jaeglé et al., 2011; Mårtensson et al., 2003; Ovadnevaite et al., 2014; Salter et al., 2014; Zábori et al., 2012; Albert et al., 2016). Some of these studies agree on a sharp SST dependence of the SSAer number fluxes, with concentrations increasing when temperature decreases below 10 °C (Salter et al., 2014; Zábori et al., 2012). In the SST range representative of the Mediterranean seawater, parameterizations derived from synthetic inorganic seawater experiments show little dependence of the SSAer number concentrations on temperature (Mårtensson et al., 2003; Salter et al., 2014). From laboratory experiments of SSAer generation from natural Mediterranean seawaters, Schwier et al. (2017)

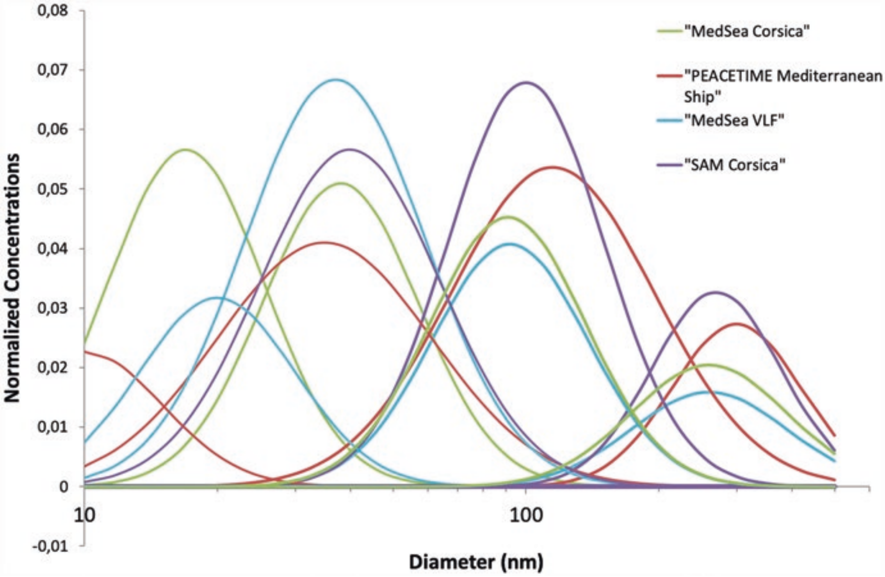

Fig. 1 Lognormal decomposition of the normalized size distributions (fractions of the total number concentration) obtained from sea spray generation experiments during four experiments (MedSea Corsica, MedSea Villefranche-sur-Mer, SAM Corsica, and PEACETIME over a ship cruise across the western Mediterranean Sea) performed with the support of ChArMEx. Thickness of lines is increasing with mode size (from nucleation to accumulation mode)

show that there is a factor of 2 increase in the total number concentration of SSAer when temperature changes from 22 to 30 °C. The same tendency was found in Jaeglé et al. (2011) for SST higher than 20 °C at the global scale, from the comparison of modelled versus observed SSAer concentrations constrained with Tropical Pacific, Tropical Atlantic, and Indian Ocean data.

Given the large variability of SST dependence of sea spray fluxes in the literature, it is likely that other factors influence this SST dependence in natural seawaters. Several laboratory sea spray generation experiments showed differences between number fluxes generated from synthetic inorganic seawaters and those generated from natural seawaters (Tyree et al., 2007; Fuentes et al., 2010; Alpert et al., 2015; Forestieri et al., 2018) or differences between fluxes measured in non-biologically versus biologically rich seawaters (Keene et al., 2017). In general, seawater enriched in biologically derived organics show higher number fluxes. In oligotrophic seawaters such as those found in the Mediterranean Sea, sea spray fluxes were measured as a function of the seawater biogeochemical properties during the PEACETIME ship campaign (a joint MERMEX-ChArMEx initiative; Guieu et al., 2020). Results show that a clear relationship was found between the number fluxes of sea spray and the abundance of nanophytoplankton cells (Sellegri et al., 2021). This relationship was also found in a data set from the South-West Pacific and reinforced when these two data sets are merged, indicating that Mediterranean seawater contains biologically originating organic material with the same properties as found in the global ocean (Sellegri et al., 2021). The mechanism behind the impact of seawater microorganisms on sea spray number emission fluxes is thought to be the production of organic compounds by these microorganisms, which affects the bubble's film stability and hence their thickness.

3 Chemical Composition of Sea Spray

Sea spray contains both inorganic sea salts and organic material. Primary emissions can acquire organic material either as bubbles traverse through the water column or at the ocean surface from the organic rich microlayer (Bigg & Leck, 2008; Blanchard, 1964; Lion & Leckie, 1981) which can exhibit different physical, chemical, and biological properties to oceanic subsurface water (Cunliffe et al., 2013; Walker et al., 2016). Concentrations of marine organic aerosol particles were found to be highly dependent on the biological productivity at the ocean surface, following a seasonal bloom cycle (O'Dowd et al., 2004; Sciare et al., 2009). Organic matter found in sea spray was mainly found as insoluble colloids during a bloom period of the North Atlantic Ocean (Facchini et al., 2008). Current mesoscale and some global atmospheric models use Chl-a in seawater as a proxy to calculate sea spray organic fractions (OMSS) (Langmann et al., 2008; Vignati et al., 2010). They are based on the results from different studies that have linked the total submicron organic mass fraction of sea spray aerosol to chlorophyll-a (Chl-a) levels observed by satellite (Vignati et al., 2010; Albert et al., 2012; Rinaldi et al., 2013). These studies use the organic fraction of marine aerosols sampled at a receptor site, and hence organic found in the samples can be of primary origin, but they can also be the result of condensation of organic gas-phase species during transport. In order to isolate the sea spray fraction of organic concentration from the total organic content of ambient marine aerosols, aerosols may be generated artificially from seawater. Using this approach, Prather et al. (2013) showed a link between the marine particles' organic content and heterotrophic bacteria abundance instead of Chl-a. Also using a sea spray generation system with Mediterranean seawaters sample, Schwier et al. (2015) showed a correlation between the organic fraction of small sea spray particles and Chl-a and heterotrophic and autotrophic bacteria abundance in seawater. The linear relationship found between the organic fraction of sea spray and seawater Chl-a was however not found during a mesocosm experiment using Mediterranean seawater in which the biological activity was sustained with the addition of nutrients (Schwier et al., 2017). The hypothesis is that the nature of the organic matter produced by microorganisms determines the amount of organic matter that is enriched in the sea surface microlayer, and therefore the ratio of organic matter to total sea spray mass transferred to the atmosphere as bubble film drops. When merging data from three different mesocosm experiments performed with Mediterranean seawaters, and excluding the mesocosms that had been enriched with nutriments, one can derive a parameterization that can be applied for the oligotrophic waters of the Mediterranean Sea (Fig. 2). Different aerosol size ranges (Aitken mode and PM_1) need to be considered separately, as a strong increase in the fraction of organics was found with decreasing particle size, as already reported by Facchini et al. (2008) for North Atlantic waters. The resulting parameterizations are the following:

$$\mathrm{OMSS}_{\mathrm{Aitken}} = 0.63 + 0.12\,\mathrm{Ln}\left(\mathrm{Chl}-a\right) \tag{1}$$

$$\mathrm{OMSS}_{\mathrm{PM1}} = 0.33 + 0.04\,\mathrm{Ln}\left(\mathrm{Chl}-a\right) \tag{2}$$

For the inorganic fraction, Schwier et al. (2015) also showed that the composition of Mediterranean SSAer was very similar to the composition of the seawater as reported by Seinfeld and Pandis (2016), although a deficit in chloride compared to the seawater chloride-to-sodium ratio was observed. Since this deficit is observed before SSAer is transported in the atmosphere, one can hypothesize that chemical reactions modifying the Na/Cl ratio already occur within the Mediterranean seawater or at the sea surface. Close to the coasts such as the Mediterranean ones, river inputs and the pollution from urbanized coastal sites with large population densities can cause an excess of phosphate and nitrate compounds (Jamet et al., 2001) that may result in specific reactions.

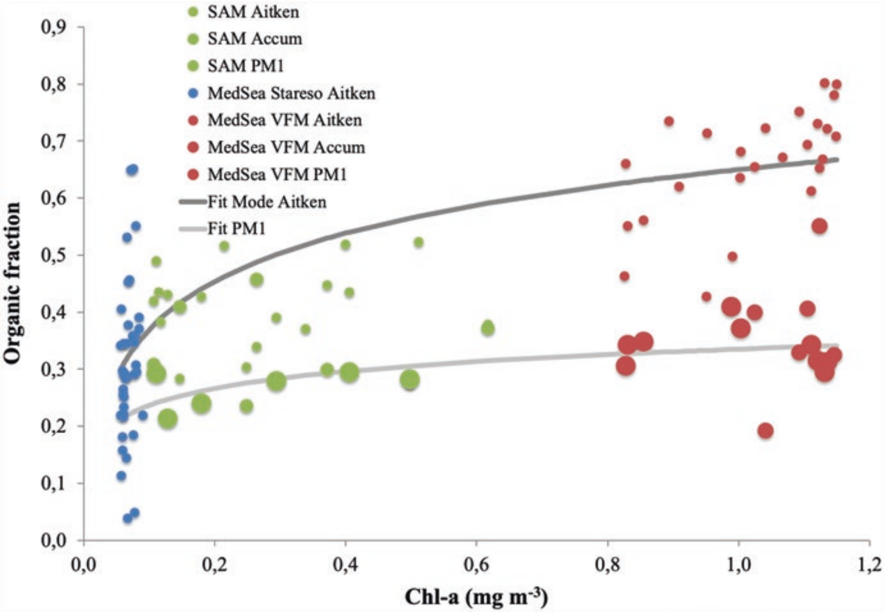

Fig. 2 Organic fraction of sea spray aerosols as a function of Chl-a concentration in the seawater measured from three different mesocosm campaigns (MedSea Stareso (Corsica, bay of Calvi), MedSea VFM (France, bay of Villefranche), and SAM (Corsica, bay of Calvi)) and for different size ranges of the sea spray (Aitken mode, accumulation mode, and total submicron aerosol PM_1). Fit of the organic fraction of each size range (Aitken and PM_1) with a logarithmic law

4 Sea Spray Emissions and Derived Cloud Condensation and Ice Nuclei

Sea spray can act as cloud condensation nuclei (CCN), and thus influences cloud properties, with an efficiency that is dependent on its size and hygroscopicity. CCN properties of sea spray generated from Mediterranean seawater were measured during the SAM experiment in May 2013, showing that the sea spray originating CCN fluxes varied like the total number of sea spray fluxes, with no impact of the organic content of sea spray on its CCN ability (around 60% of sea spray number concentration act as CCN at the 0.3% supersaturation) (Schwier et al., 2017). This conclusion was also observed from seawaters of other origins (King et al., 2012; Moore et al., 2011) and confirmed for Mediterranean seawater during the PEACETIME cruise (Sellegri et al., 2021). The lack of effect of the organic content on CCN properties of sea spray can be explained by the lack of impact of the marine organics on the shape of the sea spray size distribution (and hence its size) (see Fig. 1) and by the little variability of the organic content itself (and hence its hygroscopicity) in oligotrophic waters for a given season (see Fig. 2). The large impact of organic matter on the total sea spray number fluxes that was evidenced from diverse seawater types, including Mediterranean seawater (Sellegri et al., 2021), would however significantly influence the total number of sea spray derived CCN flux to the atmosphere as a function of the seawater biogeochemical properties.

Sea spray ice nuclei (IN) properties are thought to be lower than those of dust particles at low freezing temperatures, but may be significant at warmer temperatures and of major importance in areas where particles of terrestrial origin are absent (DeMott et al., 2016). IN properties of sea spray are poorly understood, although there is evidence that biological factors influence these properties with increased IN efficiencies for higher biological productivities (Bigg, 1973; Schnell & Vali, 1976; DeMott et al., 2016). It appears that sea spray IN properties depend differently on biological factors when considering either cold IN (for temperatures below −18 °C) or warm IN (for temperatures above −18 °C) (McCluskey et al., 2018). While cold ice nucleating sea spray particles are linked to a heat-labile microbial type in the seawater, the warm ice nucleating sea spray particles are rather linked to surface-active DOC enriched in the sea surface microlayer (McCluskey et al., 2018). This finding of two classes of IN sea spray was recently confirmed from Mediterranean seawaters and further conceptualized into a biology-dependent marine IN parameterization (Trueblood et al., 2021).

5 Conclusion and Recommendations

The relationship between sea spray and the seawater biogeochemical properties is complex, due to the presence of processes of physical, chemical, photochemical, and biological nature interacting with each other. Nowadays, parameterizations of sea spray aerosol fluxes include an organic component that is at best a function of Chl-a in the seawater, although a recent modelling study has integrated the different nature of organic classes in the seawater (originating from proteins, lipids, polysaccharides, or humic-like substances) and their surfactant properties for prescribing their transfer to the atmosphere (Burrows et al., 2014). The Mediterranean Sea is oligotrophic, and hence sea spray aerosol particles emitted from Mediterranean waters contain a low level of biologically originating carbon except during phytoplanktonic bloom conditions. Besides, sea surface temperatures are relatively high throughout the year, implying that organic matter will not have a large impact on sea spray number emissions via their impact on the temperature dependence of surfactants. However, the Mediterranean Sea experiences a large amount of radiation which effect on emissions is largely unknown and should be the focus of future research on sea spray aerosol emissions in this area of the world.

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