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## Sea-Spray-Generation Dependence on Wind and Wave Combinations: A Laboratory Study

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1 **Sea Spray Generation Dependence on Wind and**  
2 **Wave Combinations : A Laboratory Study**

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8 **Abstract** This paper investigates the effect of interacting winds and waves  
9 on the surface sea spray generation flux. To this end, the Marine Aerosol  
10 Tunnel Experiment (MATE2019) was conducted at the OSU-Pytheas large  
11 wind-wave tunnel facility at Luminy, Marseille (France), in June-July 2019.  
12 A unique range of air-sea boundary conditions was generated by configuring  
13 the laboratory with four types of wave forcing and five wind speeds ranging  
14 from 8 to 20 m s<sup>-1</sup>. The configurations included both young and developed

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15 waves spanning a wave age (defined in terms of phase speed and friction ve-  
16 locity) range between 1.3 and 9.5. Sea spray concentrations were measured  
17 over the 0.1-47.5  $\mu\text{m}$  radius range at different heights above water, and a flux-  
18 profile method using Monin and Obukhov (1954) theory allowed to estimate  
19 the sea spray generation flux from concentration profiles. Results show that  
20 the flux increases for increased wind-induced wave breaking, and the highest  
21 flux is found for steep and heavily-breaking wind-forced waves. **Scaling analysis**  
22 **shows that the sea spray generation is best correlated with the wave slope vari-**  
23 **ance ( $\langle S^2 \rangle$ ) for larger droplets (20  $\mu\text{m}$  and above, assumed predominantly**  
24 **spume droplets generated by surface tearing). For smaller droplets (7-20  $\mu\text{m}$ ,**  
25 **presumed predominantly jet droplets generated by bubble bursting) highest**  
26 **correlation is found with a non-dimensional number combining  $\langle S^2 \rangle$  and**  
27 **the friction velocity cubed  $u_*^3$ . This is reflected in the formulation of two wave**  
28 **state dependent sea spray generation functions (SSGFs), each valid for wind**  
29 **speeds 12-20  $\text{m s}^{-1}$  and droplet radii 3-35  $\mu\text{m}$ , thereby covering both jet and**  
30 **spume droplet production.**

31 **Keywords** Sea spray generation · Air-sea interaction · Wave slope variance ·  
32 Friction velocity cubed

## 33 1 Introduction

34 Sea spray aerosols formed by the interaction between air and water represent  
35 a major component of the natural aerosol mass (Jaenicke, 1984; Yoon et al.,  
36 2007) with a major role in the Earth radiation budget (Mallet et al., 2003;  
37 Mulcahy et al., 2008). In turn, the light scattering by sea spray aerosols is an  
38 important factor in the context of climate change (Lewis et al., 2004). Sea spray

39 also has a significant impact on air quality, particularly in coastal regions where  
40 anthropogenic emissions are high (Piazzola et al., 2016; Johansson et al., 2019).  
41 However, it remains difficult to numerically model these effects. Significant  
42 predictive uncertainties remain for sea spray aerosol in numerical models with  
43 significant biases observed for commonly used emission schemes (Tsyro et al.,  
44 2011; Chen et al., 2016; Neumann et al., 2016). Therefore, sea spray aerosol  
45 generation and subsequent transport in the atmosphere models need to be  
46 improved (Canepa and Builtjes, 2017).

47 Sea spray can be produced both directly and indirectly from the water  
48 surface. Indirect generation is associated with the relatively small bubble-  
49 mediated jet and film droplets produced by wave breaking, effective for wind  
50 speeds around  $4 \text{ m s}^{-1}$  and above (Blanchard, 1963; Spiel, 1994). The transition  
51 from film to jet droplet dominance is thought to occur at radii 2 - 4  $\mu\text{m}$   
52 (Cipriano and Blanchard, 1981; Woolf, 1997) with jet droplets dominating  
53 over the 2 - 20  $\mu\text{m}$  range and sometimes reaching 50  $\mu\text{m}$  or more (Newitt,  
54 1954; Andreas, 1998). The direct sea spray generation mechanisms activated  
55 at wind speeds higher than 10 - 12  $\text{m s}^{-1}$  (Monahan et al., 1986; Andreas et al.,  
56 2010) consist of surface-tearing of larger (radius  $r > 20 \mu\text{m}$ ) spume droplets  
57 that are directly ejected into the air flow.

58 The direct generation mechanisms have proven to be elusive. Anguelova  
59 et al. (1999) noted the tearing of spume drops at the wave crests by the wind,  
60 and found spume droplet generation to be strongly related to the whitecap  
61 coverage. On a microphysical scale the bag breakup of small canopies of water  
62 produced by wind surface friction at the wave crests is a potential mecha-  
63 nism for spume droplet generation (Mueller and Veron, 2009; Veron et al.,  
64 2012; Troitskaya et al., 2018). All in all, little is known about the generation  
65 of particles of radii  $> 20 \mu\text{m}$  (Veron et al., 2012; Veron, 2015). A better pa-

66 parameterization of the droplet generation flux and transport over this range is  
67 urgently needed, since a major portion of the total volume of sea spray droplets  
68 is thought to be within the 10-200  $\mu\text{m}$  radius range, as stressed by authors  
69 such as Andreas (1992).

70 The sea spray flux or sea spray generation function (SSGF) is known to  
71 depend on numerous environmental parameters such as the wind and wave  
72 fields (De Leeuw, 1986; Iida et al., 1992; Smith et al., 1993), the sea surface  
73 temperature (Mårtensson et al., 2003), and the composition and salinity of the  
74 seawater (Sellegri et al., 2006; Mehta et al., 2019). Although wind speed alone  
75 was traditionally used to parameterize the aerosol generation - cf. the SSGFs of  
76 Monahan et al. (1986); Smith et al. (1993); Andreas (1998) - there is increasing  
77 evidence that this is not sufficiently accurate, and additional environmental  
78 conditions have to be taken into account, wave parameters especially. Various  
79 authors have also considered scaling droplet generation with non-dimensional  
80 numbers such as wave age (Lafon et al., 2007; Laussac et al., 2018), or the  
81 windsea Reynolds number  $R_B$  (Zhao et al., 2006; Ovadnevaite et al., 2014;  
82 Troitskaya et al., 2018). The wave age is defined as the ratio between the wave  
83 phase velocity  $c_p$  and a wind parameter (for example the wind friction velocity  
84  $u_*$ ), and is generally used to describe wave-field development and wave-wind  
85 equilibrium.  $R_B$ , first introduced by Toba and Koga (1986) as  $u_*^2 / (\nu_a \omega_p)$  with  
86 the air kinetic viscosity  $\nu_a$  and the windsea spectrum peak angular frequency  
87  $\omega_p$ , thus describing turbulence at the wind-wave boundary. The role of wave  
88 breaking mechanisms on generation has also been a focus of study with the  
89 use of whitecap coverage to scale film, jet (Laussac et al., 2018) and spume  
90 droplet generation (Anguelova et al., 1999). In a similar fashion, wave energy  
91 dissipation - considered proportional to  $u_*^3$  (the third power of the friction  
92 velocity  $u_*$ ) by several authors - has also been considered for the study of

93 droplet and bubble generation (Newell and Zakharov, 1992; Zhao and Toba,  
94 2001; Zhao et al., 2006; Andreas, 1998, 2002; Fairall et al., 2009).

95 Of the different wave parameters used to scale sea spray generation, the  
96 wave slope has not yet been considered. Nevertheless, it is an interesting candi-  
97 didate because of its strong relation to wave breaking (Stokes, 1880; Duncan,  
98 1981; Ramberg and Griffin, 1987), whitecap production (Banner et al., 2002;  
99 Brumer et al., 2017), and therefore to sea spray bubble-bursting and surface-  
100 tearing generation mechanisms. The wave slope, however, is also thought to  
101 modulate the air flow at the wave crest (Veron, 2015; Richter et al., 2019;  
102 Husain et al., 2019), which is of crucial importance for air-sea momentum  
103 transfer and surface tearing mechanisms. Finally, the relationship between the  
104 wave slope variance and the wind speed at the wind-wave boundary is rela-  
105 tively well documented (Cox and Munk, 1956; Plant, 1982; Vandemark et al.,  
106 2004; Bringer et al., 2013; Lenain et al., 2019).

107 With the advantage of easier instrumentation and control over important  
108 environmental parameters compared to the complexity of the field, the labo-  
109 ratory has proven to be a valuable alternative to the field (open ocean) for the  
110 study of wave-wind boundary processes and sea spray generation (Toba and  
111 Koga, 1986; Fairall et al., 2009; Ortiz-Suslow et al., 2016; Troitskaya et al.,  
112 2018). Therefore, we conducted a series of experiments in the wave-wind tunnel  
113 at the **OSU Pytheas (Observatoire Sciences de L’Univers) Ocean-Atmosphere**  
114 **laboratory in Luminy, France**. In the field, a wide range of wind and wave  
115 combinations can be encountered. To reproduce a similar range in environ-  
116 mental conditions the laboratory experiments also included a unique range of  
117 wind and wave conditions, which, to the knowledge of the authors, is the most  
118 extensive range ever used for studying the SSGF in a laboratory configura-  
119 tion. The aim is to **develop** a universal wind and sea state-dependent SSGF

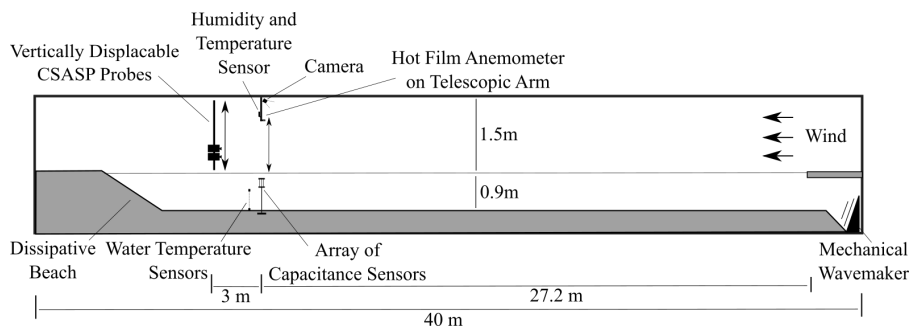
120 applicable for radii typical of jet and spume droplets. To this end, from June  
121 to July 2019, sea spray aerosol size concentrations were measured for a total  
122 of twenty wind and wave combinations as part of the Marine Aerosol Tunnel  
123 Experiment (MATE2019). The experiment is described in Sect. 2, wind and  
124 wave results are discussed in Sect. 3, and sea spray results in Sect. 4. Study  
125 results include a comparison between the laboratory data and other labora-  
126 tory and field SSGFs presented in the literature. **Finally, two new SSGFs are**  
127 **introduced in Sect. 5, one depending on the wave slope variance alone, and a**  
128 **second on both the wave slope variance and  $u_*^3$ . Both SSGFs combine labora-**  
129 **tory and field data, and are valid for wind speeds 12-20 m s<sup>-1</sup> and radii 3-35**  
130  **$\mu\text{m}$ .**

## 131 **2 Methods**

### 132 2.1 The wind-wave tunnel

133 The UMS Pytheas ocean-atmosphere interactions facility at Luminy (Mar-  
134 seille, France), schematically represented in Fig. 1, consists of a water tank  
135 with a wind tunnel on top of it. The facility currently allows the use of fresh  
136 water only. The water tank is 40 meters long, 2.6 meters wide, and has a  
137 wave-dissipating beach at the downwind end to avoid wave reflection. On the  
138 upwind end, the facility is equipped with a programmable submerged piston-  
139 like wavemaker located at the bottom of a cavity below the air tunnel and  
140 controlled by an electrohydraulic motor. Wave properties such as amplitude,  
141 frequency and slope can therefore be selected. Waves can attain amplitudes  
142 reaching 15 cm, approximately. The air channel ceiling is slightly inclined to  
143 the fetch to avoid the airflow acceleration and the related longitudinal pressure

144 gradient generated by the thickening of the wall and water surface boundary  
 145 layers (Coantic et al., 1981). To reduce turbulence as the air flow encounters  
 146 the water surface, a thin buoyant sheet is placed at the surface of the water  
 147 at the entrance of the tunnel. The wind speed in the facility is adjustable,  
 148 with winds reaching  $15 \text{ m s}^{-1}$ , approximately. Glass walls on either side of the  
 149 laboratory made it possible to monitor the experiment at the location of the  
 150 instruments (given below).



**Fig. 1** Side view of the experimental configuration in the laboratory

151 For the experiments in June-July 2019, the water depth was set to 90 cm,  
 152 leaving 150 cm between the water surface and the ceiling of the wind tunnel.  
 153 As schematically represented in Fig. 1, instruments used for the measurements  
 154 of air, water, wind and wave characteristics were placed at the 27.2 meter fetch  
 155 mark. A sensor positioned 81 cm above the mean water level (MWL) was used  
 156 to measure air relative humidity ( $RH$ ) and temperature ( $T_a$ ). Vertical profiles  
 157 of horizontal wind speed and air temperature between 15 and 66 cm above  
 158 MWL were obtained using sensors mounted on a telescopic arm. The water  
 159 temperature was measured at both 20 and 60 cm depth beneath the MWL.  
 160 A camera with 60 Hz sampling frequency placed on the tunnel ceiling allowed  
 161 estimating the whitecap coverage during the experiments from color images.  
 162 Though not schematically represented in Fig. 1 a higher-frequency 128 Hz  
 163 camera was used to film through the glass wall of the tunnel for monitoring  
 164 purposes only. Finally, at the 30 meter fetch mark, vertical profiles of the sea



165 spray number concentration were measured using two particle probes mounted  
166 onto a vertically displaceable frame.

## 167 2.2 Wave generation and characterisation

168 The water surface elevation was measured by an array of two calibrated capac-  
169 itance wave gauges placed at the 27.2 meter fetch mark. The sample frequency  
170 for the wave gauges was 256 Hz allowing to sample a wide range of wave surface  
171 elevation and slope frequency spectra. The sensors were aligned orthogonally  
172 to the general wave direction, and were placed 1 cm from each other to ensure  
173 accurate wave slope measurements.

174 In the present study, four types of waves were generated. Five different wind  
175 speeds between 8-20 m s<sup>-1</sup> were applied over each wave type, in a total of 20  
176 laboratory wind-wave configurations. A first type of waves was generated solely  
177 by the wind, resulting in pure wind waves. The other three types of waves were  
178 generated using the piston wavemaker and are referred to as short wave forcing  
179 (peak frequency  $f_p = 1.3$  Hz and wavelength  $\lambda = 0.92$  m), intermediate wave  
180 forcing ( $f_p = 1.1$  Hz and  $\lambda = 1.29$  m), and long wave forcing ( $f_p = 0.8$  Hz and  $\lambda$   
181  $= 2.4$  m). These conditions were selected to ensure that near-equilibrium with  
182 the overlying wind field was either never met, or met at different wind speeds.  
183 Deep water conditions are theoretically confirmed for all types of forcing except  
184 the long wave case. This is also the wave type that is least forced by the wind  
185 and can be considered representative of the wind over swell condition that can  
186 be found in the field. Together, the 20 tunnel wind and wave combinations  
187 span a range of wave ages  $\frac{c_p}{u_*}$  between 1.3 and 9.6.

188 The wave sensor array provides several parameters that are useful to re-  
189 late the wave properties measured at the 27.2 m fetch mark to the sea spray  
190 concentrations measured at the 30 m fetch mark. The significant wave height  
191  $H_S$  is determined using:

$$192 \quad H_s = 4\sigma_\eta$$

193 where  $\sigma_\eta$  is the standard deviation of the surface elevation ( $\eta$ ) time series  
194 measured by the wave sensor array.

195 The wave slope variance  $\langle S^2 \rangle$  is considered equal to the wave mean  
196 square slope (MSS). It is calculated from the time series of the wave slope  $S$ ,  
197 for wavelengths  $\lambda > 1$  cm (strong majority of waves dominated by gravity).  
198 The use of classical Fourier analysis allows to determine the peak frequency  
199  $f_p$  from the wave elevation energy spectra. Finally, the wave phase speed  $c_p$   
200 is determined from the phase shift between the wave gauges as the waves  
201 propagated along the wave sensor array.

### 202 2.3 Whitecap measurements

203 To quantify the wave breaking, the whitecap coverage  $W(\%)$  is estimated from  
204 video colour images taken by a camera mounted at the tunnel ceiling (cf. Fig.  
205 1). The system takes images at 60 Hz frequency with dimensions  $2704 \times 2028$ .  
206 These images are spatially referenced to allow to estimate the approximate  
207 surface area of each pixel. To identify the whitecaps, a separation method is  
208 used that applies a greyscale conversion and a subsequent intensity threshold  
209 to the image. The surface areas of the pixels above threshold are subsequently  
210 added and divided by the total image surface area for an estimate of the

211 whitecap surface coverage  $W(\%)$ . Using a time series of images, it was found  
 212 that  $W(\%)$  estimates converge to an average value within approximately 120  
 213 frames. This method for whitecap estimation is commonly used (e.g. Lafon  
 214 et al. (2007); Lenain and Melville (2017); Brumer et al. (2017)), and although  
 215 the selection of the intensity threshold may be subjective, we expect that our  
 216 whitecap coverage estimates capture the relative amount of wave breaking  
 217 induced by various wind and wave conditions.

## 218 2.4 Wind measurements

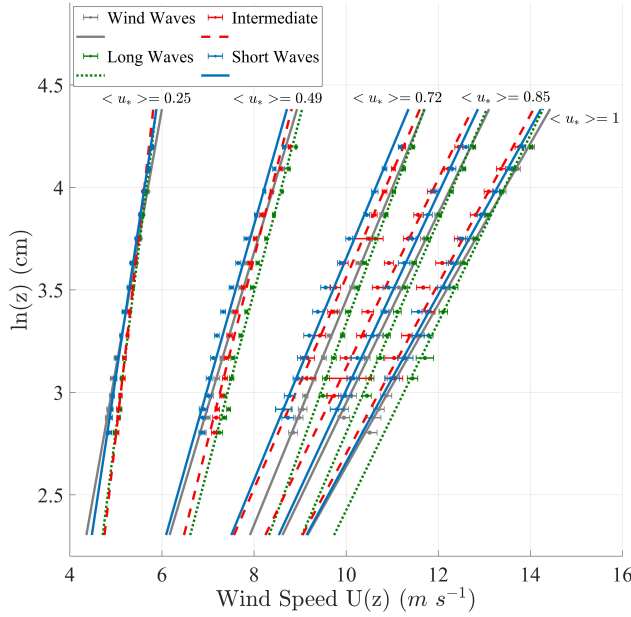
219 A hot film wind sensor (E+E Elektronik, Langwiesen, Austria) mounted onto  
 220 a telescopic arm was used to measure vertical profiles of horizontal wind speed  
 221  $U$  at the 27.2 meter fetch mark (cf. Fig. 1). This sensor was calibrated against a  
 222 reference sonic anemometer on several occasions during the experiments. Some  
 223 subsets of wind data obtained at 15 cm of the MWL were eliminated from the  
 224 dataset to avoid erroneous measurements caused by large droplets impacts on  
 225 the hot film sensor. To ensure representative wind statistics, convergence of  
 226 the wind speed time-averages was achieved for each height  $z$  along the  $U(z)$   
 227 profiles, **with averages spanning over 20 to 80 minutes in accordance with the**  
 228 **sea spray aerosol sample durations.**

229 The wind speed reference used in this study is  $U_{10}$ , i.e. the wind speed  
 230 at 10 meters above MWL. Assuming a logarithmic wind profile and neutral  
 231 conditions, Monin and Obukhov (1954) scaling allows evaluation of  $U_{10}$  (m  
 232  $s^{-1}$ ) and the friction velocity  $u_*$  (m  $s^{-1}$ ) from the measured  $U(z)$  profile values  
 233  $U(z)$ :

$$U(z) = \frac{u_*}{k} \ln\left(\frac{z}{z_0}\right) \quad (1)$$

234 where  $k = 0.4$  the von Kármán constant,  $z$  is the measurement height above  
235 MWL, and  $z_0$  the roughness length. The latter is retrieved by extending the  
236 wind profile with data from the more elevated part of the wind profile that is  
237 not significantly affected by the wave field. During the experiments,  $z_0$  esti-  
238 mates varied between 0.2 and 2.5 mm.

239 **Figure 2 presents the experimental wind profiles for all 20 laboratory config-**  
240 **urations (data points with horizontal error bars) as well as the corresponding**  
241 **theoretical profiles obtained with Eq. 1.** For clarity and indicative purposes  
242 only, are also presented the average friction velocities  $\langle u_* \rangle$  calculated over  
243 the 4 different wave types for each individual tested wind speed. The experi-  
244 mental profiles exhibit near-logarithmic behaviour, and the gradient increases  
245 for increasing reference wind speed  $U_{10}$ , in accordance with a fully developed  
246 turbulent layer near the water surface as encountered in the field. This is fur-  
247 ther evidenced by the windsea Reynolds number ( $R_B$ ) found greater than  $10^3$   
248 in the laboratory for reference wind speeds  $U_{10}$  of  $12 \text{ m s}^{-1}$  or more, thereby  
249 signifying that the air flow in the boundary layer is in a fully turbulent regime  
250 and windsea breaking occurs (Toba et al., 2006).



**Fig. 2** Measured  $U(z)$  profiles for all twenty laboratory configurations (horizontal bars) with associated theoretical profiles (lines) calculated with Eq. 1. Average friction velocities values  $\langle u_* \rangle$  pertain to the five wind speeds set in the laboratory.

## 251 2.5 Sea spray aerosol measurements

252 The measurement of sea spray in the tunnel was carried out using two CSASP  
 253 (Classical Scattering Aerosol Spectrometer) probes (Particle Measurement  
 254 Systems, Boulder, Colorado, USA). This type of probes has been proven re-  
 255 liable in numerous experiments by the authors and others (e.g., Frick and  
 256 Hoppel (2000); Savelyev et al. (2014); Petelski et al. (2014)). Our two probes  
 257 have overlapping particle radius ranges allowing a combined range of 0.1 to  
 258  $47.5 \mu\text{m}$ . Prior to the experiments, both probes were calibrated with latex  
 259 particles of known sizes. The CSASP-100-HV-ER probe samples by rotating  
 260 over 4 sets of 15 size bins at a time, in total spanning from 0.5 to  $47.5 \mu\text{m}$   
 261 radius. The CSASP-200, samples over a single set of 31 size bins ranging from

262 0.1 to 10  $\mu\text{m}$  radius. The probes send data to the controller every second,  
263 and aerosol number concentrations for each bin size are obtained by averaging  
264 over a chosen sample (integration) time. Since number concentrations were  
265 expected to decrease with increasing height above the water surface, sample  
266 times varied from a minimum of 20 minutes at  $z = 35$  cm to a maximum of  
267 80 minutes at  $z = 82$  cm in order to improve droplet count statistics. Never-  
268 theless, particle concentrations for radii greater than 35  $\mu\text{m}$  were statistically  
269 unreliable and had to be discarded. It was verified that droplet concentrations  
270 were stationary over the duration of the experiment once equilibrium for a  
271 particular laboratory configuration was established.

272 During the experiments, the probes were placed on top of each other.  
273 Droplet number concentrations were measured at five different heights for each  
274 of the twenty laboratory configurations. For the CSASP-100-HV-ER, samples  
275 were made at heights  $z = 35, 41, 51, 65$  and 82 cm above the MWL. These  
276 heights are used as reference in this study, and the CSASP-200 concentrations  
277 are interpolated to these heights. The air flow inside a long wind-tunnel is not  
278 completely homogenous, and very slight transverse counter-rotating stream-  
279 wise vortices or cells may appear (Holmes et al., 1996; Pope, 2000), with a  
280 very weak divergence or convergence zone along the middle of the width of the  
281 tunnel. As it is usually done in the IRPHE wind-wave facility, the probes were  
282 slightly shifted away from the middle axe line of the facility. For all laboratory  
283 configurations, both aerosol probes were temporarily positioned at the tunnel  
284 entrance to verify that no background aerosols entered the tunnel, which im-  
285 plies that all particles detected at the 30 m fetch mark are solely produced  
286 over the upwind 30 meter fetch length.

287 After generation at the water surface, the hygroscopic aerosols adjust their  
288 size to the ambient humidity and temperature of the air flow. This process is

289 different for salt water than for the fresh water used in the tunnel, as freshwater  
 290 droplets can evaporate more quickly and completely than saltwater droplets  
 291 that reach an equilibrium size (e.g. Pruppacher and Klett (1978); Andreas  
 292 (1989); Fairall (1990); van Eijk et al. (2001); Mueller and Veron (2014); Mehta  
 293 et al. (2019)). During the experiments, the relative humidity (RH) in the  
 294 laboratory varied between 79 and 82%, with an average water temperature at  
 295 0.2 m depth of 18°C, and an average air temperature  $T_a$  at 85 cm height of  
 296 25°C. We therefore assume that sea spray number concentrations correspond  
 297 (roughly) to  $RH = 80\%$ . Since the rate of freshwater droplet evaporation at  
 298  $RH = 80\%$  is relatively small, we assume that our measured droplets are  
 299 representative for saltwater droplets at their  $RH = 80\%$  equilibrium radii.  
 300 However, this assumption will fail if the freshwater droplets are relatively small  
 301 or have long residence times before reaching the aerosol probes, as evaporation  
 302 can become substantial (Veron, 2015). To avoid such evaporation effects over  
 303 the length of the wave-wind facility, only radii greater than  $7 \mu\text{m}$  are considered  
 304 for the source functions proposed in this study (see Sect. 5) in accordance with  
 305 Fairall et al. (2009).

306 Aerosol concentrations are often expressed as number concentrations  $dN/dr$   
 307 ( $\text{cm}^{-3} \mu\text{m}^{-1}$ ; the number of particles  $N$  of a given radius  $r$  measured per unit  
 308 air) or volume concentrations  $dV/dr$  (the volume for a given radius measured  
 309 per unit air). In this study, we use mostly number concentrations, with a  
 310 single exception (Sect. 4, Fig. 6). The conversion from number to volume  
 311 concentrations ( $\text{cm}^{-3} \text{cm}^{-3} \mu\text{m}^{-1}$ ) is made by assuming that the droplets are  
 312 spherical:

$$\frac{dV}{dr} = \frac{dN}{dr} \frac{4}{3} \pi r^3 \quad (2)$$

313 As discussed above, we may consider that the freshwater droplet distributions  
 314 measured in the tunnel are analogous to salt water droplets at  $RH = 80\%$ .

315 Since the processes that generate the freshwater droplets in the tunnel are the  
316 same as for sea spray in the field, we will denote the droplets in the tunnel as  
317 sea spray in the remainder of this paper.

### 318 **3 Tunnel wave and wind results**

#### 319 **3.1 Wave measurements**

320 Table 1 characterises all 20 laboratory configurations in terms of the significant  
321 wave height  $H_s$ , the wave slope variance  $\langle S^2 \rangle$ , the friction velocity  $u_*$ ,  
322 and the whitecap coverage  $W(\%)$ . The table shows that the wind field forces  
323 the wave field, resulting in increased wave height, wave slope variance and  
324 breaking, as evidenced by  $H_s$ ,  $\langle S^2 \rangle$  and  $W(\%)$ . In some cases however,  
325 significant wave height and wave slope decrease for high wind speeds as wave  
326 energy dissipation by wind-induced breaking becomes temporarily greater than  
327 the energy input from the wind to the waves. This is especially the case for  
328 the intermediate wave forcings for wind speeds increasing from 16 to 18 m s<sup>-1</sup>  
329 (Table 1) where wave breaking was especially high. Further evidence of the  
330 forcing of the wave field by the wind is the decrease in  $f_p$  for increasing wind  
331 speed observed at the 27.2 fetch mark, with the exception of the long wave  
332 forcing which conserves the 0.8 Hz frequency prescribed by the wavemaker  
333 **throughout** the experiments. The momentum transfer from the wind to the  
334 waves is therefore relatively low for the long wave forcing in comparison to  
335 the other wave forcings, resulting in relatively low  $U(z)$  gradients (Fig. 2) and  
336 whitecap coverages  $W(\%)$  (Table 1).



**Table 1** Wave-wind boundary characteristics for the different laboratory configurations: significant wave height  $H_s$  (cm), wave slope variance ( $\langle S^2 \rangle$ ), friction velocity  $u_*$  ( $\text{m s}^{-1}$ ), wave phase speed  $c_p$  ( $\text{m s}^{-1}$ ) and whitecap coverage  $W(\%)$

| Forcing                        | $U_{10} = 8 \text{ m s}^{-1}$ |                       |       |       |         | $U_{10} = 12 \text{ m s}^{-1}$ |                       |       |       |         |
|--------------------------------|-------------------------------|-----------------------|-------|-------|---------|--------------------------------|-----------------------|-------|-------|---------|
|                                | $H_s$                         | $\langle S^2 \rangle$ | $u_*$ | $c_p$ | $W(\%)$ | $H_s$                          | $\langle S^2 \rangle$ | $u_*$ | $c_p$ | $W(\%)$ |
| Wind                           | 3.2                           | 0.026                 | 0.32  | 0.78  | 0.13    | 5.5                            | 0.039                 | 0.54  | 0.99  | 0.29    |
| Short                          | 9.7                           | 0.037                 | 0.28  | 1.19  | 0.28    | 12.4                           | 0.053                 | 0.52  | 1.29  | 0.62    |
| Inter.                         | 13.2                          | 0.033                 | 0.21  | 1.42  | 0.32    | 16.7                           | 0.047                 | 0.46  | 1.49  | 0.53    |
| Long                           | 12.0                          | 0.024                 | 0.23  | 2.2   | 0.16    | 13.8                           | 0.03                  | 0.48  | 1.7   | 0.25    |
| $U_{10} = 16 \text{ m s}^{-1}$ |                               |                       |       |       |         |                                |                       |       |       |         |
| Wind                           | 8.3                           | 0.056                 | 0.75  | 1.1   | 0.8     | 9.63                           | 0.062                 | 0.89  | 1.25  | 1.25    |
| Short                          | 13.7                          | 0.063                 | 0.76  | 1.33  | 1.17    | 15.4                           | 0.066                 | 0.85  | 1.54  | 1.45    |
| Inter.                         | 20.1                          | 0.087                 | 0.8   | 1.50  | 2.55    | 18.4                           | 0.08                  | 0.89  | 1.59  | 4.65    |
| Long                           | 16.5                          | 0.046                 | 0.66  | 2.1   | 0.73    | 19.6                           | 0.055                 | 0.79  | 2.14  | 0.84    |
| $U_{10} = 20 \text{ m s}^{-1}$ |                               |                       |       |       |         |                                |                       |       |       |         |
| Wind                           | 11.3                          | 0.073                 | 1.05  | 1.32  | 2.0     |                                |                       |       |       |         |
| Short                          | 15.2                          | 0.072                 | 1.01  | 1.63  | 2.25    |                                |                       |       |       |         |
| Inter.                         | 17.6                          | 0.078                 | 1.0   | 1.57  | 3.19    |                                |                       |       |       |         |
| Long                           | 21.6                          | 0.065                 | 0.9   | 2.15  | 1.19    |                                |                       |       |       |         |

337 This non-linear behaviour of the parameters listed in Table 1 highlights  
338 the importance of correctly understanding the boundary processes, such as  
339 the state of wind-wave equilibrium. Though not shown here,  $f^{-5}$  and  $f^{-4}$   
340 spectral shapes were identified for the individual wave spectra, but it proved  
341 difficult to quantify the wave-wind equilibrium this way. We therefore abide by  
342 the heuristic idea that when the phase speed  $c_p$  is greater than the overlying  
343 wind speed (e.g. the friction velocity  $u_*$ ) then the underlying waves are no  
344 longer forced by the wind field. Thus we use the wave age defined as  $c_p/u_*$  to  
345 quantify the wind-wave equilibrium.

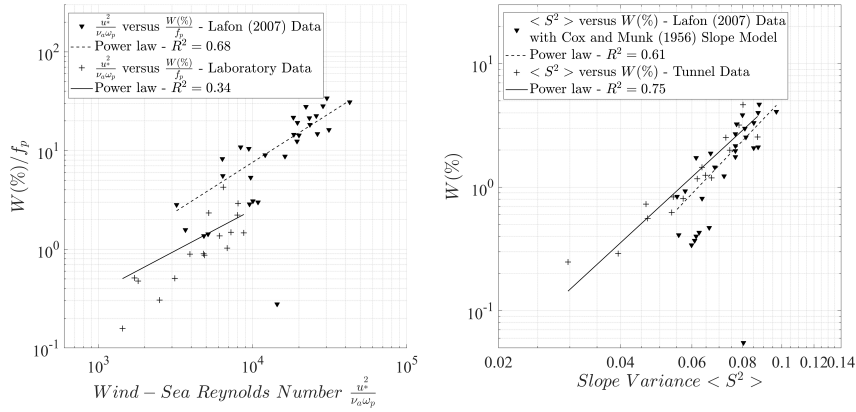
346 Our laboratory configurations yield wave age  $c_p/u_*$  values ranging from 1.3  
347 to 9.6 where the higher values correspond to the long wave forcing combined  
348 with the lowest wind speed ( $8 \text{ m s}^{-1}$ ). These values are generally lower than  
349 those recorded in the field because of the comparatively much shorter fetch  
350 length in the laboratory.

### 3.2 Whitecap measurements

As previously noted, several studies have related whitecapping to the generation of film, jet and spume droplets. For comparison, **it is therefore important to ascertain whether the whitecap production in the laboratory differs from the field.** To this end, we use data collected during the EMMA campaign which took place in Toulon-Hyeres bay, yielding wave, wind and whitecap coverage data for wind speeds in the 10 - 18 m s<sup>-1</sup> range and wave ages  $c_p/u_*$  in the 14 - 28 range Lafon et al. (2007). We compare our laboratory whitecap coverage to that measured in fetch-limited field conditions during EMMA, using a similar image-processing technique with intensity threshold separation (data available in Lafon et al. (2007)). Laboratory data obtained at 8 m s<sup>-1</sup> wind speed are not included in this comparison because the relatively small amount of wave breaking may reduce the accuracy of the whitecap coverage estimates.

Multiple independent scaling studies have proven  $R_B$  to be a successful scaling tool for  $W(\%)$  (Zhao and Toba, 2001). However, the evaluation of  $R_B$  requires the wave peak frequency  $\omega_p$ , which differs significantly between the laboratory and the field. **The relatively short fetch in the laboratory and the wavemaker settings lead to shorter wavelengths and a relatively greater number of wave crests per unit area (Mueller and Veron, 2009).** To allow a better comparison between the laboratory and the field, the  $W(\%)$  value are therefore normalised by the wave peak frequency. Figure 3a shows the relationship between the normalised  $W(\%)$  and  $R_B$ . The data from both the laboratory (+) and the field (▼) display a very similar trend yielding nearly identical slopes when fitted by a power law. In absolute magnitude there is a factor 1.5 difference between the laboratory and the field with the lower  $W(\%)$  values for the laboratory. A possible explanation is the absence of surfactants

377 and salt in the laboratory, which in the field contribute to whitecap formation  
 378 and increase whitecap lifetime (Callaghan et al., 2012). Consequently, the lower  
 379 rate of whitecap formation and lifetime in the laboratory may signify that  
 380 our whitecap coverage is more representative for the **process of active wave**  
 381 **breaking, which we refer to as the wave breaking coverage.**



**Fig. 3** Comparison between laboratory and field (Lafon et al., 2007) (a)  $f_p$ -normalised whitecap coverage  $W(\%)$  estimates as function of the windsea Reynolds number  $R_B$ , and (b)  $W(\%)$  as function of the wave slope variance  $\langle S^2 \rangle$ . Lines and corresponding  $R^2$  statistics represent power laws fitted to the data.

382 A further comparison between the laboratory and the field is obtained by  
 383 scaling  $W(\%)$  to the wave slope variance  $\langle S^2 \rangle$ . In this case,  $W(\%)$  does  
 384 not need to be normalised by the peak frequency as the wave slope variance  
 385  $\langle S^2 \rangle$  integrates a very large part of the wave frequency spectrum and  
 386 corresponding wave slopes, unlike the peak wave characteristics required for  
 387 windsea Reynolds number  $R_B$ . For the EMMA campaign data (Lafon et al.,  
 388 2007),  $\langle S^2 \rangle$  is estimated from the measured wind speed data (Cox and  
 389 Munk, 1956) :

$$\langle S^2 \rangle = 0.003 + 5.12 \times 10^{-3} U_{12.5} \pm 0.004 \quad (3)$$

390 where the wind speed  $U_{12.5}$  is calculated from the EMMA campaign  $U_{10}$  mea-  
 391 surements using classical Monin and Obukhov (1954) theory. It is worth noting  
 392 that Eq. 3 (Cox and Munk, 1956) was determined from airborne observations  
 393 of sun glitter resulting in the contribution of a wide range of wave scales in-  
 394 cluding non-breaking waves, which is to some extent comparable to the wide  
 395 range of wavelengths included in the laboratory wave slope variance.

396 Figure 3b presents the  $W(\%)$  scaling by  $\langle S^2 \rangle$ , demonstrating that lab-  
 397 oratory (+) and field ( $\blacktriangledown$ ) data almost overlap. The laboratory data is well  
 398 fitted ( $R^2 = 0.75$ ) by a the power law:

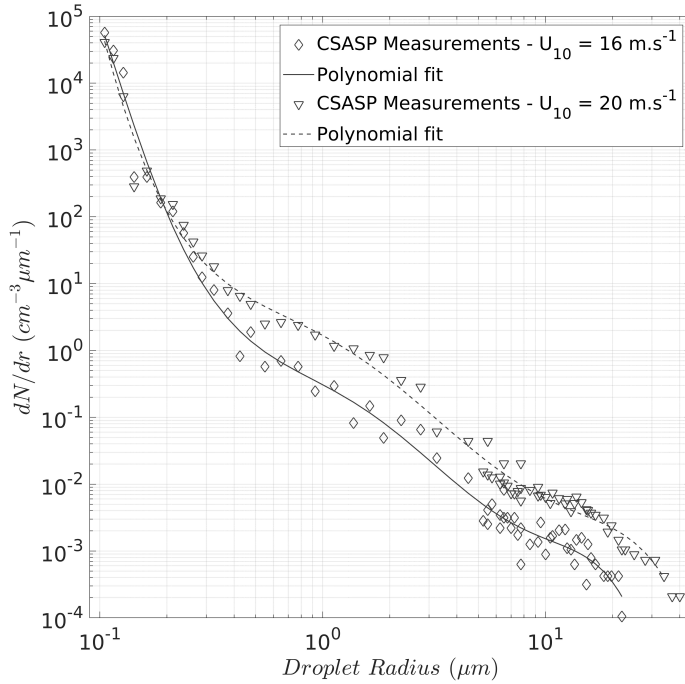
$$W(\%) = 6.02 \times 10^3 \langle S^2 \rangle^{3.025} \quad (4)$$

399 The good fit of this power law to field data (Fig. 3b) suggests that Eq. 4 may  
 400 have a validity range extending to both the laboratory and the field. The strong  
 401 correlation between  $\langle S^2 \rangle$  and  $W(\%)$  suggests that the wide range of the  
 402 wave slope spectrum inherent to  $\langle S^2 \rangle$  allows to separate breakers and non  
 403 breakers. This corroborates conclusions from a north-Pacific field campaign  
 404 (Schwendeman and Thomson, 2015), where the wave slope variance amongst  
 405 the different existing slope parameterizations is found to scale best with  $W(\%)$   
 406 by considering the slopes of the shorter waves, thought to be important for  
 407 whitecap production. In contrast, the scaling of whitecap coverage with wave  
 408 steepness (often written as  $H/(2\lambda)$ ) has proven difficult (Schwendeman and  
 409 Thomson, 2015) or even unsuccessful (Holthuijsen and Herbers, 1986).

## 410 **4 Laboratory sea spray results**

### 411 4.1 Sea spray aerosol size distribution spectra

412 Figure 4 presents two typical size distributions as measured in the laboratory.  
413 The figure shows the sea spray distributions for wind speeds  $U_{10} = 16 \text{ m s}^{-1}$   
414 and  $U_{10} = 20 \text{ m s}^{-1}$ , measured 55 cm above MWL and during the intermediate  
415 wave forcing. For convenience, polynomial functions (solid and dashed lines for  
416 16 and 20  $\text{m s}^{-1}$ , respectively) have been fitted to the distributions. The poly-  
417 nomial functions show that the number concentration of all but the smallest  
418 particles increases with increasing wind speed. This behaviour is observable  
419 at any height above MWL and for all four types of wave forcing. The con-  
420 centration increase is consistent with the increased  $W(\%)$  for increasing wind  
421 speeds (cf. Table 1) and hence, the enhanced droplet generation. Furthermore,  
422 the size distribution shows a relative abundance of particles with sizes around  
423 1 and 10  $\mu\text{m}$ . This corroborates the established size ranges of film and jet  
424 droplets, respectively. The droplet spectra for larger radii change markedly at  
425 wind speeds lower than  $12 \text{ m s}^{-1}$ , where number concentrations for  $r > 15 \mu\text{m}$   
426 are negligible. This corroborates the assumption that these larger droplets are  
427 predominantly spume droplets, and that their generation mechanism activates  
428 at wind speeds around  $12 \text{ m s}^{-1}$ . This was confirmed by our high-speed video  
429 footage, which revealed surface tearing and bag break-up only for wind speeds  
430 higher than  $12 \text{ m s}^{-1}$ . In conclusion, the above observations suggest that sim-  
431 ilar droplet generation mechanisms are active in the laboratory and in the  
432 field.

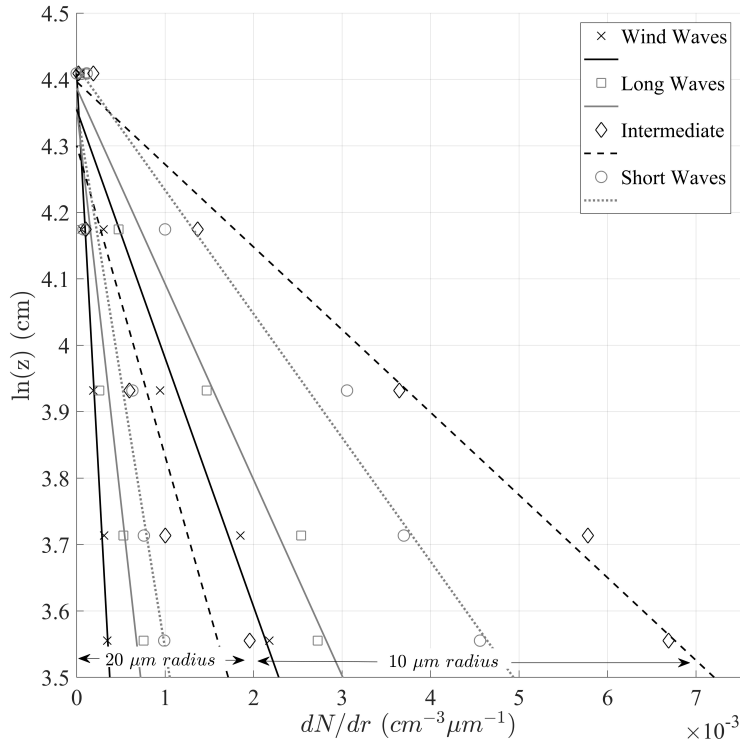


**Fig. 4** Sea spray number concentration size distribution at 55 cm above the MWL for the intermediate wave forcing.

433 Let us now consider the vertical distribution of the freshly generated parti-  
 434 cles. Figure 5 shows number concentration profiles obtained at  $U_{10} = 20 \text{ m s}^{-1}$   
 435 as a linear function of the natural logarithm of height  $\ln(z)$ . Two sets of profiles  
 436 are shown, for droplets of 10 and 20  $\mu\text{m}$  radius, and each set contains data for  
 437 the four different types of wave forcing. The symbols of individual data points  
 438 refer to the type of wave forcing. The lines denote linear functions fitted on  
 439 the experimental data. Figure 5 suggests that the vertical number concentra-  
 440 tion profiles of sea spray are near-logarithmic with height above MWL. This  
 441 behaviour is general for all radii in the 7-35  $\mu\text{m}$  range. For the three highest  
 442 wind speeds,  $R^2$  values of the individual fits all exceed 0.95. Though not pre-  
 443 sented in Fig. 5 for clarity, the standard deviation in number concentrations is

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444 highest closest to the water surface, especially for intermediate and short wave  
445 forcing, but remains very small with a maximum value approaching  $10^{-5} \text{ cm}^{-3}$   
446  $\mu\text{m}^{-1}$  at  $z = 0.35 \text{ m}$ . As the environmental conditions were stationary during  
447 the measurements for each individual laboratory configuration, this greater  
448 variability (standard deviation) in near-surface concentrations could be asso-  
449 ciated with wave-induced **and phase-locked modulation of the airflow, which**  
450 **can lead to flow separation. These mechanisms have recently been observed in**  
451 **microphysical laboratory experiments (Buckley and Veron, 2019; Richter et al.,**  
452 **2019; Husain et al., 2019)**. Whether this modulation effect also caused stronger  
453 fluctuations in the near-surface sea spray concentrations in the laboratory re-  
454 mains hypothetical as the maximum sample frequency of the particle probes  
455 does not allow to resolve these fluctuations assumed to occur at frequencies  
456 approaching  $f_p$  or higher.

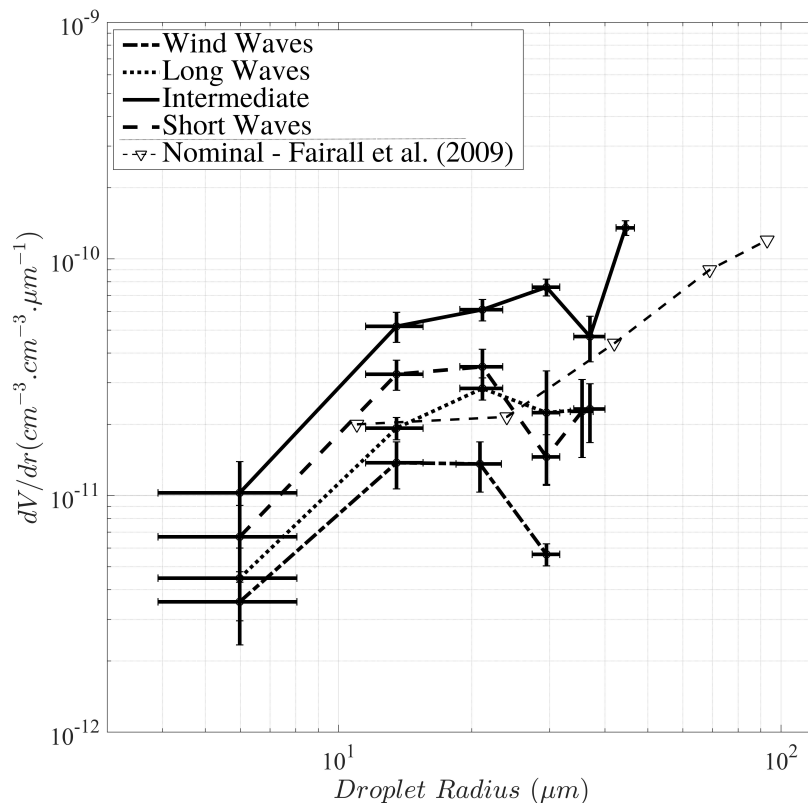


**Fig. 5** Sea spray number concentration profiles at  $20 \text{ m s}^{-1}$  winds as function of height, represented as  $\ln(z)$ . Symbols denote experimental data, lines linear fits to the data.

457 Figure 5 also demonstrates that the type of forcing affects the vertical  
 458 concentration gradients. The stronger gradients are observed for intermediate  
 459 and short wave forcings, whereas the forcing by wind alone results in a less  
 460 pronounced concentration decrease with height. To further investigate the ef-  
 461 fect of sea state, Fig. 6 focuses on the aerosol size distributions over the 6 -  
 462  $47.5 \mu\text{m}$  radius range obtained at  $z = 35 \text{ cm}$ , and for wind speed  $U_{10} = 20 \text{ m}$   
 463  $\text{s}^{-1}$ . Individual datapoints represent an average over several particle bin sizes  
 464 (spanning  $\pm 4 \mu\text{m}$ ) for clarity, and horizontal and vertical error bars illustrate  
 465 the standard deviations in average size and concentration, respectively. The  
 466 data is presented on a log-log scale, and individual datapoints are connected



467 by lines to provide an indication of the volume spectrum. As identified by the  
 468 legend in Fig. 6, we present data for the four types of wave forcings using in  
 469 the laboratory.



**Fig. 6** Sea spray volume concentrations for  $U_{10} = 20 \text{ m s}^{-1}$  at 35 cm above the mean water level, for the four forcings used in the laboratory. SPANDEX data measured at 31.5 cm above MWL corresponding to their nominal condition is presented for comparison.

470 Figure 6 depicts volume rather than number concentrations. Comparing  
 471 with Fig. 4 reveals that while large droplets are less numerous, they represent  
 472 the larger part of the volume (or mass). Presenting the data as volume con-  
 473 centrations allows us to better detail the differences between the four types of  
 474 wave forcing. Visual comparison of the four curves reveals that wind forcing  
 475 alone results in the lowest concentrations (dashed curve) and a distribution  
 476 that strongly decreases for  $r > 20 \mu\text{m}$ . The three wavemaker forcings all re-

477 sult in higher volume concentrations than the wind forcing; the order long,  
478 short and intermediate for increasing concentration corresponds well with the  
479 increase in whitecap  $W(\%)$  and wave slope variance  $\langle S^2 \rangle$  for these three  
480 types of forcing (cf. Table 1), and hence, the production of droplets.

481 A literature survey for comparison laboratory data identified the SPAN-  
482 DEX dataset (Fairall et al., 2009) as the most suitable. These authors also  
483 used a wind-wave interaction tunnel with freshwater and a mechanical wave  
484 paddle for the wave forcings. The triangles in Figure 6 (connected by the thin  
485 dashed line) represent the volume concentrations reported by Fairall et al.  
486 (2009), obtained at 31.5 cm height for 16.7 m s<sup>-1</sup> wind speed (corresponding  
487 to their nominal forcing) measured at 40 cm above MWL (roughly equivalent  
488 to 28 m s<sup>-1</sup> at  $U_{10}$ ), 1.36 Hz peak wave frequency (same as our short wave  
489 forcing), and friction velocity  $u_* = 1.44$  m s<sup>-1</sup>. Though the wind speed and  
490 friction velocity were 50 % greater than for the present study, the SPANDEX  
491 volume concentrations are of the same order of magnitude as our wave-forced  
492 data (cf. Fig. 6). This unexpected overlap may be explained by experimental  
493 differences such as tunnel dimensions (more than three times the height be-  
494 tween the water surface and the tunnel ceiling during MATE2019 compared  
495 with SPANDEX) leading to possibly different boundary effects. Another ex-  
496 planation maybe also be related to the different wave amplitudes between  
497 both experiments ( $\approx 5$  times greater in the present study) resulting in shorter  
498 distances between the probes and the wave crests in the case of MATE2019  
499 despite comparable sample heights in the present comparison.

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 500 4.2 Scaling parameters for the sea surface sea spray generation

501 In the previous section we have demonstrated that the sea spray concentra-  
 502 tion depends on properties of both the air flow and the sea state. To bridge  
 503 these two domains, we will scale the sea spray surface generation using non-  
 504 dimensional numbers representing the physical processes on the wind and wave  
 505 side of the wave-wind boundary. The non-dimensional numbers determined  
 506 from the set of parameters  $(u_*, \omega_p, c_p, H, \lambda, X, g, \rho_a, \rho_w, \nu_a, \nu_w, \Gamma)$ , where  
 507  $u_*$  is the friction velocity,  $\omega_p$  is the wave peak angular frequency,  $c_p$  is the  
 508 wave phase speed,  $H$  is the wave height,  $\lambda$  is the wavelength,  $X$  is the fetch  
 509 length,  $g$  is gravitational acceleration,  $\rho_a$  and  $\rho_w$  are air and water densities  
 510 respectively. Furthermore,  $\nu_a$  and  $\nu_w$  are air and water kinematic viscosities,  
 511 respectively, and  $\Gamma$  is the water surface tension.

512 As a first step, we focus our attention on the air kinematic viscosity. Sev-  
 513 eral studies have indicated that water droplets and water vapor can alter the  
 514 air viscosity, but the effect on the momentum flux is thought to be small,  
 515 less than 3% (Fairall et al., 2009). We therefore consider the non-dimensional  
 516 groups  $\nu_a/\nu_w$  and  $\rho_a/\rho_w$  to be constants. We also expect the surface tension  
 517  $\Gamma$  to be constant and remain negligible due to the relatively long wavelengths  
 518 considered here (strong majority of gravity waves). Then, we can express the  
 519 sea spray aerosol generation flux as a function of the non-dimensional numbers  
 520 that are the inverse wave age ( $= u_*/c_p$ ), the windsea Reynolds number  $R_B$   
 521 ( $= u_*^2/(\nu_a\omega_p)$ , determined from both wind ( $u_*$ ) and peak wave ( $\omega_p$ ) charac-  
 522 teristics), and the wave steepness. In continuity with the previous sections,  
 523 the wave slope variance  $\langle S^2 \rangle$  is used instead of the wave steepness ( $H/2\lambda$ ).  
 524 The inverse wave age is preferred to the wave age out of physical coherence, as  
 525 the former is expected to increase for increasing  $R_B$  and  $\langle S^2 \rangle$ . We create

526 a fourth scaling parameter by combining the inverse wave age, the windsea  
 527 Reynolds number and the wave slope variance:

$$528 \quad P_S = R_B \langle S^2 \rangle \frac{c_p}{u_*}^{-1}$$

529 which, considering the deep water dispersion relation  $c_p = \frac{g}{2\pi}T$  ( $T$  the wave  
 530 period), reduces to:

$$P_S = \frac{u_*^3}{\nu_a g} \langle S^2 \rangle \quad (5)$$

531 where  $P_S$ , adding to the  $P$  threshold for the activation of droplet and bub-  
 532 ble formation at the water surface (Newell and Zakharov, 1992; Fairall et al.,  
 533 2009), describes the wave slope variance-modulated wind energy input to the  
 534 waves. This number combines two experimental quantities,  $\langle S^2 \rangle$  provided  
 535 by the wave sensor array, and  $u_*^3$  provided by the wind speed vertical profiles,  
 536 providing information on the wind-dependent sea state and wind-induced dis-  
 537 sipation, respectively. This allows to study air-sea interaction processes from  
 538 a multiscale point of view, assuming that the smaller and larger scale wind  
 539 and wave components significantly contribute to boundary layer mechanisms  
 540 responsible for sea spray generation.

#### 541 4.3 Scaling of the sea spray generation

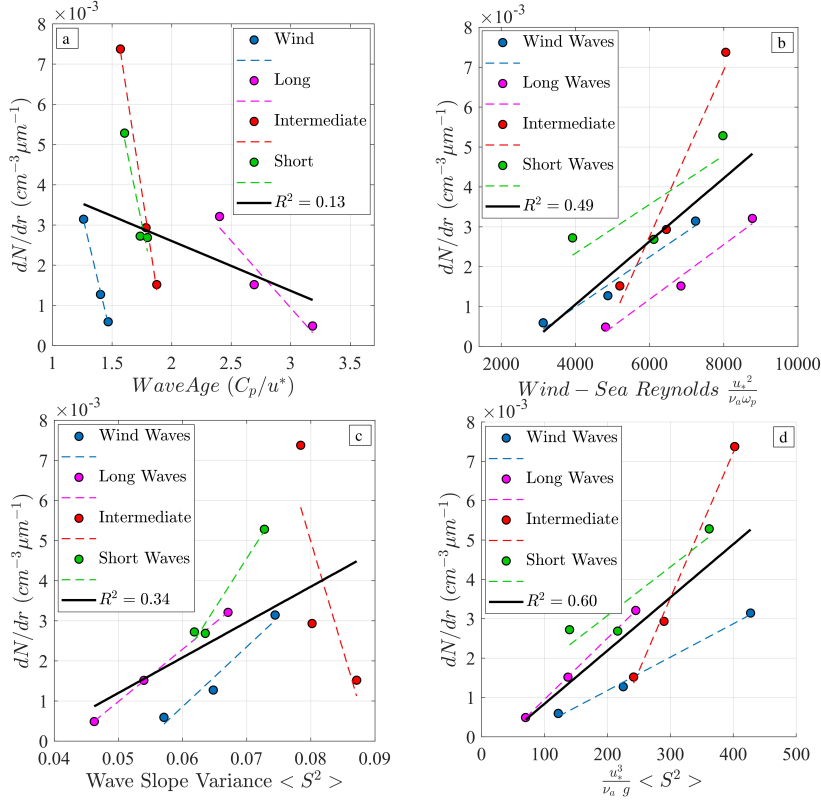
542 This section discusses the scaling of the sea spray by the four scaling pa-  
 543 rameters introduced above. For this, we use the concentrations measured 35  
 544 cm above the MWL, i.e., the lowest height of our aerosol samplers, assumed  
 545 the most representative of the generation flux and the least affected by size-  
 546 dependent vertical dispersion effects similar to Wu (1993) and Andreas (1998).  
 547 The scaling relationships are presented for droplet radii of 7.5  $\mu\text{m}$  (Fig. 7) and

25  $\mu\text{m}$  (Fig. 8), which are considered representative of the behaviour of the droplets in the 7-20  $\mu\text{m}$  and the 20-35  $\mu\text{m}$  size range, respectively. In this manner, we provide separate scaling for the sea spray predominantly produced by bubble bursting (7-20  $\mu\text{m}$ ) and surface-tearing (20-35  $\mu\text{m}$ ) mechanisms.

Figures 7a and 8a report the scaling with wave age. The presented data correspond to the wind-wave (blue dots), long wave (pink dots), intermediate wave (red dots) and short wave (green dots) forcing at 16, 18 and 20  $\text{m s}^{-1}$   $U_{10}$  wind speeds. Data of the same type of forcing are fitted to a linear function, denoted by a dashed line of the same colour. The black solid line represents a linear fit to all 12 datapoints, and the  $R^2$  value of the fit is given in the graphs ( $R^2 = 0.13$  for scaling with wave age for particles of 7.5  $\mu\text{m}$ ). Although the wave age seems to correctly scale concentrations for individual wave forcings, **no clear relationship is found between the measured concentrations and wave age when all data is included**. Therefore, we conclude that wave age is not well suited for the scaling of sea spray generation for both young wind-forced waves, and older swell-type waves.

Nevertheless, it is interesting to further discuss Figs. 7a and 8a. The overall decrease in near-surface concentrations for increasing wave age (Figs. 7a and 8a) corroborates observations made in the field, as less whitecap is produced by older swell-type waves or wind waves modulated by older waves in contrast with wind waves alone (Schwendeman and Thomson, 2015; Brumer et al., 2017). Both figures show that relatively high sea spray concentrations are found for the intermediate and short forcings, with corresponding wave age values of approximately 1.7. This can be expected considering the high values of  $W(\%)$  and  $\langle S^2 \rangle$  for these types of forcing (cf. Table 1). High droplet concentrations associated with young, steep and strongly breaking waves is typical of fetch-limited wave fields (Lafon et al., 2007). Specifically, for the

575 larger  $25 \mu\text{m}$  particles (cf. Fig. 8a) such a peak can be related to droplet  
 576 ejection microphysics as younger steeper waves induce airflow separation (Reul  
 577 et al., 2008), thought to play an important role in droplet ejection from the  
 578 wave crests (Mueller and Veron, 2009; Veron et al., 2012; Richter et al., 2019).



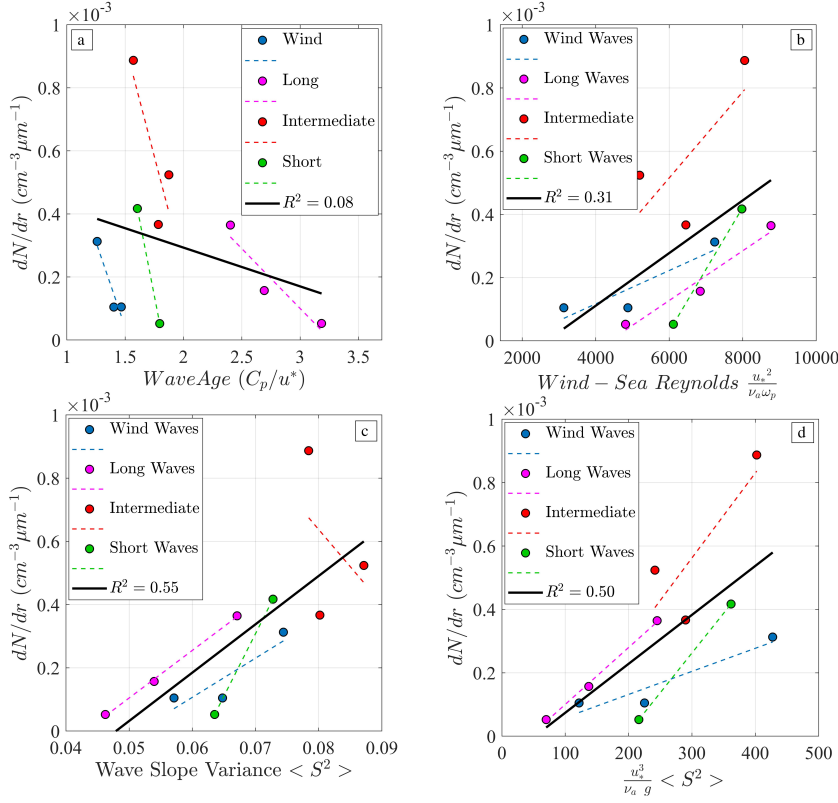
**Fig. 7** Scaling of  $dN/dr$  with (a) wave age, (b)  $R_B$ , (c)  $\langle S^2 \rangle$  and (d)  $P_S = \frac{u_*^3}{\nu_a g} \langle S^2 \rangle$  for particles of radius  $7.5 \mu\text{m}$ , 35 cm above MWL. Individual types of wave forcing are represented in color, with  $U_{10}$  wind speeds ranging from 16 to 20  $\text{m s}^{-1}$ . **Solid black lines in all panels represent linear fits to all 12 datapoints.**

579 We now turn our attention to Figs. 7b and 8b, which present the windsea  
 580 Reynolds number  $R_B$  as a scaling parameter for the sea spray concentration.  
 581 The Reynolds number has proven to be strongly related to whitecap coverage  
 582 and wave breaking (cf. Sect. 3.2) as well as sea spray concentrations for wind  
 583 waves especially (Iida et al., 1992; Toba et al., 2006; Troitskaya et al., 2018).  
 584 This seems confirmed by the overall (solid black line) trend line in Figures

585 7b and 8b, which is relatively close to the data **corresponding to** wind waves.  
 586 The data show less spread than for scaling with wave age, and most types  
 587 of forcing follow the overall trend, despite systematic differences in droplet  
 588 concentrations. The overall fit quality for the 7.5  $\mu\text{m}$  particles amounts to  $R^2$   
 589  $= 0.49$  and similar values were noted for other radii in the 7-20  $\mu\text{m}$  range.  
 590 The windsea Reynolds number  $R_B$  does not perform as well over the 20-  
 591 35  $\mu\text{m}$  radius range, as demonstrated by  $R^2 = 0.31$  for the 25  $\mu\text{m}$  particles  
 592 (Fig. 8b). The better performance for smaller radii can find explanation in  
 593 the information contained in  $R_B$  about the flow velocity and more specifically  
 594 the turbulence intensity at the wind-wave boundary layer. We expect this to  
 595 be key for the smaller droplets that are more subject to turbulent diffusion  
 596 and less affected by gravitational settling than the larger spume droplets (e.g.  
 597 Andreas et al. (2010)).

598 The scaling of spray concentrations by the wave slope variance  $\langle S^2 \rangle$   
 599 is presented in Figs. 7c and 8c. The most striking feature of these figures  
 600 is the behaviour for the intermediate wave forcing, which yields decreasing  
 601 aerosol concentrations with  $\langle S^2 \rangle$ , whereas the other types of forcing all  
 602 yield increasing concentrations. This behaviour can be traced back to the ear-  
 603 lier observation (Sect. 3.1) that the intermediate forcing yields a decrease in  
 604 significant wave height with increasing  $\langle S^2 \rangle$  (and a less clear increase of  
 605  $W(\%)$ , which was attributed to (exceptionally) efficient wave energy dissipa-  
 606 tion that for the other three types of forcing. This contrasting behaviour for  
 607 the intermediate forcing causes the overall fit quality for smaller particles to  
 608 be less than when scaling with  $R_B$  ( $R^2$  of 0.34 versus 0.49). Over the 20-35  $\mu\text{m}$   
 609 range however, the wave slope variance offers the best scaling performance for  
 610 all four scaling parameters ( $R^2 = 0.55$ ). The wave slope variance thus appears

611 well adapted for the scaling of near-surface spume droplet concentrations (Fig.  
612 8c).



**Fig. 8** Scaling of  $dN/dr$  with (a) wave age, (b)  $R_B$ , (c)  $\langle S^2 \rangle$  and (d)  $P_S = \frac{u_*^3}{\nu_a g} \cdot \langle S^2 \rangle$  for particles of radius  $25 \mu\text{m}$ ,  $35 \text{ cm}$  above MWL. Individual wave forcings are denoted by colours, for  $U_{10}$  wind speeds ranging from  $16$  to  $20 \text{ m s}^{-1}$ . **Solid black lines in all panels represent linear fits to all 12 datapoints.**

613 Finally, we scale near-surface concentrations as a function of the combina-  
614 tion of non-dimensional numbers  $\frac{u_*^3}{\nu_a g} \cdot \langle S^2 \rangle$ . Results show that the combined  
615 number  $P_S$  scales the particle concentrations better than either the wave age  
616 or the windsea Reynolds number. In comparison with the wave slope variance  
617 the combined number performance is better over the  $7\text{-}20 \mu\text{m}$  range ( $R^2 = 0.60$   
618 versus  $R^2 = 0.34$  for  $r = 7.5 \mu\text{m}$ ), but about equal for particles greater than  
619  $20 \mu\text{m}$  ( $R^2 = 0.5$  versus  $R^2 = 0.55$  for  $r = 25 \mu\text{m}$ ). However, the combined  
620 number results in similar trends for all four types of wave forcing, including



621 the intermediate forcing (compare panels c and d). This suggests that the  
 622 combined number is capable of scaling sea spray generation for a wider range  
 623 of environmental conditions than  $\langle S^2 \rangle$  alone. We therefore conclude that  
 624 the combined number is well suited for the simultaneous scaling of breaking  
 625 wave (bubble bursting) and surface tearing mechanisms.

#### 626 4.4 Combined scaling parameters

627 Our fourth scaling parameter is similar to the non-dimensional numbers sug-  
 628 gested by Brumer et al. (2017) and Lenain and Melville (2017). The latter  
 629 authors combine the wave steepness, the wave age and a Reynolds number  
 630 depending on the significant wave height  $H_s$  ( $R_H = \frac{H_s u_*}{\nu}$ ). A similar number  
 631 was also used by Brumer et al. (2017) for the scaling of whitecap coverage.  
 632 Preliminary laboratory studies on the scaling of sea spray concentrations in  
 633 the 7-35  $\mu\text{m}$  radius range by the number proposed by Lenain and Melville  
 634 (2017) suggest that the scaling does not significantly change when substitut-  
 635 ing  $R_H$  with  $R_B$ . However, significantly better scaling is achieved when the  
 636 wave steepness ( $H/2\lambda$ ) is replaced with  $\langle S^2 \rangle$ .

637 As mentioned in the introduction,  $u_*^3$  has proven to be a worthy candidate  
 638 to scale wave energy dissipation and input from the wind field necessary for  
 639 bubble and droplet production (Newell and Zakharov, 1992; Zhao and Toba,  
 640 2001; Zhao et al., 2006; Andreas, 1998, 2002; Fairall et al., 2009). Though not  
 641 detailed in the present scaling analysis, a preliminary study allowed to evaluate  
 642 the scaling performance of  $u_*^3$  for near-surface concentrations. Results reveal  
 643 similar performance to the combined number with good scaling results over the  
 644 7-20  $\mu\text{m}$  range ( $R^2 = 0.56$  for  $r = 7.5 \mu\text{m}$ ) with less satisfactory performance  
 645 for larger droplets ( $R^2 = 0.23$  for  $r = 25 \mu\text{m}$ ). Associating scaling performances

646 of  $u_*^3$  (best for  $r < 20 \mu\text{m}$ ) with  $\langle S^2 \rangle$  (best for  $r > 20 \mu\text{m}$ ) in the combined  
 647 number (Eq. 5) allows good scaling of sea spray aerosol generation over the 7-35  
 648  $\mu\text{m}$  range, suggesting that the combination of both allows to scale production  
 649 in a wider range of conditions than  $\langle S^2 \rangle$  alone.

#### 650 4.5 The laboratory generation flux

651 In this section, we will consider a suitable scaling parameter for the spray gen-  
 652 eration flux ( $dF/dr$ ). We will derive this flux from the sea spray concentration  
 653 profiles measured in the laboratory (cf. Fig. 5), at each resolved particle size.  
 654 We only use the data obtained for the higher wind speeds (16, 18 and 20 m  
 655  $\text{s}^{-1}$ ), but we include all four types of wave forcing. As a first step we approach  
 656 the vertical concentration profiles with a function depending on the natural  
 657 logarithm of measurement height (cf. Sect. 4.1 and Fig. 5):

$$N^r(z) = N_*^r \ln(z) + C^r \quad (6)$$

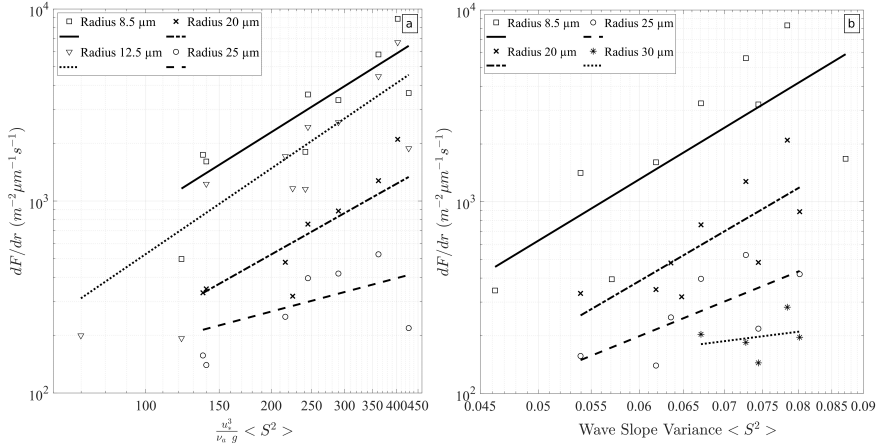
658 where radius-dependent the aerosol concentration at the surface  $N_*^r$  ( $\text{m}^{-3}$ ) is  
 659 determined by extending the measured aerosol concentration profiles to MWL.  
 660  $r$  denotes the dependence on the droplet radius. The sea spray flux  $dF/dr$  ( $\text{m}^{-2}$   
 661  $\text{s}^{-1}$ ) is subsequently obtained by multiplication with the friction velocity  $u_*$ :

$$\frac{dF}{dr} = N_*^r u_* \quad (7)$$

662 We evaluate  $N_*^r$  for each radius (bin) greater than 7  $\mu\text{m}$  measured by our  
 663 aerosol probes. This method is commonly used for the estimation of the sea  
 664 spray flux in the field (Petelski et al., 2014), as well as for numerical LES (Large  
 665 Eddy Simulation) data (Richter et al., 2019). However, the use of a logarithmic

666 function to describe the concentration gradient is debated, and other authors  
667 have preferred a power law (e.g., Fairall et al. (2009); Ortiz-Suslow et al. (2016)).  
668 Rather than entering this debate, we abide by the adequate fit of logarithmic  
669 functions to our data (cf. Fig. 5).

670 As discussed in the previous section, both the wave slope variance and  
671 the combined number are good candidates to scale droplet generation. The  
672 combined number is found suitable over the entire study radius range, and  
673 the wave slope variance for spume droplets. Thus, Figure 9 shows the size-  
674 dependent sea spray generation flux  $dF/dr$  (evaluated using Eq. 7) as function  
675 of both the combined number (panel (a)) and the wave slope variance (panel  
676 (b)), for various representative droplet radii between 8.5 and 30  $\mu\text{m}$ . Individual  
677 data for a specific radius was fitted to a power law presented as lines in Fig.  
678 9. As expected, the figure demonstrates that the combined number and the  
679 wave slope variance are also good scaling parameters for the generation flux.  
680 In accordance with the positive slopes in Figs. 7 and 8 (panels c and d),  
681 increasing values of combined number or wave slope variance lead to a stronger  
682 flux. Although it is difficult to conclude visually from Fig. 9, a comparison of  
683 panels a and b reveals the better scaling performance of the combined number  
684 for smaller radii, as reflected in the quality of the fit for 8.5 particles  $\mu\text{m}$ .  
685 Finally, and interesting feature in both Figs. 9a and 9b is the increase in the  
686 curve slopes for increasing radius as function of the scaling parameters. The  
687 lower slopes are found for the larger droplets. These slopes, seem to converge  
688 towards a maximum slope in the jet droplet range as the curves become almost  
689 parallel for  $r < 15 \mu\text{m}$ . This suggests uneven radius-dependent flux strength  
690 variations over the sea spray spectrum according to the forcing.



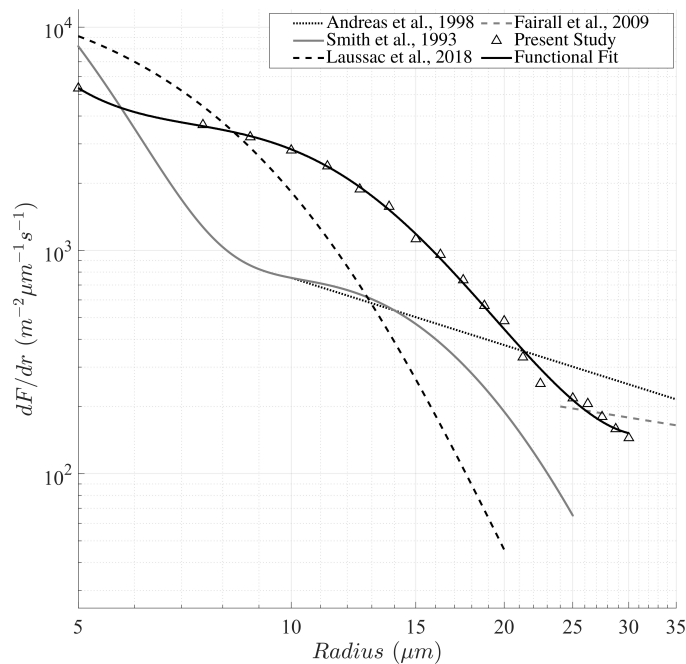
**Fig. 9** The laboratory  $dF/dr$  sea spray flux as a function of (a) the combined number  $\frac{u_*^3}{\nu_a g} < S^2 >$  and (b) the wave slope variance  $< S^2 >$ . Lines denote power law fits to data for individual radii.

#### 691 4.6 Comparison with other studies

692 This section presents a comparison of our generation flux to others reported  
 693 in the literature. Since most existing fluxes rely on data acquired in the field,  
 694 we must ascertain that our laboratory data can be compared to the field.  
 695 Comparing air-sea interaction processes and resulting sea spray generation in  
 696 laboratory conditions to the field is not a straightforward task, due to the  
 697 greater complexity of the natural environment. It is important that physical  
 698 mechanisms at play in the laboratory, as well as the resulting sea spray gen-  
 699 eration fluxes, be similar to those observed in the field. The former question  
 700 has already been discussed in this paper; the wind flow (Sect. 2.4), the wave  
 701 field (Sect. 3.1), and the whitecap coverage (Sect. 3.2) were all found to fol-  
 702 low the same physics as in the field. As for the sea spray concentrations, Sect.  
 703 4.1 demonstrated the expected behaviour with increasing concentrations for in-  
 704 creasing wind speed, a dependence on wave state, as well as a near-logarithmic

705 concentration profile later used to calculate the laboratory generation fluxes  
 706 (Eqs. 6 and 7).

707 The above reasoning leads us to compare our laboratory sea spray gen-  
 708 eration flux to four (field and laboratory) flux formulations published in the  
 709 literature. For this comparison we have evaluated our generation flux for the  
 710 laboratory pure wind forcing for 18 probe size bins over the 5-30  $\mu\text{m}$  radius  
 711 range, with friction velocity  $u_* = 1.05 \text{ m s}^{-1}$  and wind speed  $U_{10} = 20 \text{ m s}^{-1}$  (cf.  
 712 Table 1). The flux is depicted in Fig. 10 as the black solid line. The triangles  
 713 denote the radii for which the  $dF/dr$  was evaluated, to which a polynomial  
 714 function is fitted to facilitate comparison with the other generation spectra.  
 715 Figure 10 also represents fluxes from the literature; Smith et al. (1993) denoted  
 716 by S93 and the grey solid line, Andreas (1998) denoted by A98 and the dotted  
 717 line, Laussac et al. (2018) denoted by L18 and the dashed line, and Fairall  
 718 et al. (2009) denoted by F09 and the grey dashed line. The latter flux was  
 719 obtained from laboratory data, the other three from field data. The Andreas  
 720 (1998) and Smith et al. (1993) flux models solely depend on wind speed  $U_{10}$   
 721 and are evaluated here for  $U_{10} = 20 \text{ m s}^{-1}$ . The F09 flux is estimated for  $u_* =$   
 722  $1.35 \text{ m s}^{-1}$ , and is only valid for  $r \geq 24 \mu\text{m}$ . Finally, L18 flux strength depends  
 723 on the whitecap coverage, in turn defined in terms of the wave age, and has a  
 724 0.5-20  $\mu\text{m}$  radius validity range. The closest match with laboratory conditions  
 725 (wave age equal to 1.25) was obtained by estimating the L18 flux for a wave  
 726 age value of 3.9 using a fetch model adapted to the north-west Mediterranean  
 727 (Lafon et al., 2004), corresponding with a MIRAMER campaign sample point  
 728 located at 12 km fetch (Demoisson et al., 2013; Laussac et al., 2018) used to  
 729 parameterise the L18 SSGF.



**Fig. 10** Vertical sea spray concentration fluxes at  $U_{10} = 20 \text{ m s}^{-1}$ .

730 Figure 10 shows that the various flux estimates have a spread of an order  
 731 of magnitude. While this may seem significant, the spread is quite typical as  
 732 evidenced by reviews of the various source functions reported over the years  
 733 (Andreas, 1998; Lewis et al., 2004; Veron, 2015). Our flux function has the same  
 734 strength as S93, A98 and L18 for particles of  $5 \mu\text{m}$ , but suggests a stronger  
 735 flux than these other functions as radius increases. Specifically, our flux curve  
 736 includes a shoulder (or peak) around  $10\text{-}15 \mu\text{m}$ , also seen in S93, which we  
 737 attribute to a peak in jet droplet production. A similar shoulder is observed  
 738 in the aerosol size distributions measured at  $55 \text{ cm}$  height above the surface  
 739 (cf. Fig. 4). Moving forward to radii in excess of  $15 \mu\text{m}$ , the typical domain  
 740 of spume droplets, the strength of L18 and S93 generation functions decrease  
 741 rapidly. In both cases, this decrease was attributed to an underestimation of

742 spume droplet production, for L18 because the experiment was hampered by  
743 the near-absence of high wind speed events, and because for L18 and S93 the  
744 height of the measurement towers prevented efficient dispersion of the spume  
745 droplet location. In each case this can result in poor count statistics, and the  
746 larger droplet concentrations are often discarded as done for  $r > 35 \mu\text{m}$  in  
747 the present study. Examples of efforts to compensate such effects include the  
748 reformulation of S93 in A98, where the generation flux was corrected over the  
749 spume droplet range (Andreas, 1998). For the larger radii ( $r > 25 \mu\text{m}$ ), our flux  
750 function yields equal strength as F09. Because these largest particles reflect  
751 the limits of our PMS probes it is difficult to conclude, but both our flux curve  
752 and F09 seem to indicate that the decrease in strength becomes less strong  
753 with radius. This may point to the presence of another shoulder around  $30$   
754  $\mu\text{m}$ , reflecting a peak in spume droplet generation.

## 755 5 Formulation of the new sea spray generation function

756 The previous sections demonstrated that the laboratory generation flux de-  
757 pends on both wind and wave characteristics, with different responses for  
758 individual radii. The dependence on radius is traditionally accounted for by  
759 parameterising the SSGF with one or more modes centered around modal radii  
760 (Monahan et al., 1986; Demoisson et al., 2013; Ovadnevaite et al., 2014; Laus-  
761 sac et al., 2018). These modes can be represented with normal, log-normal,  
762 or other distributions that are commonly used to reproduce measured aerosol  
763 distribution characteristics. Modes are often associated with specific aerosol  
764 processes, such as the generation of jet or spume droplets. **We adopt this**  
765 **methodology by introducing two modes at 7 and 25  $\mu\text{m}$  radius, which we**  
766 **consider representative for jet and spume droplets, respectively. The choice**

of these centre radii was inspired by the shapes of the flux and concentration curves shown in Figs. 4 and 10, respectively. As mentioned in Sect. 2.5, we have excluded droplets with radii less than  $7 \mu\text{m}$  radius for our analysis, but these droplets are evidently present in the tunnel (Fig. 4). On the basis of the shape of the aerosol size distribution shown in Fig. 4, we add a third mode centered around  $2.5 \mu\text{m}$ . Taking  $3 \mu\text{m}$  as the lower limit, the flux strength of the L18 SSGF is used over  $3\text{-}7 \mu\text{m}$ . A smooth transition between the field-determined L18 and the laboratory data is attained by seeking a best fit between L18 flux distributions over the  $3\text{-}7 \mu\text{m}$  range, and the laboratory flux distributions. The  $7 \mu\text{m}$  radius marks the transition between both fluxes (cf. Fig. 11). To achieve this combination of flux functions, the whitecap coverage used in L18 (originally calculated from the wave age) (Laussac et al., 2018) is recalculated here as a function of present study scaling parameters (cf. Sect. 4.2). In the following, the L18  $W(\%)$  is estimated from  $\langle S^2 \rangle$  using Eq. 4. Section 4.3 revealed that the flux can be successfully scaled by a combined number  $P_S$ , but that the  $\langle S^2 \rangle$  also yields acceptable results, especially for larger particles. Therefore, we formulate two SSGFs in terms of three normally distributed modes, valid for droplets between  $3\text{-}35 \mu\text{m}$  radius and wind speeds  $U_{10}$  ranging from  $12\text{-}20 \text{ m s}^{-1}$ :

$$\frac{dF_X}{dr_{80}} = \sum_{i=1}^n \frac{F_i(X)\tau^{-1}}{\sigma_i\sqrt{2\pi}} \exp\left(-\frac{1}{2}\left(\frac{r_{80} - \mu_i}{\sigma_i}\right)^2\right) \quad (8)$$

where  $X$  is the scaling parameter ( $P_S$  or  $\langle S^2 \rangle$ ),  $r_{80}$  is the particle radius at  $RH = 80\%$ ,  $\frac{dF_X}{dr_{80}}$  is the size dependent SSGF determined from environmental parameters contained in  $F_i(X)$ , and  $\tau$  is the whitecap decay rate.  $\mu_i$  and  $\sigma_i$  are the mean radius and standard deviation of each of the three modes. The SSGFs can be given in terms of  $r_{80}$ , because the underlying laboratory data was obtained for  $80\%$  humidity (cf. the discussion in Sect. 2.5). In formulating the SSGFs, we found best results were obtained using normal modes, whereas other



793 authors have preferred lognormal modes (e.g. Ovadnevaite et al. (2014)). Pos-  
 794 sibly, this signals that normal modes are more suitable for the larger droplets  
 795 studied here. Numerical values for Eq. 8 are specified in Table 2. In addition,  
 796 the whitecap decay rate was given a value of 3.53 (Laussac et al., 2018). De-  
 797 spite a focus on the 16-20 m s<sup>-1</sup> range in Sect. 4, we are confident that the  
 798 SSGFs are valid over the 12-20 m s<sup>-1</sup> range as a result of the strong relation-  
 799 ship between numbers  $\langle S^2 \rangle$  and  $P_S$  and the physical mechanisms known to  
 800 drive production (cf. sects. 2.4 and 3.2) for these wind speeds.

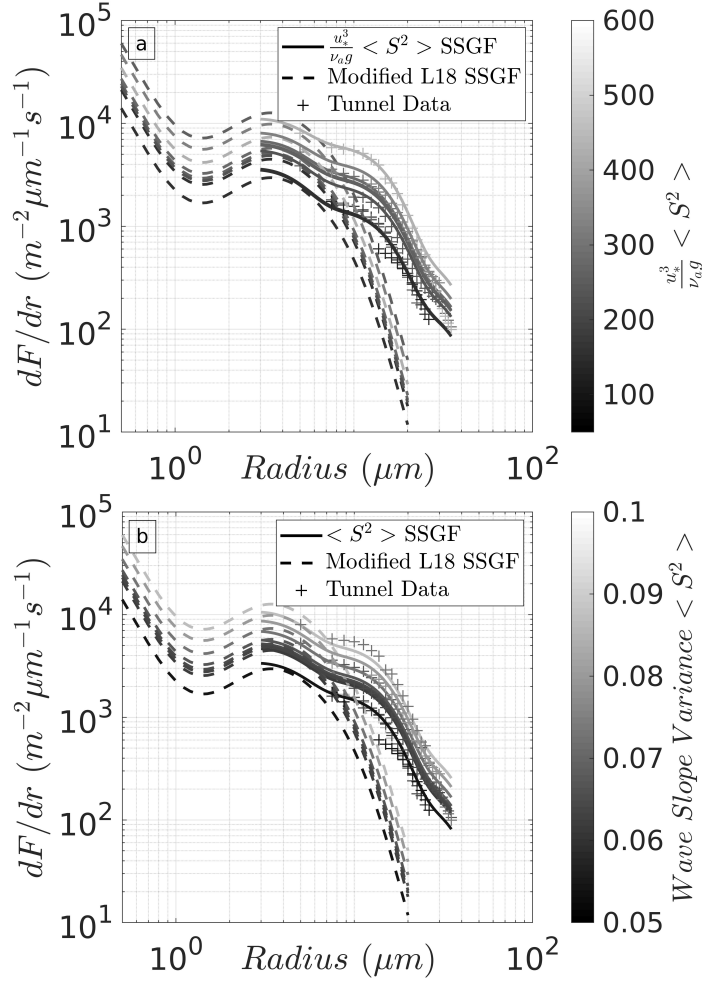
801 Table 2 shows that flux amplitudes are expressed as power laws of  $\langle S^2 \rangle$   
 802 and  $P_S$ , as suggested with the apparent power law behaviour between scaling  
 803 parameters and fluxes presented in Fig. 9.

**Table 2** Parameters for the two SSGFs.

| i | $\sigma_i$ | $\mu_i$ | $F_i(\frac{u_*^3}{\nu_{ag}} \langle S^2 \rangle)$                      | $F_i(\langle S^2 \rangle)$                      |
|---|------------|---------|--|---|
| 1 | 2.1        | 2.5     | $4.37 \times 10^2 (\frac{u_*^3}{\nu_{ag}} \langle S^2 \rangle)^{0.92}$ | $4.94 \times 10^7 (\langle S^2 \rangle)^{2.45}$ |
| 2 | 7          | 7       | $6.84 \times 10^1 (\frac{u_*^3}{\nu_{ag}} \langle S^2 \rangle)^{1.41}$ | $7.88 \times 10^7 (\langle S^2 \rangle)^{2.3}$  |
| 3 | 12         | 25      | $4.75 \times 10^1 (\frac{u_*^3}{\nu_{ag}} \langle S^2 \rangle)^{1.11}$ | $1.3 \times 10^7 (\langle S^2 \rangle)^{2.39}$  |

804 The two new SSGFs are presented in Figure 11 as two series of solid lines,  
 805 where panels a and b denote results for the SSGF in terms of the combined  
 806 number  $P_S$  and  $\langle S \rangle$  respectively. These lines are colour-coded according to  
 807 the values of the scaling parameters; the colour scale is indicated on the right  
 808 of the individual panels. The range of values was chosen to correspond with  
 809 the laboratory configurations for all types of wind speeds between 16 and 20 m  
 810 s<sup>-1</sup>. The experimental flux data obtained in these configurations is visualised  
 811 by the color-coded plus signs in Fig. 11. Finally, Figure 11 also presents the  
 812 flux spectra prescribed by the L18 SSGF (Laussac et al., 2018) over the 3-  
 813 7  $\mu\text{m}$  range, which, as mentioned above, was modified so that the whitecap  
 814 coverage  $W(\%)$  scaling parameter (originally calculated from the wave age),

815 was recalculated to depend on either  $P_S$  or  $\langle S^2 \rangle$ . The recalculated L18 flux  
 816 spectra are represented by the color-coded dashed lines.



**Fig. 11** The altered L18 SSGF (dashed lines) and the (a) combined number  $P_S$  and (b) wave slope variance  $\langle S^2 \rangle$  SSGFs. Line colors depend on respective non-dimensional (a)  $P_S$  and (b)  $\langle S^2 \rangle$  values.

817 Figure 11 demonstrates that the two SSGFs reproduce the experimental  
 818 flux data obtained in the tunnel quite well, although differences of up to a  
 819 factor 2 remain. In view of the already noted significant spread in individual  
 820 flux functions reported in the literature (Tsyro et al., 2011; Chen et al., 2016;  
 821 Neumann et al., 2016), we consider this performance adequate. The figure

822 also demonstrates that the two modes at 7 and 25  $\mu\text{m}$  adequately reproduce  
823 the shape of the tunnel spectra, and this better than flux functions with less  
824 modes in the large particle size domain (e.g., S93 and L18). Finally, Fig. 11  
825 also shows that both SSGFs connect reasonably well to L18. While this was  
826 expected with a 2.5  $\mu\text{m}$  mode inspired by L18 flux strengths, this result offers  
827 a perspective for a future coupling between the new SSGFs and L18, thereby  
828 extending a universal function to the 0.1-35  $\mu\text{m}$  range. These results also offer  
829 a perspective for future improvements on the combination of laboratory and  
830 field sea spray measurements.

## 831 **6 Discussion and conclusion**

### 832 **6.1 Generation mechanisms**

833 The aim of this study is to investigate the sea spray generation processes for  
834 different wind and wave combinations. We performed our studies in a wave-  
835 wind interaction laboratory and thus, our first task was to demonstrate that  
836 important generation mechanisms such as wave breaking (cf. Sect. 3.2) and  
837 airflow characteristics (cf. Sect. 2.4) are similar in the laboratory and in the  
838 field. Sea spray concentrations over the 7-35  $\mu\text{m}$  radius range are found to  
839 depend on both wind speed and the sea state, and are found to decrease near-  
840 logarithmically for increasing distance from the water surface. The concentra-  
841 tion spectra (Fig. 4) exhibit peaks at approximately 2.5 and 7  $\mu\text{m}$  similar to  
842 observations in the field (Laussac et al., 2018), attributed to regimes of film  
843 and jet droplet generation predominance, respectively. A third peak was  
844 identified from the volume concentration spectra (Fig. 6) and corresponds to  
845 the spume droplet size range. This peak was absent for wind speeds under 12 m

846  $s^{-1}$  (Sect. 4.1), in accordance with the threshold for spume droplet production  
847 (Monahan et al., 1986; Andreas et al., 2010). The larger droplet concentrations  
848 are found in conditions with high values for whitecap coverage and wave slope  
849 variance. These conditions correspond to intermediate and short wave forcing  
850 in the laboratory (cf. Table 1). In contrast, long wave forcing or forcing by  
851 wind alone result in lower concentrations.

## 852 6.2 Scaling sea spray generation with $\langle S^2 \rangle$

853 The sea spray generation flux is calculated assuming logarithmic concentration  
854 profiles (Sect. 4.5), motivated by the near-logarithmic profiles obtained in the  
855 laboratory (Fig. 5). The sea spray generation is scaled against four parameters  
856 that reflect the wind-wave conditions: wave age, windsea Reynolds number  $R_B$ ,  
857 wave slope variance  $\langle S^2 \rangle$ , and a combined number  $\frac{u_*^3}{\nu_a g} \langle S^2 \rangle$ , denoted  
858  $P_S$ , describing the wave slope variance-modulated wind energy input to the  
859 waves. Whilst the wave age is a useful heuristic tool to estimate wave-wind  
860 equilibria, our results indicate that it does not allow satisfactory scaling of  
861 sea spray generation for different wind and wave combinations (cf. Figs. 7 and  
862 8). The windsea Reynolds number  $R_B$ , also combining wind and peak wave  
863 characteristics, provides good results for smaller droplets (7-20  $\mu\text{m}$  range),  
864 comparable to the performance for  $\langle S^2 \rangle$ . However,  $\langle S^2 \rangle$  significantly  
865 outperforms  $R_B$  for spume droplets with radii greater than 20  $\mu\text{m}$  (cf. Fig. 8).  
866 This difference may be expected because  $R_B$  only includes information about  
867 peak wave statistics, whereas  $\langle S^2 \rangle$  is related to turbulence strength at the  
868 air-sea interface and wave breaking phenomena on a range of spatial scales.  
869 This is in agreement with an increasing number of microphysical studies that  
870 relate wave slope to turbulent events such as airflow separation and wind shear,

871 which are thought to drive the surface tearing of spume droplets at the wave  
 872 crests (Banner and Melville, 1976; Kawai, 1981; Reul et al., 2008; Mueller and  
 873 Veron, 2014; Buckley and Veron, 2019).

874 Of the four tested scaling parametersn the combined number  $P_S$  gives the  
 875 best overall performance: it is particularly well suited for the scaling of sea  
 876 spray generation for radii in the 7-20  $\mu\text{m}$  range, where bubble bursting gen-  
 877 eration dominates, but the larger spume droplets are also well scaled. The  
 878 spume droplet scaling performance reflects the presence of  $\langle S^2 \rangle$ , and the  
 879 scaling performance for smaller droplets may be attributed to  $u_*^3$  as a result  
 880 of its well documented relationship with wave energy dissipation. The con-  
 881 cept of combined scaling parameters has also been explored by other authors  
 882 (Brumer et al., 2017; Lenain and Melville, 2017) (cf. Sect. 4.4). A comparative  
 883 study (Sect. 4.3) shows that minor differences in the definition of the com-  
 884 bined number (e.g. by using a Reynolds number defined in terms of significant  
 885 wave height) do not impact on the performance, in contrast to the significant  
 886 difference obtained when replacing wave steepness with the multiscale wave  
 887 slope variance.

888 From a practical point of view, it is interesting to compare scaling perfor-  
 889 mance to the difficulty to measure the scaling parameters. Both wave age and  
 890 the windsea Reynolds number  $R_B$  require a wind parameter and relatively  
 891 common wave parameters. The wave age does not scale the flux properly for  
 892 the range of tested forcings, and  $R_B$  scaling performances are mainly limited  
 893 to the smaller jet droplet particles. The measurement difficulty increases when  
 894 considering the multiscale information (wave spectrum) required for  $\langle S^2 \rangle$ ,  
 895 and the combined number  $P_S$  adds further complexity by requiring the wind  
 896 friction velocity  $u_*$ . The use of approximate equations to estimate  $\langle S^2 \rangle$  from

897 more readily-accessible parameters (e.g. Eqs. 3 and 4) may thus be considered,  
 898 albeit with loss of scaling performance.

899 6.3  $\langle S^2 \rangle$  and traditional scaling parameters

900 As mentioned in the previous section, it may be difficult to measure  $\langle S^2 \rangle$   
 901 in the field. The use of the approximate equations to infer  $\langle S^2 \rangle$  from more  
 902 accessible parameters such as wind speed  $U_{10}$  (Eq. 3), or whitecap coverage  
 903  $W(\%)$  (Eq. 4) may then be considered, as the wave slope variance  $\langle S^2 \rangle$   
 904 has often been considered proportional to both the wind speed at  $U_{10}$  and the  
 905 friction velocity  $u_*$  (Cox and Munk, 1956; Lenain et al., 2019). In a similar  
 906 fashion, it is important to note that many authors obtain  $W(\%)$  from wind  
 907 speed Monahan and Muircheartaigh (1980). Though not explicitly presented  
 908 here,  $\frac{u_*^3}{\nu_a g} \langle S^2 \rangle$  and  $\langle S^2 \rangle$  (and to a lesser extent  $R_B$ ) outperform the  
 909 measured  $U_{10}$ ,  $u_*$  and  $W(\%)$  for the scaling of sea spray generation in the  
 910 tested laboratory configurations, especially for larger droplets.  $W(\%)$  presents  
 911  $R^2$  values ranging between 0.2 and 0.45 over the 7-35  $\mu\text{m}$  range, with best  
 912 results over the jet droplet range.  $U_{10}$  and  $u_*$  scaling performances are lower  
 913 than for  $W(\%)$  overall, except for the smaller radii of the jet droplet range  
 914 where  $U_{10}$  and  $u_*$  show good correlation with sea spray generation, with  $R^2$   
 915 values reaching maxima of 0.49 and 0.54 (for  $r = 7 \mu\text{m}$ ), respectively. These  
 916 better results over the jet droplet range are coherent with the performances  
 917 of the combined number  $P_S$  with the  $u_*^3$  term. Furthermore, this shows that  
 918  $\langle S^2 \rangle$  carries additional information (despite the good correlation between  
 919  $\langle S^2 \rangle$  and  $W(\%)$  - cf. Eq. 4) about the wave field relative to these three  
 920 parameters, such as the presumed role of the wave slope in the modulation of  
 921 the air flow and surface tearing mechanisms, as discussed in Sect 4.1.

922 Unlike  $\langle S^2 \rangle$ ,  $U_{10}$  and  $u_*$  are not found to be appropriate tools for the  
923 upscaling and extrapolation of sea spray generation and whitecap coverage  
924 from the laboratory to the field, similar to observations made by Toba et al.  
925 (2006) when comparing tunnel and field data. Although  $\langle S^2 \rangle$  outperforms  
926  $U_{10}$  and  $u_*$ , there are conditions in which the wind parameters scale the aerosol  
927 concentrations as successfully. This pertains to smaller droplets ( $r < 12.5 \mu\text{m}$ )  
928 and relatively low wind speeds of 8 and 12  $\text{m s}^{-1}$ , and a wave field that is forced  
929 by the wind. In these circumstances, sea spray consists almost exclusively of  
930 the bubble-mediated jet droplets produced from breaking waves under condi-  
931 tions of strong wave-wind equilibrium. Further analysis of our laboratory data  
932 reveals that the linear relation between  $U_{10}$  and  $\langle S^2 \rangle$  presented by Cox and  
933 Munk (1956) (Eq. 3) is best observed for the conditions corresponding to pure  
934 wind forcing. This is coherent with the good scaling performance of  $U_{10}$  and  $u_*$   
935 under these conditions, as mentioned above. However, notable deviations from  
936 this linear relationship are observed with other types of wave forcing, which we  
937 attribute to the sensitivity of  $\langle S^2 \rangle$  to wave-wind equilibria (quantified here  
938 with wave age). **The monochromatic properties of the longer wave conditions**  
939 **that experience less wind forcing, resulting in low wave energy at the higher**  
940 **frequencies of the wave spectrum, thereby affecting the value of  $\langle S^2 \rangle$ . In**  
941 **this respect, a future laboratory study with more realistic wave fields such as**  
942 **the JONSWAP spectra (Hasselmann et al., 1973) would be worthwhile.**

#### 943 6.4 Validation against field data

944 The sea spray generation fluxes measured in the laboratory are compared to  
945 other existing source functions obtained in the field and in laboratory con-  
946 ditions. Our data compare favourably with the SPANDEX laboratory data

947 reported by Fairall et al. (2009). Flux estimates differ typically by less than  
948 an order of magnitude when compared to flux functions obtained in the field  
949 (Smith et al., 1993; Andreas, 1998), which is in accordance with the typi-  
950 cal spread in the literature (Andreas, 1998; Veron, 2015). More substantial  
951 differences are noted from the spume droplet domain, when we compare our  
952 flux estimates to a function that does not accommodate these large particles  
953 explicitly (Laussac et al., 2018). These results prove that the laboratory is a  
954 good instrument for determining generation fluxes. This inspires us to continue  
955 the tunnel experiments, since the tunnel is better suited for the study of sea  
956 spray production for a wide range of wave fields and in high winds represen-  
957 tative of extreme events. Also, the tunnel facilitates the complex experiments  
958 required to understand the generation processes at the microphysical scale,  
959 such as airflow separation, and the effects of sea spray on heat, moisture and  
960 momentum fluxes at the air-sea interface. Ideally, such experiments should be  
961 performed with salt water, for easier comparison with field experiments, be-  
962 cause saltwater and freshwater lead to differences in the aerosol concentrations  
963 and distributions (Fairall et al., 2009; Mehta et al., 2019).

## 964 6.5 Sea spray generation functions (SSGFs) derived from laboratory 965 experiments

966 The laboratory experiments culminated in two sea spray generation functions  
967 (SSGFs) (cf. Fig. 11 and Table 2). These SSGFs describe the droplet genera-  
968 tion spectrum as a sum of three normally distributed modes, and are validated  
969 for wind speeds ranging from 12-20 m s<sup>-1</sup> and droplet radii 3-35  $\mu\text{m}$ . The radius  
970 range spans the major part of the film and jet ranges, as well as an apprecia-  
971 ble start of the spume droplet range. One SSGF solely dependent on the wave



972 slope variance, and to our knowledge is the first of its kind. Wave slope vari-  
973 ance measurements in the field are scarce, and for practical purposes it may  
974 be necessary to estimate the wave slope variance from more readily available  
975 variables such as wind speed (Eq. 3, Cox and Munk (1956)), or from mea-  
976 sured or modelled wave spectra (Elfouhaily et al., 1997). As shown in Section  
977 6.3, such approximations may reduce the accuracy of the SSGF. The second  
978 SSGF depends on a combined scaling parameter in terms of both  $\langle S^2 \rangle$   
979 and  $u_*^3$ . Though obtaining accurate measurements or predictions of  $u_*^3$  may  
980 be challenging in the field (and thus renders the use of the second SSGF even  
981 more difficult than the one depending solely on  $\langle S^2 \rangle$ ), this non-dimensional  
982 number is undeniably the best scaling tool for the generation of film and jet  
983 droplets, and also scales the larger spume droplets satisfactorily. In extrapolat-  
984 ing the  $P_S$ -dependent SSGF down to  $12 \text{ m s}^{-1}$  the flux may be underestimated  
985 at these lower wind speeds, following the suggestion by Andreas (1998) that  
986 the rate of spume droplet generation increases with  $u_*^2$  and not  $u_*^3$  for winds  
987 below  $15 \text{ m s}^{-1}$ .

## 988 6.6 Outlook

989 Recent improvements in the integration of higher order wave field properties  
990 in numerical wave models (Cathelain, 2017) and the coupling of wave, atmo-  
991 spheric and circulation models (Pianezze et al., 2018) provide an impetus for  
992 the study of sea spray generation processes. Present study results have shown  
993 that the integration of the wave field is essential for the better scaling of the sea  
994 spray generation. As we work towards universal SSGFs valid for a wide range  
995 of (complex) environmental conditions, a further understanding is required  
996 of the effect of multiscale (i.e. millimeter to meter scale) boundary processes

997 sea spray fluxes. This may include first and higher order statistics, such as  
998 for the consideration of overlapping wave fields, the directional spreading of  
999 wave spectra (Peureux et al., 2018) and wave-current interactions affecting  
1000 surface wave properties (Ardhuin et al., 2017). As the present study has high-  
1001 lighted the importance of the wave slope variance in modelling droplet fluxes,  
1002 we signal an urgent need for a better understanding of small-scale sea surface  
1003 characteristics (Jähne and Riemer, 1990). Unfortunately, field measurements  
1004 of these characteristics have been relatively sparse (Munk, 2009), not in the  
1005 least because the spatial resolution of airborne and satellite-based sensors is  
1006 limited and the presence of whitecaps complicates the retrieval of slope vari-  
1007 ance estimates (Cox and Munk, 1956; Lenain et al., 2019).

1008 **Finally, having demonstrated that the tunnel is a good proxy to the field in**  
1009 **determining sea spray fluxes, we would now like to extend the comparison to**  
1010 **the aerosol concentrations at some height above the water surface. However,**  
1011 **this is not a simple task since the differences in dispersion characteristics,**  
1012 **background concentrations and production footprints between the tunnel and**  
1013 **the field need to be elucidated. Efforts to do so are underway, and will be**  
1014 **reported elsewhere.**

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