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1	The "bag breakup" spume droplet generation mechanism at high
2	winds. Part I. Spray generation function.
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27 This paper describes the results of an experimental and theoretical investigation into 28 the mechanisms by which spume droplets are generated by high winds. The 29 experiments were performed in a high-speed wind-wave flume at friction velocities 30 between 0.8 m/s and 1.5 m/s (corresponding to a 10-m wind speed of 18-33 m/s under 31 field conditions). High-speed video of the air-water interface revealed that the main 32 types of spray-generating phenomena near the interface are "bag breakup" (similar to 33 fragmentation of droplets and jets in gaseous flows at moderate Weber numbers), 34 breakage of liquid ligaments near the crests of breaking surface waves, and bursting 35 of large submerged bubbles. Statistical analysis of these phenomena showed that at 36 wind friction velocities exceeding 1.1 m/s (corresponding to a wind speed of 37 approximately 22.5 m/s) the main mechanism responsible for the generation of spume 38 droplets is bag-breakup fragmentation of small-scale disturbances that arise at the air-39 water interface under the strong wind. Based on the general principles of statistical 40 physics, it was found that the number of bags arising at the water surface per unit area 41 per unit time was dependent on the friction velocity of the wind. The statistics 42 obtained for the bag breakup events and other data available on spray production 43 through this type of fragmentation were employed to construct a spray generation 44 function (SGF) for the bag breakup mechanism. The resultant bag breakup SGF is in 45 reasonable agreement with empirical SFGs obtained under laboratory and field 46 conditions.

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48

50 **1. Introduction**

51 Sea spray is a typical element of the marine atmospheric boundary layer and is of 52 great importance to marine meteorology, atmospheric chemistry, and climate studies. It is 53 considered a crucial factor in the development of hurricanes and severe extratropical storms, 54 which are responsible for the enhancement of energy flux from the ocean to the atmosphere (cf., e.g., Andreas and Emanuel, 2001; Andreas, 2011; Bao et al., 2011; Bianco et al., 2011; 55 56 Soloviev et al., 2014; Takagaki et al., 2012; Takagaki et al., 2016). According to the concept 57 of re-entrant spray put forward by Andreas and Emanuel (2001), the contribution of spray to 58 the energy flux is dominated by spume droplets: spray mechanically torn off the crests of 59 breaking waves, which rapidly sediments under the effects of gravity before a significant 60 fraction of its volume has had time to evaporate. The spray-mediated momentum flux is also dominated by the spume droplets, which are the main contributors to the volume flux of sea 61 62 spray (Fairall et al., 1994; Andreas 1998). However, it remains challenging to arrive at 63 estimates for the efficiency of the spray-mediated fluxes because the number and parameters 64 of spume droplets ejected from the water surface into the atmosphere at high winds are 65 uncertain due to both difficulties in taking measurements under storm conditions and uncertainties in the mechanisms of spray generation. As a result, empirical spray generation 66 67 functions (SGF), which describe the size spectra of spray ejected per unit area per unit time, 68 can differ for the spume droplets by up to six orders of magnitude in different observations 69 (cf., e.g., a compilation of experimental data by Veron (2015) and Andreas (2002)).

Koga (1981) reported the first observations of the process by which spume droplets are generated. This work showed the development of small liquid ligaments, mainly on the crests of breaking waves, that stretch and break, producing one or two droplets. An SGF based on this mechanism was developed by Mueller and Veron (2009). The second mechanism of sea spray production is via the bursting of bubbles formed at the crests of

breaking waves, as studied by Blanchard (1963), Spiel (1994a, 1994b, 1995, 1997, 1998), and Lhuissier and Villermaux (2012). Recently, Veron et al. (2012) reported on an alternative mechanism: fragmentation of water surface disturbances in the "bag breakup" regime. On the basis of high-speed video, Troitskaya et al. (2017) classified the spray-generating phenomena, estimated their efficiency and proved that bag-breakup fragmentation is the dominant mechanism of spume droplet production at high winds. In this paper, part I of this study, we construct the SGF for this dominant mechanism and compare it with the available data.

82 The structure of the paper is as follows. The technical details of the experimental 83 setup and methods of data acquisition and processing are given in Section 2. The 84 classification of phenomena responsible for the generation of spume droplets is described and 85 illustrated in Section 3. Section 3 expands on material briefly presented in Troitskaya et al. 86 (2017) and the Supplement to this paper. The results of statistical analysis of the spray-87 generating phenomena at different wind speeds are presented in Section 4, with a detailed 88 description of statistics relating to the most efficient bag breakup mechanism. Section 4 also 89 presents possible parameterizations of bag breakup statistics and discusses approaches to 90 applying the data obtained in laboratory experiments to field conditions. The data obtained 91 are employed for constructing an SGF for the bag breakup mechanism in Section 5. In 92 Section 6, the resultant bag breakup SGF is compared with available SGFs that were designed for application under laboratory and field conditions. In Appendix A the 93 dependence of the specific number of "bags" on wind friction velocity u_* is derived from the 94 95 general principals of statistical physics. Here, it is necessary to quote extensively from the 96 Supplement to Troitskaya et al. (2017) for completeness. Appendices B and C supply the 97 mathematical details of the derivation of the SGF.

99 **2.** Experimental setup and methods of measurement

100 **2.1.** The experimental setup and parameters of the experiment

Experiments were performed at the wind-wave flume of the Large Thermally Stratified Tank of the Institute of Applied Physics of the Russian Academy of Sciences (IAP RAS). The airflow channel has a cross-section 0.4 m × 0.4 m over the water surface and the length of 10 m. The centerline velocity range is 3–25 m/s. The tank is filled with fresh water, with a temperature ranging from 15 to 20°C. The measured value of the surface tension was $\sigma = (7.0 \pm 0.15) 10^{-2}$ N/m. The facility and the parameters of the air flow and surface waves are described in detail in Troitskaya et al. (2012).

108 To characterize the air flow above the water surface, we use the parameters of the 109 atmospheric turbulent boundary layer: the wind friction velocity, u_* , roughness height, z_0 , and 110 10-m wind speed, defined as follows:

111
$$U_{10} = \frac{u_*}{\kappa} \ln \frac{H_{10}}{z_0}$$

112 where $\kappa = 0.4$ (the von Karman constant) and $H_{10} = 10$ m.

113 In the wind-wave flume, u_* is in the range 0.2–2 m/s and U_{10} is 7–36 m/s. The 114 dependence of z_0 on u_* in the tank follows the Charnock formula:

115
$$z_0 = \alpha \frac{u_*^2}{g}$$
, (1)

116 with the Charnock constant α =0.0057 ± 0.0005 (cf. Fig. 1a).

117 The wind wave field parameters in the flume were measured by three wire gauges 118 positioned in the corners of an equilateral triangle with 2.5 cm sides; the data sampling rate 119 was 100 Hz. The frequency spectra have sharp peaks depending on u^* and the fetch. The 120 dependency of the peak frequency ω_p on u^* in the working section at a fetch of 6.5 m is 121 plotted in Fig. 1b. The experimental points are best fitted by the power function:

122
$$\omega_p = \Omega_{u0} u_*^{\gamma}, \qquad (2)$$

123 where $\Omega_{u0}=12.4 \text{ m}^{1/2}\text{s}^{-3/2}$ with the 95% confidence interval between 12.2 m^{1/2}s^{-3/2} and 12.6 m^{1/2}s^{-3/2}, and $\gamma=0.5 \pm 0.04$.

125 **2.2.** Optical scheme and experimental techniques for investigating spray-generating

126 phenomena using the shadow method

127 Using the shadow method for visualization, video of the air-water interface was 128 captured by a NAC Memrecam HX-3 high-speed digital video camera from two angles: a top 129 view of the channel at 6.5 m fetch and a side view at 7.5 m fetch. For the side view, the 130 camera was placed in a waterproof box attached to the side wall of the channel at 7.5 m fetch 131 (the horizontal shadow method, Fig. 2a). The optical axis of the camera lens was located 5 132 cm above the water surface and was directed horizontally. The distance from the camera to 133 the shooting area was 65 cm. A 300-W LED spotlight was mounted at the side of channel 134 section 8 at a distance of 50 cm from the wall and a height of less than 5 cm from the surface 135 of the water. A diffuser screen was placed on the side wall of the channel opposite the camera. 136 The 85–mm focal length lens provided an image size of 75×66 mm (1024×904 px, 73μ m 137 pixel size), the recording rate was 10,000 fps, and the exposure time was 50 µs. Detailed side 138 view records of spray-generating phenomena were obtained for wind speeds from 18 m/s to 139 33 m/s.

To obtain statistics on the spray-generating phenomena, top-view video was filmed using underwater lighting (the vertical shadow method, Fig. 2b). The video was captured through the transparent top wall at 6.5-m fetch. The camera was mounted vertically at a distance of 207 cm from the water surface. The 85–mm focal length lens provided an image size of 147×377 mm (576 × 1472 px, 256 µm pixel size), the recording rate was 4,500 fps.

146 **3.** Classification of phenomena responsible for generation of spume droplets

147 The combination of the two shooting angles revealed the phenomena responsible for 148 the generation of spume droplets. A brief description and classification of these are given in 149 Troitskaya et al. (2017). This section presents an extended description and new illustrations.

Experiments were performed at airflows with friction velocities from 0.8 m/s to 1.51 m/s corresponding to 10-m wind speeds between 18 m/s and 33 m/s under field conditions according to Foreman and Emeis (2010). Analysis of the images enables us to specify three types of phenomena responsible for the generation of the spume droplets near the wave crest: breakage of liquid ligaments, bursting of large (approximately 1-cm diameter) submerged bubbles, and bag breakup.

156 **3.1. Breakage of liquid ligaments**

157 Fig. 3 illustrates the mechanism by which droplets are generated through breakage of 158 liquid ligaments, as discovered and studied by Koga (1981). Recently, Mueller and Veron 159 (2009) constructed an SGF based on this mechanism. According to Koga (1981), the Kelvin-160 Helmholtz instability at the air-water boundary leads to the development of liquid ligaments, 161 mainly at the crests of breaking waves, which stretch and then break into droplets. An 162 example of droplet formation by this mechanism under field conditions is shown in Fig. 3e. 163 The details of the structure of the breaking wave crest as shown in insets to Fig. 3e are similar 164 to the structures shown in Fig. 3a-d. Fig. 3b-d confirm that each breakage of a ligament produces a few droplets with diameters of 1-2 mm, which typically fall to the water close to 165 166 the breaking crest.

167 **3.2. Bursting of large submerged bubbles**

Entrainment of air at breaking wave crests leads to the formation of a large number of bubbles, which emerge because of their positive buoyancy and burst into droplets as they reach the water surface (Fig. 4). A detailed model of this phenomenon was recently 171 developed by Lhuissier and Villermaux (2012). The mechanism of the spray production due 172 to bursting of smaller bubbles (less than 10 µm) has been studied by Blanchard (1963) and Spiel (1994a, 1994b, 1995, 1997, 1998). Wu (1981) considered bursting bubbles to be the 173 174 main source of ocean spray with the radii below 50 µ. According to experiments by Lhuissier 175 and Villermaux (2009, 2012), bursting of a bubble begins with a local reduction of the film 176 thickness and the formation of a hole. The rim bounding the hole moves along the curved 177 surface of the bubble during its expansion. Resulting centrifugal acceleration causes the 178 development of Rayleigh-Taylor instability, accompanied by the formation of ligaments that fragment into droplets. Fig. 4 shows that, in the presence of strong wind and waves at the 179 180 water surface, the bursting of a submerged bubble touching the water surface occurs as in air 181 and water at rest, as investigated by Lhuissier and Villermaux (2009, 2012).

182 **3.3. Bag breakup.**

183 A sequence of top-view (Fig. 5a) and side-view (Fig. 5b) frames from two different 184 videos illustrates another effective mechanism for generating spume droplets at high wind. It 185 starts with an increase in the small-scale elevation of the surface (Fig. 5a, 0 ms, Fig. 5b, 0 ms), 186 which then transforms into a small liquid "sail", inflates into a canopy bordered by a thicker 187 rim (the thick rim) (Fig. 5a, 4.7, 8.2 ms, Fig. 5b, 3.4 ms), and finally ruptures to produce 188 spray (Fig. 5a, 10.5 ms, Fig. 5b, 5.6 ms). In some cases, the initial elevated area was 189 transformed into a more complex structure comprising several inflating canopies (Fig. 5c-d). 190 The above-described process by which this occurs is well-known in industrial fluid dynamics 191 as the bag breakup regime of liquid fragmentation in gaseous flows (cf., e.g., Gelfand, 1996). 192 This regime of spume droplet generation at the crests of wind waves was recently observed in 193 a laboratory flume by Veron et al. (2012). Below, we will use the term "bag" to refer to the 194 observed structure.

195 The process of rupture of the canopy looks similar to the process of bubble bursting 196 investigated by Lhuissier and Villermaux (2009, 2012): it also involves a hole bounded by a 197 rim (the thin rim). A rim moving along the curved film of the canopy during the expansion of 198 the hole leads to the formation of ligaments and drops through the development of Rayleigh-199 Taylor instability. The thick rim remains after the bubble has burst and then experiences 200 fragmentation into droplets that are large in comparison to those formed at the rupture of the 201 canopy. We emphasize that the distinguishing feature of a bag from a bubble is the presence 202 of two rims limiting the film. As a result, the size distribution of droplets can be expected to 203 show two typical scales.

204 Fig. 6a–d schematically illustrate the typical stages of fragmentation of the air–water 205 interface in the bag breakup mode, similar to droplets in gaseous flows. Note that research 206 has not yet been able to determine the detailed appearance of the initial disturbance that will 207 be transformed into a bag. In Fig. 6a, it is assumed that the growth of the initial elevation in 208 the water surface is governed by shear flow instability. This assumption is indirectly 209 supported by estimates of the sizes of bags given in Section 4.3. With the increase of the 210 surface elevation, the airflow becomes asymmetrical: the pressure minimum is shifted to the 211 leeward side of the elevated region of water (Fig. 6b), eventually turning it to a liquid sail 212 (Fig. 6c). This process is similar to the transformation of droplets in gaseous flow into a thin 213 disk moving across the flow. The result is the distortion of the shape of the elevated region of 214 water, leading to inflation of the canopy (Fig. 6d), which then raptures (Fig. 6e) and bursts, 215 producing spray.

The statistical analysis below shows that bag breakup appears to be the dominant mechanism of spume droplet generation at high winds.

218

4. Statistics of local phenomena responsible for generation of spume droplets

4.1. Method and analysis of statistics of spray-generating phenomena

221 Statistical data for the spray-generating phenomena (breaking ligaments, bursting 222 underwater bubbles, and bag breakup events) was retrieved from the sequence of video 223 frames using software that allows the selection and counting of objects in images semi-224 automatically (see details in Troitskaya et al., 2017). For the bag breakup events, the software 225 also enables one to obtain the geometrical and kinematic parameters of the objects (cf. Fig. 7), 226 including the initial size, D_1 , of the bag, defined as the distance between edge markers in a 227 frame showing the nucleation of the bag; the bag final size, D_2 , defined as the distance 228 between edge markers in a frame showing film puncture; and the bag lifetime from the 229 moment of its nucleation until the moment of film puncture, τ . The velocities of the bag 230 edges and center, u_1 , and u_2 , were calculated as the distance between, respectively, the 231 midpoints of edge markers or the centers of the canopy on the initial and final frames, divided 232 by τ . Below, we also use the initial and final bag radii, $R_1 = D_1/2$ and $R_2 = D_2/2$.

233 Fig. 8a shows the dependence on wind friction velocity, u_* , of the specific number 234 (per unit time per unit area) of spray-generating phenomena obtained using semi-automatic 235 processing. The numbers of processed images are given in Table 1. The number of frames 236 required for the collection of statistics decreased with increasing wind speed, while the 237 number of bags increased. One can see that the specific numbers of local events of any type 238 (ligaments, bursting bubbles, or bags) increase with increasing u_* , with bags showing the 239 greatest growth rate. Note that in the multi-bag regime, each canopy of the complex object 240 was treated in the statistics as one bag. For $u \le 1.1$ m/s, the numbers of the spray-generating 241 phenomena are approximately equal, beyond which the number of bubble bursts is less than 242 the number of ligaments and bags. For $u \ge 1.1$ m/s, the number of bags exceeds the number of 243 ligaments. Given that breaking of one ligament produces only one or two droplets (see Fig. 3) but bag fragmentation produces hundreds of droplets (see Fig. 5) we conclude that, for $u^* >$ 1.1 m/s, bag breakup becomes the dominant mechanism of spume droplet production.

It should be noted that the activation thresholds for the friction velocity were obtained at the laboratory facility. Under field conditions at considerably larger wind fetches and different wind wave regimes, these values may differ. It may be possible to makes estimates of these different values using the approach of Toba and Koga (1986). They suggested parameterizing the strongly nonlinear phenomena in the boundary layers near the air–sea interface using the windsea Reynolds number (this term was suggested later by Toba et al. (2006)):

253
$$\operatorname{Re}_{B} = \frac{u_{*}^{2}}{\omega_{p}v},$$
(3)

254 where ω_p is the peak frequency in the spectrum of surface wind waves, and v is the kinematic 255 viscosity of the air. Iida et al. (1992) and Zhao et al. (2006) showed that the parameter Re_B 256 was effective for scaling the spray droplet production rate under both laboratory and field conditions, and Toba and Koga (1986), Zhao et al. (2006) and Toba et al. (2006) successfully 257 258 used it to scale the wind-sea breaking rate, whitecap coverage and transfer coefficients for 259 momentum and CO₂. Following these studies, we used this parameter for scaling the specific 260 number of the spray-generating phenomena. The peak frequency, ω_p , in the flume was 261 measured directly. At the fetch of the working section, the dependence of peak frequency, ω_{p} , 262 on u_* is given by Eq. (2).

Fig. 8b plots the specific numbers of bags, bursting bubbles and projections versus the windsea Reynolds number, Re_B . This shows that the threshold for activation of bag breakup, as well as other spray-generating phenomena, is $Re_B \approx 4000$, and that bag breakup becomes the dominant spray-production mechanism for $Re_B > Re_{Bcr} \approx 8000$. Note that, according to Toba and Koga (1986), $Re_{Bcr} \approx 8000$ should be a universal number applicable both under 268 laboratory and field conditions, despite only being retrieved from laboratory data. Given that 269 the gravity wave dispersion relation yields $\omega_p c_p = g$ where c_p is the phase velocity of surface

270 waves with a frequency
$$\omega_p$$
, $\operatorname{Re}_B = \frac{u_*^3}{g v \Omega \sqrt{C_D}}$ where $C_D = u_*^2 / U_{10}^2$ is the sea surface drag

271 coefficient and $\Omega = U_{10}/c_p$ is the wave-age parameter. Using the empirical expression of 272 Foreman and Emeis (2010) for $u_*(U_{10})$, $u_* = 0.051U_{10} - 0.14$, for estimating C_D yields the 273 equations for Re_B in terms of U_{10} :

274
$$Re_{B} = \frac{(0.051U_{10} - 0.14)^{2} U_{10}}{g v \Omega}$$
(4)

275 It follows from Eq. (4) that U_{10} decreases with decreasing Ω for a fixed Re_B , and so 276 the threshold wind velocity at which spume spray production will start will decrease with the 277 development of the wave field, which is accompanied by decreasing Ω . This is illustrated in 278 Fig. 8c, where the wind speed is plotted against the wave-age parameter Ω for constant Re_B 279 equal to 4000 and 8000, corresponding to the threshold for activation of the bag breakup 280 mechanism and the condition where bag breakup becomes the dominant mechanism for 281 production of the spume droplets. It follows from Fig. 8c that for Ω between 1 and 3, typical values for open ocean conditions, the bag breakup activation threshold is between 8 m/s and 282 283 10 m/s, and at a wind speed between 9.5 m/s and 13 m/s, the mechanism becomes dominant. 284 It is interesting to note that the first range corresponds to number 5 of the Beaufort scale, 285 when, according to the state-of-the-sea scale of Petrsen (1927), "many white horses are 286 formed; chance of some spray."

It should be emphasized that only droplets with sizes exceeding 10 μm are being
discussed here. For smaller droplets, the main generating mechanism is bubble bursting (see,
e.g., Wu, 1981), and the contribution of bag-breakup fragmentation is uncertain.

290 **4.2. Statistics of bag breakup events**

291 To describe the statistics on the number of bag breakup events, we use a 292 phenomenological approach based on the Gibbs (1902) method, initially introduced in 293 equilibrium statistical mechanics. The central concept of this method is the canonical 294 ensemble, or the ensemble of states of a large system described in the statistical approach. According to Rumer and Ryvkin (1980), the concept of the Gibbs canonical ensemble allows 295 296 its universal application to any large system and not just thermodynamic systems consisting 297 of atoms and molecules. Based on this approach, it is possible to derive an expression for the 298 specific number of bag breakup events (see details in the appendix to Troitskaya et al. (2017) 299 and Appendix A) as follows:

$$300 \quad \langle N \rangle = Q_0 u_*^2 \exp\left(-\frac{U_0^2}{u_*^2}\right), \tag{5}$$

The constants in Eq. (5), $U_0=2$ m/s with a 95%-confidence interval between 1.87 m/s and 2.13 m/s, $Q_0=9.27\cdot10^2$ m⁻⁴s with a 95%-confidence interval between 5.91·10² m⁻⁴s and 1.45·10³ m⁻⁴s, are determined as the best fit to the experimental data shown in Fig. 9a.

304 Note, that the state of the air-sea system is characterized by one more parameter, the 305 wind fetch. Although the form of the expression for $\langle N \rangle$ obtained from the general principals 306 of statistical physics remains valid, some changes in the constants are expected with a change 307 in fetch. This should be taken into account when constructing models for field conditions. We 308 here consider a possible parameterization of the specific number of bags with the windsea Reynolds number Re_B (Eq. (3)) introduced by Toba and Koga (1986) and reformulate Eq. (5) 309 accordingly, introducing the dimensionless parameter $V = \frac{U_0^2}{\omega_n v}$. Given that, according to 310 311 Toba and Koga (1986), the dimensionless V and Q_0 determined by the state of the air-sea interface under the action of wind are functions of Re_B , Eq. (5) yields: 312

313
$$\langle N \rangle = Q_0 \left(Re_B \right) \frac{Re_B}{V \left(Re_B \right)} exp \left(-\frac{V \left(Re_B \right)}{Re_B} \right).$$

314 Suppose that $Q_0(Re_B)$ and $V(Re_B)$ are the power functions of Re_B , then:

315
$$\langle N \rangle = M_0 R e_B^{\mu_0} exp\left(-\frac{M_1}{R e_B^{\mu_1}}\right).$$
 (6)

To determine constants in Eq. (6) we used the results of Zhao et al. (2006), who showed that the production rate of the spume droplets is proportional to $Re_B^{1.5}$. Given that bag breakup is the dominant mechanism for spume droplet production at Re_B >8000 and assuming that one bag produces on average a certain number of droplets, we can expect that the dependencies of the spume droplet production rate and the specific number of bags on Re_B have the same asymptotics $Re_B^{1.5}$ at large enough Re_B , when the bag breakup spray production mechanism dominates. The best fit to the experimental data in Fig. 9b then gives:

323
$$\langle N \rangle = M_0 R e_B^{3/2} exp\left(-\frac{M_1}{R e_B^{3/2}}\right),$$
 (7)

The best fit to the data in Fig. 9b gives the following constants in Eq.(7): $M_0=2.58\cdot10^{-4}\text{m}^{-2}\text{s}^{-1}$ with a 95%-confidence interval between $2.22\cdot10^{-4}\text{m}^{-2}\text{s}^{-1}$ and $3.00\cdot10^{-4}\text{m}^{-2}\text{s}^{-1}$, $M_1=6.93\cdot10^{5}$ with a 95%-confidence interval between $6.22\cdot10^{5}$ and $7.64\cdot10^{5}$, and the relative error in the specific number of bags defined by the 95% confidence interval is approximately 15%.

We also considered another option to translate our laboratory data for field conditions is a simple rescaling of the specific number of bags using the $Re_B^{3/2}$ -dependence of the spray production rate. Eq. (3) for Re_B yields, for a certain u_{*} :

331
$$\langle N \rangle_{field} = \langle N \rangle_{lab} \left(\frac{\omega_{p-lab}}{\omega_{p-field}} \right)^{3/2}$$
.

332 The dispersion relation for the surface gravity waves: $\omega = \sqrt{gk}$ yields $\omega_{p-field} = g/c_p$. This 333 gives

334
$$\omega_{p-field} = g\Omega/U_{10} = g\Omega\sqrt{C_D}/u_*.$$
(8)

Using Eq. (2) for ω_{p-lab} and Eq. (8) for $\omega_{p-field}$ yields the specific number of bags under field conditions:

337
$$\langle N \rangle_{field} = Q_0 \frac{u_*^2}{U_0^2} \left(\frac{12.4 u_*^{0.5}}{g \Omega \sqrt{C_D}} \right)^{1.5} \exp\left(-\frac{U_0^2}{u_*^2} \right).$$
 (9)

We compared the values of $\langle N \rangle$ calculated according to Eqs. (7) and (9), including for hurricane conditions. In accordance with the direct measurements of a wave field in hurricane conditions made by Wright et al. (2001), the wave age parameter, Ω , was taken to be between 2.5 and 3.5. Fig. 9c compares the dependencies of the specific number of bags on wind speed U_{10} , as calculated using Eqs. (7) and (9). For C_D , in both cases we used an approximation of non-monotonous dependence on U_{10} after Holthuijsen et al. (2012), where the latest drop sonde measurements of the drag coefficient were summarized:

345
$$C_D = \begin{cases} (0.057 - 0.48/U_{10})^2 \text{ for } U_{10} < 40 \text{ m/s} \\ (2.37/U_{10} - 0.012)^2 \text{ for } U_{10} > 40 \text{ m/s} \end{cases}$$

A comparison of the curves in Fig. 9c shows that values for <N> when using Eqs. (7) and (9)
differ significantly at lower winds, but are very similar at high winds.

It should be emphasized that Eqs. (7) and (9) were obtained on the basis of a limited dataset obtained in a laboratory experiment in a straight channel with a very short wind fetch and, in this regard, should be considered preliminary. The data are not yet sufficient to clearly favor one of the two approaches, and in part II of this study, we will compare the estimates of the exchange coefficients obtained by using each of them. Further refinement of these expressions can be expected as data is accumulated in experiments at large wind fetches, including those with artificial fetch enhancement, as suggested by Takagaki et al. (2017).

4.3. Statistical distributions of the geometrical parameters of bags

356 Semi-automatic processing of the video allowed us to study the statistical distribution

of bag size (radii at nucleation, R_1 , and film rupture, R_2), velocity (of edges, u_1 , and centers, u_2), and typical lifetime between nucleation and film puncture, τ , for different air flow velocities. Fig. 10a–c show that the probability density of these quantities normalized to the median values can be well-approximated by the gamma distribution:

361
$$P_n(x) = \frac{n^n}{\Gamma(n)} x^{n-1} e^{-nx}$$
(10)

 $x = X / \langle X \rangle$ in Eq. (10) represents one of the physical variables R_1 , R_2 , u_1 , u_2 and τ , 362 363 normalized by its mean value: $\langle R_1 \rangle$, $\langle R_2 \rangle$, $\langle u_1 \rangle$, $\langle u_2 \rangle$, $\langle \tau \rangle$ and $\Gamma(n)$ is Euler's Gamma 364 function. Eq. (10) generalizes the gamma distribution used in Troitskaya et al. (2017) to the 365 case of fractional parameters. For R_1 and R_2 , n=7.53, for u_1 and u_2 , n=13.30, and for τ , 366 n=3.70. It is interesting to note that the bag parameters are described by the gamma distribution similarly to completely different objects, for example, droplets produced by 367 fragmentation of ligaments or liquid film (cf. Marmottant and Villermaux, 2004; Lhuissier 368 369 and Villermaux, 2012).

The dependence of the average values on the friction velocity, u_* , of the air flow is shown in Fig. 11a–c. There is a clear decrease in the size and lifetime of the bags and increase in the velocity of the edges and center with increasing wind speed. The corresponding empirical dependencies can be approximated as follows:

374
$$\langle R_1 \rangle (u_*) = 5.9 u_*^{-1},$$
 (11)

375
$$\langle R_2 \rangle (u_*) = 9.6 u_*^{-1},$$
 (12)

$$376 \quad \langle u_1 \rangle (u_*) = 1.96 + 1.21 \, u_*, \tag{13}$$

377
$$\langle u_2 \rangle (u_*) = 1.1 + 4.2 \, u_*$$
, (14)

378
$$\langle \tau \rangle (u_*) = 7.7 \ u_*^{-2},$$
 (15)

379 where u_* is measured in m/s, $\langle R_1 \rangle$ and $\langle R_2 \rangle$ in mm and $\langle \tau \rangle$ in ms.

380 These dependences of the bag parameters on wind friction velocity can be explained if it is assumed that the water surface perturbations from which they develop arise as a result of 381 382 the shear instability of the water and air layers near the interface. The thickness of these 383 layers in air and water, δ_a and δ_w , can be estimated as the scales of the buffer layers of turbulent boundary layers, $(20 \div 30) v_a / u_*$ and $(20 \div 30) v_w / u_* \sqrt{\rho_w / \rho_a}$, respectively (here v_a 384 and v_w are the kinematic viscosity coefficients of air and water, and ρ_a and ρ_w are their 385 386 densities). Note that these quantities are of the same order. The velocity difference in shear layers in air is ~ $(10 \div 12)u_*$ and in the water is ~ $(10 \div 12)u_*\sqrt{\rho_a/\rho_w}$ (see, e.g., Hinze, 1959). 387 The spatial scale of the most unstable disturbances is scaled by the thickness of the shear 388 layer, and then it is $\sim v_a/u_*$, in agreement with the dependence of the average bag size on 389 wind friction velocity in Eqs. (11) and (12). Accordingly, the lifetimes of bags of these sizes 390 in a flow with velocity scaled by u_* are proportional to v_a / u_*^2 , in agreement with Eq. (15). 391

Fig. 11d shows the plane (R_1, R_2) , where points correspond to individual bags. These values are proportional, with a correlation coefficient of 0.97. This indicates that the evolution of the bag form is self-similar, i.e., it approximately preserves its form across a range of sizes.

Finally, we can construct the frequency distribution of bag sizes. We present it here as the function of the radius of the bag at the moment of rupture $R = R_2$, which will be used below for constructing an SGF. Combining Eqs. (7) or (9) for the average specific number of bags, $\langle N \rangle$, their size distribution Eq. (10) and the dependence Eq. (12) of the average size $\langle R_2 \rangle (u_*)$ on wind friction velocity gives:

$$401 \qquad F_{bag}\left(R, u_{*}\right) = \frac{\langle N \rangle}{\langle R_{2} \rangle(u_{*})} \frac{n^{n}}{\Gamma(n)} \left(\frac{R}{\langle R_{2} \rangle(u_{*})}\right)^{n-1} e^{-n\left(\frac{R}{\langle R_{2} \rangle(u_{*})}\right)}, \tag{16}$$

402 with *n*=7.53.

403

404 **5.** Construction of a function for spray generation due to the bag breakup mechanism

It is now possible to construct the bag breakup SGF, i.e., the number of spray droplets with radii in the range $[r; r+\Delta r]$ generated per unit time per unit area due to the bag breakup mechanism. There are two ways of producing droplets through bag breakup: (i) rupture of the canopy of the inflated bag (Fig. 5a, 10.5 ms and Fig. 5b, 5.6 ms); and (ii) fragmentation of the rim that survives briefly after the rupture of the bag (Fig. 5b, 11.7 ms). Here we first construct the size spectra of droplets produced by each of these two mechanisms.

411

5.1 The statistical distribution of the canopy droplets

412 When considering the statistics of the droplets produced by rupture of the bag canopy, 413 we used the results of a detailed study by Lhuissier and Villermaux (2012) concerning a 414 similar mechanism for the generation of spray through the bursting of a submerged bubble 415 touching the surface. Visually, the fragmentation dynamics of the two cases look similar, 416 since they are governed by the same mechanism governed by surface tension. Lhuissier and 417 Villermaux (2012) obtained a size spectrum for droplets (the average number of droplets 418 versus the droplet radius, r) generated through rupture of the bubble cap (the film above the 419 bubble connected to the bulk of the water via a surrounding meniscus) with curvature radius, *R*, as follows: 420

421
$$F_1(r,R) = \frac{N_{film}(R)}{\langle r \rangle(R)} P_m\left(\frac{r}{\langle r \rangle(R)}\right), \qquad (17)$$

422 where $P_m(\mathbf{x})$ is the gamma distribution in Eq. (10) with m = 11.

423 Based on thorough optical measurements, Lhuissier and Villermaux (2012) obtained the 424 dependence of the average diameter, $\langle d \rangle$, and total number of droplets from the burst of a 425 bubble, $N_{film}_{drops}(R)$, on *R*. The power best fit to the experimental data taken from Fig. 20a of

426 Lhuissier and Villermaux (2012) yields the following empirical equation for $\langle d \rangle$ on *R*:

427
$$\langle d \rangle = 0.19L \left(\frac{R^{3/8}h^{5/8}}{L}\right)^{0.8},$$
 (18)

428 Here, *h* is the thickness of the cup of the bubble at the moment of rupture. According to429 Lhuissier and Villermaux (2012):

$$430 h = L \left(\frac{R}{L}\right)^2, (19)$$

431 where $L=2 \ 10^4$ mm.

432 The best fit to the experimental data on $N_{film}_{drops}(R)$, taken from Lhuissier and Villermaux's

433 (2012) Fig. 20b gives:

434
$$N_{film}_{drops} = 2.24 \cdot 10^{-3} \left(\left(\frac{\rho_w g R^2}{\sigma} \right) \left(\frac{R}{h} \right)^{7/8} \right)^{1.18}$$
 (20)

Given that Eqs. (18) and (19) yield the following empirical dependence of the average radius of droplets $\langle r \rangle = \langle d \rangle / 2$ on the bubble radius:

437
$$\langle r \rangle (R) = aL \left(\frac{R}{L}\right)^{\alpha}; a = 0.094; \alpha \approx 4/3.$$
 (21)

and given that Eqs. (19) and (20) yield for the total number of droplets from the burst of abubble

440
$$N_{film}_{drops} = b \left(\frac{R}{L}\right)^{\beta}; b = 2.24 \cdot 10^{-3} \left(\frac{\rho_w g L^2}{\sigma}\right)^{1.18}; \beta \approx 4/3,$$
 (22)

441 for a bag, *R* is interpreted as its curvature radius at the moment of rupture.

Finally, the total number of droplets with radii in the range $[r, r+\Delta r]$ produced by rupture of the bag canopies per unit area per unit time is the convolution of the size spectra of bags (Eq. (16)) with the size spectra of droplets generated due to the rupture of the canopy of the inflated bag (Eq. (17)). Below we will use the term "canopy droplets" to distinguish them from the "canopy droplets" originating from bursting bubbles. The derivation of the equation for the generation function for the canopy droplets, $\frac{dF_c(r)}{dr}$, is presented in Appendix B.

448

5.2 The statistical distribution of rim droplets.

To describe the size spectrum of droplets resulting from fragmentation of the rim, we consider it as a liquid ligament of a certain thickness prescribed by the radius of the bag, *R*. According to Marmottant and Villermaux (2004), the statistical distribution of droplets produced by fragmentation of such objects follows the gamma distribution (Eq. (10)), with $x=r/r_1$, n=4. Here $r_1=0.4r_0$, where r_0 is defined as the radius of a sphere with a volume equal to the volume of the initial filament (the rim in our case), $V: V = 4\pi r_0^3/3$.

Before constructing an SGF for the rim droplets, we need to determine the 455 456 relationship of the rim volume, V, with the measured bag radius, R. For this purpose, we use 457 its similarity with the well-known bag breakup regime of secondary fragmentation of droplets 458 in gaseous flows. For this case, Chou and Faeth (1998) showed that the rim volume is equal 459 to 0.56 of the initial volume of a droplet. In the case of bag-breakup fragmentation of the air-460 water interface, the initial volume of the object that is going to be fragmented is not defined, 461 and we use the following argument. We model the initial shape of a bag as a semi-circular disc with a radius R_1 and thickness h_1 . The initial volume of the object is $V_1 = \pi R_1^2 h_1 / 2$. At 462 the moment of rupture, the shape of a bag with radius R_2 is approximated to a liquid ring 463 (torus) with a thickness h_2 , which holds the liquid film. The torus volume is $V = \pi^2 R_2 h_2^2 / 2$. 464 Based on observations by Chou and Faeth (1998), we assume that the thickness of the rim 465 466 does not change in the course of its evolution, i.e., $h_1 = h_2 = H$. We also suppose that the ratio 467 of the rim volume, V_{i} to the initial liquid volume, V_{i} remains as found by Chou and Faeth

468 (1998) for the bag breakup regime of the secondary fragmentation of droplets, i.e. $V=0.56V_1$.

469 We then have $0.56\pi R_1^2 H/2 = \pi^2 R_2 H^2/2$ and $H = 0.56 R_1^2/(\pi R_2)$.

Given the strong correlation between R_1 and R_2 seen in Fig. 11d (the coefficient of determination is above 0.95), we assume a linear relationship between R_1 and R_2 . The linear best fit line shown in Fig. 11d gives $R_2 \approx 1.66R_1$. Finally, $H = 0.56R_2 / (1.66^2 \pi) = 0.065R_2$, and the rim volume is $V = \pi^2 R_2 H^2 / 2 = 0.021R_2^3$. The radius of an equivalent sphere is $r_0 = \sqrt[3]{3V/4\pi} = 0.17R_2$ and the scale in the gamma distribution for the "rim" droplets according to Marmottant and Villermaux (2004), $r_1=0.4r_0=\gamma R_2$ where $\gamma=0.068$.

476 The average number of rim droplets from one bag, N_{rim}_{drops} , can be found as the ratio of

477 the rim volume, V, to the average volume of a drop, $\langle V \rangle$, resulting from its fragmentation. 478 Given that the size statistics of the rim droplets is described by the gamma distribution with 479 the parameter 4, we have:

480
$$\langle V \rangle = \frac{4\pi}{3} r_1^3 \int_0^\infty x^3 \frac{4^4}{3!} x^3 e^{-4x} dx = \frac{15}{8} \frac{4\pi}{3} r_1^3$$

481 For $r_1=0.4r_0$, we have $N_{rim}_{drops} = V / \langle V \rangle = 8.3$.

482 Finally, the size spectrum of droplets (average number of droplets over the whole483 range of radii, *r*) generated through fragmentation of the rims is as follows:

484
$$F_2(r,R) = \frac{N_{rim}}{\gamma R} P_k\left(\frac{r}{\gamma R}\right), \qquad (23)$$

485 where P_k (x) is the gamma distribution (Eq. (10)) with k=4, $\gamma=0.068$, $N_{rim}_{drops} = 8.3$. Here and

486 below we use the notation R instead of R_2 .

487 The total number of rim droplets with radii in the range $[r, r+\Delta r]$ produced by bags per 488 unit area per unit time is the convolution of the size spectra of bags (Eq. (16)) with the size 489 spectra of droplets generated due to the fragmentation of the rim (Eq. (23)). The expression
490 for the generation function for the rim droplets is derived in Appendix C.

491 The complete bag breakup SGF is the sum of the contributions of the canopy and rim
492 droplets given in Appendices B and C (Eqs. (B6) and (C5), respectively)

$$493 \qquad \frac{dF(r,u_*)}{dr} = \langle N \rangle \left(\frac{3.3 \cdot 10^{-9}}{L} \left(\frac{\rho_w g L^2}{\sigma} \right)^{1.18} \left(\frac{r}{\theta} \right)^{7.3} e^{-5.2 \sinh\left(\frac{3}{7}\ln\frac{r}{\theta}\right)} + \frac{1.5 \cdot 10^{-4} N_{rim}}{\Theta} \left(\frac{r}{\Theta} \right)^{4.5} e^{-3.94 \sinh\left(\frac{1}{2}\ln\frac{r}{\Theta}\right)} \right).$$

$$494 \qquad (24)$$

In Eq. (24), $\langle N \rangle$ is defined by Eqs. (5) and (7). Expressions for θ and Θ are derived in Appendices B and C, respectively: $\theta = 0.001 (\langle R_2 \rangle (u_*))^{4/3} L^{-1/3}$, $\Theta = 0.0021 \langle R_2 \rangle (u_*)$, where $\langle R_2 \rangle (u_*)$ is expressed by Eq. (12) and L=20 m (cf. Lhuissier and Villermaux, 2012). The uncertainty of the SGF given by Eq. (24) as well as the specific number of bags, $\langle N \rangle$, is about 15% with a 95% confidence interval.

500

509

501 6. Properties of the spray generation function for bag breakup and comparison with 502 laboratory and field data

The SGF defined by Eq. (24) is shown in Fig. 12a. It has two clear peaks, corresponding to canopy droplets with average radii of approximately 100 µm and giant rim droplets with average radii of approximately 1 mm; this is shown directly in Fig. 12b, which plots $\frac{dF_{rim}(r,u_*)}{dr}$ and $\frac{dF_{film}(r,u_*)}{dr}$ separately. The rim-droplet peak at r = 500-1000 µm is the distinctive feature of the bag breakup spray generation mechanism. Giant droplets torn off the wave crests have also been observed in laboratory experiments reproducing hurricane

510 Note that although the number of giant rim droplets is small, we can expect that they will

conditions (Veron et al., 2012; Ortiz-Suslow et al., 2016; Iida et al., 1992, Fairall et al., 2009).

significantly support the spray volume flux. This is confirmed by a strongly enhanced peak in the SGF as a volume flux at sizes of about 1000 μ m, corresponding to the rim droplets (cf. Fig. 12c). It should also be noted that the maxima in size spectra of the droplets change with wind speed, similarly to Mueller and Veron (2009), because the average size of the bag, $<R_2>(u*)$, which in turn scales both canopy and rim droplets (θ , Eq. (B7) and Θ , Eq.(C6)), depends on u*.

517 We compare in Fig. 13 the bag breakup SGF (Eq. (24)) with available experimental 518 SGFs derived from laboratory data by Iida et al. (1992), Fairall et al., 2009, Veron et al. 519 (2012) and Ortiz-Suslow et al. (2016). We used the wind friction velocity as the control 520 parameter since this allows direct comparison with earlier data. The data points of Fairall et al. 521 (2009), Veron et al. (2012) and Ortiz-Suslow et al. (2016), reproduced in Fig. 13, confirm 522 that they observed the presence of the giant droplets with sizes of hundreds of micrometers in 523 the air flow, in agreement with the bag breakup SGF. The earlier SGF suggested by Iida et al. 524 (1992) shows a very slow decrease for droplet radii exceeding 200 µm, which could also 525 indicate the presence of the giant droplets. The absolute values of SGFs are within the 526 experimental uncertainty of the data of Ortiz-Suslow et al. (2016), estimated by the authors as 527 one order of magnitude (see Fig. 10a in Ortiz-Suslow et al., 2016). Note that the difference 528 between the data of Veron et al. (2012) and Ortiz-Suslow et al. (2016) for similar values of u_* 529 is about one order of magnitude, which can be explained by the difference in the conditions 530 of the experiments and the transformation used to infer the SGF from measurements of 531 droplet concentration at different levels (see Ortiz-Suslow et al., 2016). Fig. 13 shows that the 532 main difference of the bag breakup SGF from the data of Veron et al. (2012) and Ortiz-533 Suslow et al. (2016) is an overestimation of numbers of droplets with radii above 250-534 $300 \,\mu\text{m}$ and an underestimation of the number of droplets with radii below $200-250 \,\mu\text{m}$. This 535 may be due to several factors. First of all, strictly speaking, in Fig. 13 we are comparing

536 different features. Veron et al. (2012) and Ortiz-Suslow et al. (2016) did not directly measure 537 the number of droplets ejected from the water surface as assumed by the bag breakup SGF, 538 but, rather, they estimated the number of drops injected from a certain level in terms of a 539 certain model, namely 2.5 H_s in Ortiz-Suslow et al. (2016) and H_s in Veron et al. (2012), 540 where H_S is the significant wave height. At these levels, the concentration of the largest 541 droplets may be significantly lower versus ejection from the surface. This hypothesis is indirectly confirmed by the lower SGF in Ortiz-Suslow et al. (2016) compared to Veron et al. 542 543 (2012). Underestimation of the number of droplets with radii less than 200–250 µm by the 544 bag breakup SGF can be explained by a contribution from alternative mechanisms of spray 545 production (e.g., bursting of large underwater bubbles described in Section 3.2), which can be 546 more effective in producing smaller droplets. Notice, that the values of SGFs, retrieved from 547 the data of Fairall et al. (2009) significantly exceed those in Ortiz-Suslow et al. (2016) and 548 Veron et al. (2012). Similarly, the bag breakup SGF predicts lower values in comparison with 549 the data of Fairall et al. (2009). Possibly these differences originate from the peculiarities of 550 the mixed regime of surface waves in the experiments by Fairall et al. (2009) combined paddle-generated waves with wind waves. Alternatively, in the present experiments and in 551 552 the experiments by Ortiz-Suslow et al. (2016) and Veron et al. (2012), pure wind wave 553 regime was used.

We have also verified the bag breakup SGF against data obtained in the field. Among numerous SGFs for spume droplets (see references in Veron (2015) and Andreas (2002)) we selected those suggested by Andreas (1998), Fairall et al. (1994) and Zhao et al. (2006), which are close in magnitude and fit the criteria of reliability suggested by Andreas (2002). The bag breakup SGF was calculated using Eq. (24) with the specific number of bag breakup events defined by Eqs. (7) and (9). Figs. 14a–b show quite good correspondence between our "theoretical+lab-experiment SGF" and the "empirical SGFs" of Andreas (1998), Fairall et al. 561 (1994), and Zhao et al. (2006) in the radius interval $30\mu < r < 300\mu$ m. It is by no means 562 surprising that giant rim droplets with r > 300mm are missed in SGFs by Andreas (1998), 563 Fairall et al. (1994), and Zhao et al. (2006), where SGFs were derived through extrapolation 564 of data obtained at winds below 20 m/s (cf. Andreas, 1992), when the bag breakup 565 mechanism was not activated.

566

567 **7. Summary**

568 This paper focuses on the mechanisms of production of spume droplets, i.e., sea spray 569 torn off wave crests by wind. The study is based on laboratory experiments in a high-speed 570 wind-wave flume, where measurements were performed for airflow with a friction velocity in 571 the range 0.8–1.5 m/s. According to data by Powell et al. (2003) and Richter et al. (2016), 572 this corresponds to the typical range of turbulent shear stresses observed in the turbulent 573 boundary layer under severe tropical storm and hurricane conditions. High-speed video 574 capture enabled us to investigate how droplets are torn from the crests of surface waves. 575 Since the typical timescale of this process is a few milliseconds, frame rates from 4,500 fps to 576 10,000 fps were used. Capturing video from two directions (side and top view) enabled us to 577 study the process of spume droplet generation in detail. The video revealed that the 578 generation of spume droplets near the wave crest is caused by several local phenomena. It is 579 possible to classify the observed phenomena into three types. One is the development of 580 liquid ligaments, mainly on the crests of breaking waves, which stretch and then break into 581 droplets, as previously observed by Koga (1981). A second is the production of spume 582 droplets through the bursting of submerged bubbles, as was investigated in detail by Lhuissier 583 and Villermaux (2012). The third effective mechanism of spume droplet generation was bag 584 breakup, first observed by Veron et al. (2012). During a bag breakup event, an increase in the

585 small-scale elevation of the water surface results in the formation of a kind of small sail, 586 which is then inflated into a canopy bordered by a thicker rim and finally ruptures to produce 587 spray.

588 Statistical analysis of the videos showed that bag breakup is the dominant mechanism 589 of spume droplet generation at high winds, when u_* exceeds approximately 1.0 m/s. This 590 mechanism was therefore studied in more detail. The dependency of the specific number of 591 bag breakup events, $\langle N \rangle$ (per unit area per unit time), on wind friction velocity, u_* , in the 592 turbulent boundary layer was derived from the processing of top-view high-speed video 593 frames. These statistics were interpreted using a phenomenological approach based on the 594 methods of statistical physics, specifically the Gibbs canonical ensemble, and the function for $\langle N \rangle$ in terms of u_* was then derived. Expressing $\langle N \rangle$ in terms of the windsea Reynolds number, 595 $\operatorname{Re}_{B} = \frac{u_{*}^{2}}{\omega_{v}V}$, yields the dependence on wind fetch required for extrapolation to field 596 597 conditions. The statistical distributions of the sizes, speeds, and lifetimes of the observed bags 598 at different airflow velocities were also investigated, and it was found that all the probability 599 density functions could be well-approximated by the gamma distribution with different 600 parameters.

601 The statistics obtained for the bag breakup events were used to construct an SGF for 602 the bag breakup mechanism of spray production. First, we took into account that the droplets 603 from a bag are generated via two mechanisms: rupture of the canopy of the inflated bag (the 604 canopy droplets) and fragmentation of the rim that remains after fragmentation of the canopy 605 (rim droplets). To obtain the size distribution of the canopy droplets, we used the results of a 606 detailed study by Lhuissier and Villermaux (2012) on spray generation through the bursting 607 of submerged bubbles, which is very similar to rupture of the canopy of the bag, since both 608 these phenomena are governed by surface tension. To derive the size distribution of the rim

609 droplets, we used the similarity of the bags observed near the crests of breaking surface 610 waves and the development of liquid droplets in gaseous flows in the bag breakup regime. 611 We used the geometrical parameters of bags that have been thoroughly investigated by Chou 612 and Faeth (1998) and the statistics of droplets produced by fragmentation of liquid ligaments 613 suggested in Marmottant and Villermaux (2004). The resultant bag breakup SGF was the sum 614 of the generation of both functions for the canopy and rim droplets. The maxima of these 615 functions slightly decrease with wind friction velocity and are significantly different: for 616 u = 1-2 m/s, the maximum of the canopy part of the SGF as the volume flux corresponds to a 617 drop radius of approximately 100 µm, while rim fragmentation producing giant droplets has a 618 maximum corresponding to approximately 1000 µm.

619 The bag breakup SGF derived from our laboratory data was compared with spume 620 droplet SGFs designed for application under both field and laboratory conditions. We 621 compared with the SGFs of Iida et al. (1992), Veron et al. (2012), and Ortiz-Suslov et al. 622 (2016), which were derived from specially designed laboratory experiments. This comparison 623 showed a reasonable agreement of the bag breakup SGF with the data of Veron et al. (2012) 624 and Ortiz-Suslov et al. (2016), although the bag breakup SDF predicts a larger average 625 droplet size. We propose that this may be explained by a lower number of the largest droplets 626 at the measuring points used in the experiments of Veron et al. (2012) and Ortiz-Suslov et al. 627 (2016). The SGF suggested by Iida et al. (1992) shows a very slow decrease for droplet radii 628 exceeding 200 µm, which may also indicate the presence of giant droplets.

We also compared the bag breakup SGF with SFGs developed for field conditions. We employed the bag breakup SGF rescaled to field conditions using a parameterization of the number of bag breakup phenomena by the windsea Reynolds number, as suggested by Toba and Koga (1986). In our estimates, we assumed that the inverse wave age parameter under field conditions, $\Omega = U_{10}/c_n$, was equal to 2.5–3.5 in accordance with the field data of Wright et al. (2001) and used a surface drag coefficient taken from Holthuijsen et al. (2012). The bag breakup SGF was in reasonable agreement in magnitude with SFGs by Andreas (1992, 1998) and Fairall et al. (1994). We suggested two versions of the model of the fetch dependence of the specific numbers of the bag-breakup events and related SGF, Eq. (7) and Eq.(9). Now the data are not sufficient to give advantage to one of these expressions, however Eq.(7) looks promising, because it predicts realistic essential spray production at wind speeds 10-15 m/s.

641 The agreement of the bag breakup SGF with both lab and field data confirms our 642 basic hypothesis regarding the dominant role of bag breakup in hurricanes. This result has 643 numerous prospective applications and forms a new basis for modeling the sea-spray and air-644 sea fluxes. For example, the effect of bag breakup on water-surface resistance can explain the 645 non-monotonous dependence of the surface drag coefficient on wind speed in hurricane 646 winds. Besides, the boosting of exchange processes by giant droplets could be responsible for 647 the significant increase in the air-sea enthalpy flux at high winds that is needed to explain the 648 fast development of intensive hurricanes. These questions are the subject of part II of this 649 study.

650

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Appendix A. Statistics of bag breakup events: the Gibbs canonical ensemble approach

662

This is a summary of the material in the Supplement to Troitskaya et al. (2017).

Originally, the concept of the Gibbs canonical ensemble was developed for a thermodynamic system in weak thermal contact with a heat bath, i.e., an environment that remains at an unchanged temperature due to the negligible feedback of the thermodynamic system to the heat bath. The statistics of the system are determined using the Gibbs or canonical distribution, and the probability that the energy of the system is in the range [E, E+ dE] is

669 $dW = A\exp(-E/\vartheta)dE$.

670 The factor A=1/9, according to the normalization condition.

However, the Gibbs method can be applied universally (see Rumer and Ryvkin, 1980) 671 672 and here it is applied to describing the statistics of bags. We consider the canonical ensemble, 673 which consists of all energy states of the air-ocean interface including those where it can 674 potentially be transformed into bags and then atomized into spray. The analog of the heat 675 bath is the marine atmospheric boundary layer (MABL), which prescribes the state of the 676 interface. Note that the comparatively weak feedback of the state of the air-sea interface to the state of the MABL (an analog of the weakness of the thermal contact of the 677 678 thermodynamic system to the heat bath) is provided by the small (4-5%, according to 679 Golitsyn (2001)) fraction of wind energy spent to generate surface waves and accompanying 680 phenomena in comparison with turbulent dissipation in the MABL.

The heat bath parameter, \mathcal{G} , can be derived from the Boussinesq (1877) analogy between the velocity fluctuations in a turbulent flow and the thermal motions of molecules in a gas. For molecular motion, according to Gibbs (1902), \mathcal{G} is proportional to the temperature of the heat bath, i.e., to the average kinetic energy of the molecules. In the problem being considered here, the analog to the temperature of the gas is the kinetic energy density of turbulent fluctuations in the MABL, where the latter is scaled by the wind friction velocity, i.e., $\vartheta \sim u_*^2$.

688 When the energy of the system under consideration, *E*, exceeds the threshold, *E*₀, of 689 activation of the bag breakup regime, the number of bags arising per unit time per unit area of 690 the water surface is a function of the energy state of the air-ocean interface *N*(*E*), and the 691 average specific number of bags $\langle N \rangle$ is equal to the integral of *N*(*E*)*dW* over all states with 692 energy exceeding the activation threshold, *E*₀. In the vicinity of the threshold, 693 $N(E) = N'(E_0)(E - E_0)$ and integrating for *N*(*E*)*dW* yields $\langle N \rangle = N'(E_0)\mathcal{P}e^{-\frac{E_0}{\mathcal{P}}}$. Since $\mathcal{P} \sim u_*^2$,

694
$$\langle N \rangle (u_*) = Q_0 u_*^2 \exp\left(-\frac{U_0^2}{u_*^2}\right),$$

695 where Q_0 and U_0 are empirical constants to be derived by fitting of the data.

696

697 Appendix B. Derivation of the SGF for the canopy droplets

The total number of droplets with radii in the range $[r, r+\Delta r]$ produced by rupture of the canopy of a bag per unit area per unit time is the convolution of the size spectra of bags (Eq. (16)) with the size spectra of droplets generated due to the rupture of the canopy of the inflated bag (Eq. (17)):

$$\frac{dF_{c}(r,u_{*})}{dr} = \frac{\langle N \rangle(u_{*})}{\langle R_{2} \rangle(u_{*})} \times \\
\times \int_{0}^{\infty} \frac{n^{n}}{\Gamma(n)} \left(\frac{R}{\langle R_{2} \rangle(u_{*})}\right)^{n-1} e^{-\frac{nR}{\langle R_{2} \rangle(u_{*})}} N_{film} \left(R\right) \frac{m^{m}}{\Gamma(m)} \left(\frac{r}{\langle r \rangle(R)}\right)^{m-1} e^{-\frac{mr}{\langle r \rangle(R)}} \frac{dR}{\langle r \rangle(R)}$$
(B1)

- Here, the dependence $\langle R_2 \rangle (u_*)$ is given by Eq. (12), $\langle r \rangle (R)$ by Eq. (21), n=7.53, m=11. The
- 104 limits of integration are chosen equal to 0 and ∞ because the Gamma distribution is a well-
- 705 localized function that decreases exponentially at infinity.
- 70

Substituting expressions for
$$\langle r \rangle (R)$$
 and $N_{film}_{drops}(R)$ from Eqs. (21) and (22) to Eq. (B1)

and replacing the variables in the integrand, $R = \langle R_2 \rangle (u_*) x$, gives:

$$\frac{dF_{c}(r,u_{*})}{dr} = \langle N \rangle (u_{*}) \frac{n^{n}m^{m}}{\Gamma(n)\Gamma(m)} \left(\frac{n}{m}\right)^{m-1} \frac{b}{aL} \left(\frac{\langle R_{2} \rangle (u_{*})}{L}\right)^{\beta-\alpha} \left(\frac{r}{\delta}\right)^{\frac{m+n-\alpha+\beta-1}{\alpha+1}} \times \\
\times \int_{0}^{\infty} x^{n+\beta-\alpha m-1} \exp\left(-n\left(\frac{r}{\delta}\right)^{\frac{1}{\alpha+1}} (x+x^{-\alpha})\right) dx$$
(B2)

709 where
$$\delta = \frac{na(\langle R_2 \rangle (u_*))^{\alpha}}{mL^{\alpha-1}}$$

- 710 Approximate integration of Eq. (B2) using the method of steepest descent (cf., e.g. Nayfeh,
- 711 1981) is applicable if $n\eta >>1$, where $\eta = (r/\delta)^{1/(\alpha+1)}$. In this case, the factor in Eq. (B2),

712
$$G(\eta) = \eta^{m+n-\alpha+\beta-1} \int_{0}^{\infty} x^{n+\beta-\alpha m-1} \exp\left(-n\eta\left(x+x^{-\alpha}\right)\right) dx, \qquad (B3)$$

713 is transformed to the following simple expression:

714
$$G(\eta) \approx G_0 \eta^{m+n-\alpha+\beta-\frac{3}{2}} \exp\left(-g_1 \eta + \frac{g_2}{\eta}\right), \tag{B4}$$

715 with

716
$$G_{0} = \alpha^{\frac{n+\beta-\alpha m-1/2}{\alpha+1}} \sqrt{\frac{2\pi}{n(\alpha+1)}}; \ g_{1} = n(\alpha+1)\alpha^{-\frac{\alpha}{\alpha+1}}; \ g_{2} = \frac{(n+\beta-\alpha m)^{2}}{2n(\alpha+1)}\alpha^{-\frac{1}{\alpha+1}}$$
(B5)

717 Selection of the parameters G_0 , g_1 and g_2 in Eq. (B4) by best fitting of the exact expression 718 Eq. (B3) for $G(\eta)$, namely $G_0=0.46$, $g_1=15.12$, $g_2=0.44$, provides an accurate approximation 719 of the factor $G(\eta)$ that is applicable to all relevant values of η . 721 Eq. (B2) gives:

720

$$\frac{dF_{c}(r,u_{*})}{dr} = \langle N \rangle (u_{*}) \frac{n^{n}m^{m}}{\Gamma(n)\Gamma(m)} \left(\frac{n}{m}\right)^{m-1} \frac{b}{aL} \left(\frac{\langle R_{2} \rangle (u_{*})}{L}\right)^{\beta-\alpha} \left(\frac{r}{\delta}\right)^{\frac{m+n-\alpha+\beta-3/2}{\alpha+1}} \times G_{0} \exp\left(-g_{1}\left(\frac{r}{\delta}\right)^{\frac{1}{(\alpha+1)}} + g_{2}\left(\frac{\delta}{r}\right)^{\frac{1}{(\alpha+1)}}\right)$$

$$723 \quad \text{where } \delta = \frac{na\left(\langle R_{2} \rangle (u_{*})\right)^{\alpha}}{mL^{\alpha-1}}.$$

Simple algebra yields the following symmetrical form of the exponent:

$$\frac{dF_{c}(r,u_{*})}{dr} = \langle N \rangle (u_{*}) \frac{n^{n}m^{m}}{\Gamma(n)\Gamma(m)} \left(\frac{n}{m}\right)^{m-1} \frac{b}{aL} \left(\frac{\langle R_{2} \rangle (u_{*})}{L}\right)^{\beta-\alpha} \left(\frac{r}{\theta}\right)^{\frac{m+n-\alpha+\beta-3/2}{\alpha+1}} \times \\ \times G_{0} \left(\frac{g_{2}}{g_{1}}\right)^{\frac{m+n-\alpha+\beta-3/2}{2}} \exp\left(-2\sqrt{g_{1}g_{2}}\sinh\left(\frac{1}{\alpha+1}\ln\frac{r}{\theta}\right)\right) \qquad ,$$

726 where

727
$$\theta = \frac{na(\langle R_2 \rangle (u_*))^{\alpha}}{mL^{\alpha-1}} \left(\frac{g_2}{g_1}\right)^{\frac{\alpha+1}{2}}.$$

728 Substituting numbers for n=7.53, m=11,

729
$$a = 0.094, \ \alpha = 4/3 \ b = 2.24 \cdot 10^{-3} \left(\frac{\rho_w g L^2}{\sigma}\right)^{1.18}, \ \beta \approx 4/3, \ G_0 = 0.46, \ g_1 = 15.12, \ \text{and} \ g_2 = 0.44$$

finally gives:

731
$$\frac{dF_c(r,u_*)}{dr} = \frac{3.3 \cdot 10^{-9}}{L} \langle N \rangle (u_*) \left(\frac{\rho_w g L^2}{\sigma}\right)^{1.18} \left(\frac{r}{\theta}\right)^{7.3} e^{-5.2 \sinh\left(\frac{3}{7}\ln\frac{r}{\theta}\right)}, \tag{B6}$$

732
$$\theta = \frac{0.001(\langle R_2 \rangle (u_*))^{4/3}}{L^{1/3}}.$$
 (B7)

733

735 Appendix C. Derivation of the expression for the SGF for rim droplets

The total number of rim droplets with radii in the range $[r, r+\Delta r]$ produced by bags per unit area per unit time is the convolution of the size spectra of bags (Eq. (16)) with the size spectra of droplets generated due to the fragmentation of the rim (Eq. (23)):

$$739 \qquad \frac{dF_{rim}(r,u_*)}{dr} = \frac{\langle N \rangle(u_*)N_{rim}}{\langle R_2 \rangle(u_*)} \int_0^\infty \frac{n^n}{\Gamma(n)} \left(\frac{R}{\langle R_2 \rangle(u_*)}\right)^{n-1} e^{-\frac{nR}{\langle R_2 \rangle(u_*)}} \frac{k^k}{\Gamma(k)} \left(\frac{r}{\gamma R}\right)^{k-1} e^{-\frac{kr}{\gamma R}} \frac{dR}{\gamma R}$$

740 Replacing the variables in the integrand, $R = \langle R_2 \rangle (u_*) x$, yields:

$$741 \qquad \frac{dF_{rim}(r,u_*)}{dr} = \frac{\langle N \rangle(u_*)N_{rim}}{\gamma \langle R_2 \rangle(u_*)} \frac{n^n}{\Gamma(n)} \frac{k^k}{\Gamma(k)} \left(\frac{k}{n}\right)^{\frac{n-k}{2}} \left(\frac{r}{\gamma \langle R_2 \rangle(u_*)}\right)^{\frac{n+k}{2}-1} \int_0^\infty x^{n-k-1} e^{-\sqrt{\frac{knr}{\gamma \langle R_2 \rangle(u_*)}} \left(x+\frac{1}{x}\right)} dx$$

742

743 (C1)

Introducing the new variable, $\varepsilon = \sqrt{r/\Delta}$, where $\Delta = \gamma \langle R_2 \rangle (u_*)$, and using the method of steepest descent, similarly to Appendix B, we can transform the factor

746
$$\Phi(\varepsilon) = \varepsilon^{n+k-2} \int_{0}^{\infty} x^{n-k-1} e^{-\varepsilon \sqrt{kn} \left(x+\frac{1}{x}\right)} dx \text{ in (C1) to}$$
747
$$\Phi_{app}(\varepsilon) = \Phi_{0} \varepsilon^{n+k-\frac{5}{2}} e^{-\phi_{0}\varepsilon+\frac{\phi_{1}}{\varepsilon}},$$
(C2)

748 with

749
$$\phi_1 = 2\sqrt{kn}, \ \phi_2 = \frac{(n-k)^2}{4\sqrt{kn}}, \ \Phi_0 = \sqrt{\frac{\pi}{\sqrt{kn}}}.$$
 (C3)

750 Similarly to in Appendix B, we can find ϕ_1 , ϕ_2 , and Φ_0 , which enables an accurate fit for $\Phi(\varepsilon)$

- 751 by use of function (C2): $\phi_1 = 11.07$, $\phi_2 = 0.35$, and $\Phi_0 = 1.02$.
- 752 Substituting Eq. (C2) into Eq. (C1) gives:

753
$$\frac{dF_{rim}(r,u_{*})}{dr} = \frac{\langle N \rangle(u_{*})N_{rim}}{\Delta} \frac{n^{n}}{\Gamma(n)} \frac{k^{k}}{\Gamma(k)} \left(\frac{k}{n}\right)^{\frac{n-k}{2}} \left(\frac{r}{\Delta}\right)^{\frac{n-k}{2}-\frac{5}{4}} \Phi_{0} \exp\left(-\phi_{1}\left(\frac{r}{\Delta}\right)^{\frac{1}{2}} + \phi_{2}\left(\frac{\Delta}{r}\right)^{\frac{1}{2}}\right)$$
(C4)

Simple algebra yields the symmetrical form of the exponent in Eq. (C4):

$$755 \qquad \frac{dF_{rim}(r,u_*)}{dr} = \frac{\langle N \rangle(u_*)N_{rim}}{\Theta} \frac{n^n}{\Gamma(n)} \frac{k^k}{\Gamma(k)} \left(\frac{k}{n}\right)^{\frac{n-k}{2}} \left(\frac{r}{\Theta}\right)^{\frac{n+k}{2}-\frac{5}{4}} \Phi_0\left(\frac{\phi_2}{\phi_1}\right)^{\frac{n+k}{2}-\frac{1}{4}} e^{-2\sqrt{\phi_1\phi_2}\sinh\left(\frac{1}{2}\ln\frac{r}{\Theta}\right)},$$

756 where $\Theta = \gamma \langle R_2 \rangle (u_*) \phi_2 / \phi_1$.

757 Substituting numbers for n=7.53, k=4, $\gamma=0.068$, $\phi_I=11.07$, $\phi_2=0.35$, and $\Phi_0=1.02$ finally 758 yields:

759
$$\frac{dF_{rim}(r,u_*)}{dr} = \langle N \rangle (u_*) \frac{1.5 \cdot 10^{-4} N_{rim}}{\Theta} \left(\frac{r}{\Theta}\right)^{4.5} e^{-3.94 \sinh\left(\frac{1}{2}\ln\frac{d}{\Theta}\right)},$$
(C5)

760
$$\Theta = 0.0021 \langle R_2 \rangle (u_*).$$
 (C6)

762 **References**

- Andreas, E. L., 1992: Sea spray and the turbulent air-sea heat fluxes. J. Geophys. Res., 97,
- 764 11429–11441, https://doi.org/10.1029/92JC00876.
- Andreas, E. L., 1998: A New Sea Spray Generation Function for Wind Speeds up to 32 m s^{-1} .
- 766 J. Phys. Oceanogr., 28, 2175–2184, https://doi.org/10.1175/1520-
- 767 0485(1998)028<2175:ANSSGF>2.0.CO;2.
- Andreas, E. L., 2002: A review of spray generation function for the open ocean. Atmosphere-
- 769 Ocean Interactions Volume 1, Perrie, W., Ed., WIT Press, 1–46.
- Andreas, E. L., 2011: Fallacies of the Enthalpy Transfer Coefficient over the Ocean in High
- 771 Winds. J. Atmos. Sci., 68, 1435–1445, https://doi.org/10.1175/2011JAS3714.1.
- Andreas, E. L., and K. A. Emanuel, 2001: Effects of Sea Spray on Tropical Cyclone Intensity.
- 773 J. Atmos. Sci., 58, 3741–3751, https://doi.org/10.1175/1520-
- 774 0469(2001)058<3741:EOSSOT>2.0.CO;2.
- 775 Bao, J.-W., C. W., Fairall, S. A. Michelson, and L. Bianco: 2011: Parameterizations of sea-
- spray impact on the air-sea momentum and heat fluxes. *Mon. Weather Rev.*, **139**, 3781–3797,
- 777 https://doi.org/10.1175/MWR-D-11-00007.1.
- 778 Bianco, L., J.-W. Bao, C. W. Fairall, and S. A. Michelson, 2011: Impact of sea spray on the
- surface boundary. Bound.-Layer Meteor., 140, 361–381, https://doi.org/10.1007/s10546-011-
- 780 9617-1.
- 781 Blanchard, D. C., 1963: The electrification of the atmosphere by particles from bubbles in the
- 782 sea. *Prog. Oceanogr.*, **1**, 71–202, https://doi.org/10.1016/0079-6611(63)90004-1.
- 783 Boussinesq, J. V., 1877: Essai sur la theorie des eaux courantes. Mémoires présentés par
- 784 Divers savants à l'Acad. des Sci. Inst. Nat. Fr., XXIII, 1–680.

Chou W. H., and G. M. Faeth, 1998: Temporal properties of secondary drop breakup in the
bag breakup regime. *Int. J. Multiphase Flow*, 24, 889–91, https://doi.org/10.1016/S0301-

787 9322(98)00015-9.

- Fairall, C. W., J. D. Kepert, and G. J. Holland, 1994: The effect of sea spray on surface
- energy transports over the ocean, *Global Atmos. Ocean Syst.*, **2**, 121–142.
- 790 Fairall, C. W., M. L. Banner, W. L. Peirson, W. Asher, and R. P. Morison, 2009:
- 791 Investigation of the physical scaling of sea spray spume droplet production, J. Geophys. Res.
- 792 *Ocean.*, **114**, C10001, doi.org/10.1029/2012JC007983.
- Foreman, R. J., and S. Emeis, 2010: Revisiting the Definition of the Drag Coefficient in the
- 794 Marine Atmospheric Boundary Layer. J. Phys. Oceanogr., 40, 2325–2332,
- 795 https://doi.org/10.1175/2010JPO4420.1.
- Gelfand, B. E., 1996: Droplet breakup phenomena in flows with velocity lag. *Prog. Energ.*
- 797 *Combust. Sci.*, **22**, 201–265, https://doi.org/10.1016/S0360-1285(96)00005-6.
- Gibbs, J. W., 1902: *Elementary Principles in Statistical Mechanics*. Charles Scribner's Sons,
 239 pp.
- 800 Golitsyn, G. S., 2010: The energy cycle of wind waves on the sea surface *Izv. Atmos. Ocean.*
- 801 *Phys.*, **46**, 6-13, https://doi.org/10.1134/S0001433810010020.
- 802 Hinze, J.O., 1959: *Turbulence*. 2nd Edition, McGraw Hill, 586 pp.
- 803 Holthuijsen, L. H., M. D. Powell, and J. D. Pietrzak, 2012: Wind and waves in extreme
- 804 hurricanes. J. Geophys. Res. Ocean., 117, C09003, https://doi.org/10.1029/2012JC007983.
- 805 Iida, N., Y. Toba, and M. Chaen, 1992: A new expression for the production rate of sea water
- 806 droplets on the sea surface. J. Oceanogr., **48**, 439–460, https://doi.org/10.1007/BF02234020.
- 807 Koga, M. 1981: Direct production of droplets from breaking wind-waves—its observation by

- 808 a multi-colored overlapping exposure photographing technique. *Tellus*, **33**, 552–563,
- 809 https://doi.org/10.3402/tellusa.v33i6.10776.
- 810 Lhuissier, H., and E. Villermaux, 2009: Bursting bubbles. *Phys. Fluids*, **21**, 091111,
- 811 https://doi.org/10.1063/1.3200933.
- 812 Lhuissier, H., and E. Villermaux, 2012: Bursting bubble aerosols. J. Fluid Mech., 696, 5–44,
- 813 https://doi.org/10.1017/jfm.2011.418.
- Marmottant, P., and E. Villermaux, 2004: On spray formation. *J. Fluid Mech.*, 498, 73–111,
 https://doi.org/10.1017/S0022112003006529.
- 816 Mueller, J. A., and F. Veron, 2009: A Sea State–Dependent Spume Generation Function. J.
- 817 *Phys. Oceanogr.*, **39**, 2363–2372, https://doi.org/10.1175/2009JPO4113.1.
- 818 Nayfeh, A. H., 1981: Introduction to perturbation techniques. Wiley & Sons, 666 pp.
- 819 Ortiz-Suslow, D. G., B. K. Haus, S. Mehta, and N. J. Laxague, 2016: Sea Spray Generation in
- 820 Very High Winds. J. Atmos. Sci. 73, 3975–3995, https://doi.org/10.1175/JAS-D-15-0249.1.
- 821 Petersen, P., 1927: Zur Bestimmung der Windstärke auf See. Für Segler, Dampfer und
- 822 Luftfahrzeige. Annalen der Hydrographie und Maritimen Meteorologie, 55, 69-72.
- 823 Powell, M. D., P. J. Vickery, and T. A. Reinhold, 2003: Reduced drag coefficient for high
- wind speeds in tropical cyclones. *Nature*, **422**, 279–283, https://doi.org/10.1038/nature01481.
- 825 Richter, D. H., R. Bohac, and D. P. Stern, 2016: An assessment of the flux profile method for
- 826 determining air-sea momentum and enthalpy fluxes from dropsonde data in tropical cyclones.
- 827 J. Atmos. Sci., **73**, 2665–2682, https://doi.org/10.1175/JAS-D-15-0331.1.
- 828 Rumer Yu. B. and M. S. Ryvkin, 1980: Thermodynamics, statistical physics, and kinetics
- 829 (English translation). Mir, 600 pp.

- 830 Soloviev, A. V, R. Lukas, M. Donelan, B. K. Haus, and I. Ginis, 2014: The air-sea interface
- and surface stress under tropical cyclones. Sci. Rep. 4, 5306,
- 832 https://doi.org/10.1038/srep05306.
- 833 Spiel, D. E., 1994a: The number and size of jet drops produced by air bubbles bursting on a
- fresh water surface. J. Geophys. Res., 99, 10289–96, https://doi.org/10.1029/94JC00382.
- 835 Spiel, D. E., 1994b: The sizes of jet drops produced by air bubbles bursting on sea- and fresh-
- 836 water surfaces. *Tellus B*, **46**, 4, 325–338, https://doi.org/10.3402/tellusb.v46i4.15808.
- 837 Spiel, D. E., 1995: On the births of jet drops from bubbles bursting on water surfaces. J.
- 838 *Geophys. Res.*, **100**, 4995–5006, https://doi.org/10.1029/94JC03055.
- 839 Spiel, D. E., 1997: More on the births of jet drops from bubbles bursting on seawater surfaces.
- 840 J. Geophys. Res., 102, 5815–5821, https://doi.org/10.1029/96JC03582.
- 841 Spiel DE., 1998: On the births of film drops from bubbles bursting on seawater surfaces. J.
- 842 *Geophys. Res.*, **103**, 24907–24918, https://doi.org/10.1029/98JC02233.
- 843 Takagaki, N., S. Komori, N. Suzuki, K. Iwano, T. Kuramoto, S. Shimada, R. Kurose, and K.
- 844 Takahashi, 2012: Strong correlation between the drag coefficient and the shape of the wind
- sea spectrum over a broad range of wind speeds. *Geophys. Res. Lett.*, **39**,
- 846 https://doi.org/10.1029/2012GL053988.
- 847 Takagaki, N., S. Komori, N. Suzuki, K. Iwano, and R. Kurose, 2016: Mechanism of drag
- 848 coefficient saturation at strong wind speeds. *Geophys. Res. Lett.*, **43**, 9829–9835,
- 849 https://doi.org/10.1002/2016GL070666.
- 850 Takagaki, N., S. Komori, M. Ishida, K. Iwano, R. Kurose, and N. Suzuki, 2017: Loop-Type
- 851 Wave-Generation Method for Generating Wind Waves under Long-Fetch Conditions. J.
- 852 Atmos. Oceanic Technol., **34**, 2129–2139, https://doi.org/10.1175/JTECH-D-17-0043.1.

- Toba, Y. and M. Koga, 1986: A parameter describing overall conditions of wave breaking,
- 854 whitecapping, sea-spray production and wind stress. Oceanic Whitecaps, E.C. Monahan and
- 855 G. MacNiocaill, Eds., D. Reidel Publishing Company, 37–47.
- Toba, Y., S. Komori, Y. Suzuki, and D. Zhao, 2006: Similarity and dissimilarity in air-sea
- 857 momentum and CO₂ transfers: The nondimensional transfer coefficients in light of windsea
- 858 Reynolds number, *Atmosphere-Ocean Interactions*, 2, W. Perrie, Ed., WIT Press, 53–82.
- 859 Troitskaya, Y. I., D. A. Sergeev, A. A. Kandaurov, G. A Baidakov, M. A. Vdovin, and V. I.
- 860 Kazakov, 2012: Laboratory and theoretical modeling of air-sea momentum transfer under
- severe wind conditions. J. Geophys. Res. Ocean., 117, C00J21,
- 862 https://doi.org/10.1029/2011JC007778.
- 863 Troitskaya, Y. I., A. Kandaurov, O. Ermakova, D. Kozlov, D. Sergeev, and S. Zilitinkevich,
- 864 2017: Bag-breakup fragmentation as the dominant mechanism of sea-spray production in
- 865 high winds. Sci. Rep., 7, 1614, https://doi.org/10.1038/s41598-017-01673-9.
- 866 Veron F., 2015: Ocean Spray. Annu. Rev. Fluid Mech., 47, 507–538,
- 867 https://doi.org/10.1146/annurev-fluid-010814-014651.
- 868 Veron, F., C. Hopkins, E. L. Harrison, and J. A. Mueller, 2012: Sea spray spume droplet
- production in high wind speeds. *Geophys. Res. Lett.*, **39**, L16602,
- 870 https://doi.org/10.1029/2012GL052603.
- 871 Wright, C. W., and Coauthors, 2001: Hurricane Directional Wave Spectrum Spatial Variation
- 872 in the Open Ocean. J. Phys. Oceanogr., **31**, 2472–2488, https://doi.org/10.1175/1520-
- 873 0485(2001)031<2472:HDWSSV>2.0.CO;2.
- Wu, J., 1981: Evidence of sea spray produced by bursting bubbles. *Science*, **212**, 324.
- 875 Zhao, D., Y. Toba, K. Sugioka, and S. Komori, 2006: New sea spray generation function for
- spume droplets. J. Geophys. Res. Ocean., 111, C02007.

877 Tables

Frequency	Friction velocity	Number	Total
of the fan	(m/s)	of	number of
(Hz)		records	frames
30	0.753	1	33452
31	0.814	1	33452
32	0.874	2	66248
33	0.931	5	164636
34	0.987	15	490996
35	1.04	11	360452
36	1.09	11	360452
37	1.15	7	229588
38	1.20	5	164316
39	1.24	3	98884
40	1.29	3	98884
42	1.38	1	33452
45	1.51	2	66248
	Total:	67	2234512

Table 1. Parameters of the experiments.

879

881 **Figure caption list**

Fig. 1. Wind and wave parameters in the flume. (a) The roughness height due to the friction velocity in the flume, with the best fit from Eq. (1). (b) The dependency of the peak frequency in the wind wave spectra on wind friction velocity, with the best fit from Eq. (2).

- **Fig. 2.** Schematic diagram of experimental setup.
- **Fig. 3.** Generation of droplets through the development and breakage of liquid ligaments. (a) Stretching of the ligament, t=0 ms. (b) Formation of the droplet, t=3.9 ms. (c) Separation of the first droplet and formation of the second, t=9.3 ms. (d) Formation of the third and fourth droplets, t=17.3 ms, wind speed $U_{10}\approx 25$ m/s, image dimensions 67.48×101.13 mm. (e) Spray generation at the crest of the breaking waves (photo taken by Troitskaya at the Gorky reservoir, Volga river, Oct. 1, 2011). Wind speed $U_{10}\approx 9$ m/s. Insets 1 and 2 show the magnified details of the wave crest.
- Fig. 4. The burst of a large bubble in strong wind conditions, $U_{10}=25$ m/s. The top panel: (a) The floating bubble, t=0 ms. (b) Formation of a hole in the liquid film, t=3.2 ms. (c) Expansion of the hole, t=5.1 ms. (d) Droplet formation, t=8.4 ms. The bottom panel is the side view of the bubble burst.
- 897 Fig. 5. The formation and rupture of a bag. A single bag: (a) side view, (b) top view. A 898 "multi-bag": (c) side view, (d) top view. $U_{10}=25$ m/s.
- **Fig. 6.** Schematic diagram of the formation and fragmentation of a bag. (a) Formation of the initial disturbance, (b) increase of the initial disturbance, (c) sail-shaped disturbance, (d) formation of the bag, (e) rupture of the bag. The thin dotted lines are the streamlines in the reference frame following the elevation in the water surface.
- 903 Fig. 7. Semi-automatic registering of the evolution of a bag: (a) initial frame showing bag
 904 nucleation, (b) intermediate frame with annotated markers, edge positions are interpolated,
 905 (c) final frame showing the moment of canopy puncture. 1- bag nucleation, 2 canopy

906 puncture, 3 - manually defined markers. The friction velocity of the airflow is 1.04 m/s, and907 the average wind direction is from top to bottom.

Fig. 8. Dependence of the specific numbers (per unit time per unit area) of the spraygenerating phenomena (a) on the wind friction velocity and (b) on the windsea Reynolds number. Open circles – bursting of floating bubbles, squares - liquid filaments, closed circles – bag breakup events (Fig. 8a is adopted from Troitskaya et al. (2017)). (c) 10-m wind speed versus wave-age parameter $\Omega = U_{10}/c_p$ at a fixed windsea Reynolds numbers, *ReB*, equal to 4000, corresponding to the first appearance of bag breakup (solid curve) and 8000 (dashed curve).

Fig. 9. Approximation of the experimental dependence of specific number of bags on friction velocity (a) using Eq. (5) and (b) on the windsea Reynolds number using Eq.(7). Open symbols are data obtained by processing individual 33,000-frame videos records, and closed symbols are the averaged data; the error bars are defined by the standard deviation. (c) Estimated specific number of bags using field conditions versus U_{10} for the wave-age parameter Ω =2.5 (black curves) and Ω =3.5 (grey curves); solid curves - Eq. (7), dashed curves - Eq. (9).

Fig. 10. (a) The frequency distribution of bag size at the moment of nucleation $(R_1/\langle R_1 \rangle)$ and at the moment of rupture $(R_2/\langle R_2 \rangle)$. (b) The frequency distribution of the velocity of the motion of bag edges $(u_1/\langle u_1 \rangle)$ and centers $(u_2/\langle u_2 \rangle)$. (c) The frequency distribution of bag lifetime $(\tau/\langle \tau \rangle)$.

925 Curves are the Gamma distribution for (a) n = 7.53, (b) n = 13.30, and (c) n = 3.70.

926 Fig. 11. Dependencies of averaged values on the friction velocity of the air flow: (a) the 927 initial (closed circles) and final size (open diamonds) of bags, (b) the velocity of the edges 928 (closed circles) and centers (open diamonds) of bags, and (c) the lifetime of bags. Lines are 929 the power best fit from Eqs. (11-15). (d) Proportionality in the sizes of bags. **Fig. 12.** (a) The bag breakup SGF. (b) The SGFs for the canopy (dash-dot curve) and rim (dashed curve) droplets and their aggregate (gray solid curve) for u = 1.5 m/s. (c) Bag breakup SGF as the volume flux. For (a) and (c), u = 1.5 m/s to 2 m/s with an increment 0.1 m/s.

934 Fig. 13. Comparison of the bag breakup SGF (solid and dashed lines) with empirical 935 estimations of SGFs under laboratory conditions. Symbols: Iida et al. (1992) - open circles; Ortiz-Suslow et al. (2016) - open squares - u = 1.75 m/s ($U_{10} = 36$ m/s), closed circles - u = 1.97936 937 m/s (U_{10} =40.5 m/s), upward-pointing triangles - u_* =2.19 m/s (U_{10} =45 m/s), closed squares -938 u = 2.43 m/s ($U_{10} = 49.5 \text{ m/s}$), downward-pointing triangles - u = 2.66 m/s ($U_{10} = 54 \text{ m/s}$); Veron 939 et al. (2012) - crosses - u = 1.98 m/s ($U_{10} = 41.2$ m/s), X - u = 2.33 m/s ($U_{10} = 47.1$ m/s). Fairall 940 et al. (2009) - thin lines with symbols: open diamonds u = 1.35 m/s, closed diamonds u =941 1.44 m/s, closed triangles u = 1.64 m/s. For the bag breakup SGF, u_* varies between 1.6 m/s 942 and 2.4 m/s with an increment of 0.2 m/s. Solid lines correspond to u*=2 m/s (lower) and 943 u = 2.4 m/s (upper).

Fig. 14. Comparison of the bag breakup SGF with empirical SGFs under field conditions by Andreas (1998) (diamonds), Fairall et al. (1994) (triangles), Zhao et al. (2006) (circles) at U_{10} =30 m/s (a) and U_{10} =35 m/s (b). In the bag breakup SGF, the wave-age parameter Ω=2.5 (black curves) and Ω=3.5 (grey curves); solid curves - Eq. (7), dashed curves - Eq. (9).

Figures



Fig. 1. Wind and wave parameters in the flume. (a) The roughness height due to the friction velocity in the flume, with the best fit from Eq. (1). (b) The dependency of the peak





Fig. 2. Schematic diagram of experimental setup.





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Fig. 3. Generation of droplets through the development and breakage of liquid ligaments. (a) Stretching of the ligament, t=0 ms. (b) Formation of the droplet, t=3.9 ms. (c) Separation of the first droplet and formation of the second, t=9.3 ms. (d) Formation of the third and fourth droplets, t=17.3 ms, wind speed $U_{10}\approx 25$ m/s, image dimensions 67.48×101.13 mm. (e) Spray generation at the crest of the breaking waves (photo taken by Troitskaya at the Gorky reservoir, Volga river, Oct. 1, 2011). Wind speed $U_{10}\approx 9$ m/s. Insets 1 and 2 show the magnified details of the wave crest.

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The floating bubble, *t*=0 ms. (b) Formation of a hole in the liquid film, *t*=3.2 ms. (c)

Expansion of the hole, t=5.1 ms. (d) Droplet formation, t=8.4 ms. The bottom panel is the side view of the bubble burst.





(b)





Fig. 5. The formation and rupture of a bag. A single bag: (a) side view, (b) top view. A "multi-bag": (c) side view, (d) top view. $U_{10}=25$ m/s.



Fig. 6. Schematic diagram of the formation and fragmentation of a bag. (a) Formation of the
initial disturbance, (b) increase of the initial disturbance, (c) sail-shaped disturbance, (d)
formation of the bag, (e) rupture of the bag. The thin dotted lines are the streamlines in the
reference frame following the elevation of the water surface.

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1003	Fig. 7. Semi-automatic registering of the evolution of a bag: (a) initial frame showing bag
1004	nucleation, (b) intermediate frame with annotated markers, edge positions are interpolated
1005	(c) final frame showing the moment of canopy puncture. 1- bag nucleation, 2 - canopy

1006 puncture, 3 - manually defined markers. The friction velocity of the airflow is 1.04 m/s, and

1007 the average wind direction is from top to bottom.



1008 (a) (b) (c) 1009 **Fig. 8.** Dependence of the specific numbers (per unit time per unit area) of the spray-1010 generating phenomena (a) on the wind friction velocity and (b) on the windsea Reynolds 1011 number. Open circles – bursting of floating bubbles, squares - liquid filaments, closed circles 1012 – bag breakup events (Fig. 8a is adopted from Troitskaya et al. (2017)). (c) 10-m wind speed 1013 versus wave-age parameter $\Omega = U_{10}/c_p$ at a fixed windsea Reynolds numbers, Re_B , equal to 1014 4000, corresponding to the first appearance of bag breakup (solid curve) and 8000 (dashed 1015 curve).

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1023 **Fig. 9**. Approximation of the experimental dependence of specific number of bags on friction 1024 velocity (a) using Eq. (5) and (b) on the windsea Reynolds number using Eq.(7). Open 1025 symbols are data obtained by processing individual 33,000-frame records, and closed 1026 symbols are the averaged data; the error bars are defined by the standard deviation. (c) 1027 Estimated specific number of bags using field conditions versus U_{10} for the wave-age 1028 parameter Ω =2.5 (black curves) and Ω =3.5 (grey curves); solid curves - Eq. (7), dashed 1029 curves - Eq. (9).





Fig. 10. (a) The frequency distribution of bag size at the moment of nucleation $(R_1/\langle R_1 \rangle)$ and at the moment of rupture $(R_2/\langle R_2 \rangle)$. (b) The frequency distribution of the velocity of the motion of bag edges $(u_1/\langle u_1 \rangle)$ and centers $(u_2/\langle u_2 \rangle)$. (c) The frequency distribution of bag lifetime $(\tau/\langle \tau \rangle)$. Curves are the Gamma distribution for (a) n = 7.53, (b) n = 13.30, and (c) n = 3.70.



Fig. 11. Dependencies of averaged values on the friction velocity of the air flow: (a) the
initial (closed circles) and final size (open diamonds) of bags, (b) the velocity of the edges
(closed circles) and centers (open diamonds) of bags, and (c) the lifetime of bags. Lines are
the power best fit from Eqs. (11-15). (d) Proportionality in the sizes of bags.



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1042Fig. 12. (a) The bag breakup SGF. (b) The SGFs for the canopy (dash-dot curve) and rim1043(dashed curve) droplets and their aggregate (gray solid curve) for u = 1.5 m/s. (c) Bag1044breakup SGF as the volume flux. For (a) and (c), u = 1.5 m/s to 2 m/s with an1045increment 0.1 m/s.



1048 Fig. 13. Comparison of the bag breakup SGF (solid and dashed lines) with empirical 1049 estimations of SGFs under laboratory conditions. Symbols: Iida et al. (1992) - open circles; 1050 Ortiz-Suslow et al. (2016) - open squares - u = 1.75 m/s ($U_{10} = 36$ m/s), closed circles - u = 1.971051 m/s (U_{10} =40.5 m/s), upward-pointing triangles - u_* =2.19 m/s (U_{10} =45 m/s), closed squares u = 2.43 m/s ($U_{10} = 49.5 \text{ m/s}$), downward-pointing triangles - u = 2.66 m/s ($U_{10} = 54 \text{ m/s}$); Veron 1052 1053 et al. (2012) - crosses - u = 1.98 m/s ($U_{10} = 41.2$ m/s), X - u = 2.33 m/s ($U_{10} = 47.1$ m/s). Fairall 1054 et al. (2009) - thin lines with symbols: open diamonds u = 1.35 m/s, closed diamonds u =1055 1.44 m/s, closed triangles u = 1.64 m/s. For the bag breakup SGF, u_* varies between 1.6 m/s 1056 and 2.4 m/s with an increment of 0.2 m/s. Solid lines correspond to u*=2 m/s (lower) and 1057 u = 2.4 m/s (upper).



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1060 **Fig. 14.** Comparison of the bag breakup SGF with empirical SGFs under field conditions by 1061 Andreas (1998) (diamonds), Fairall et al. (1994) (triangles), Zhao et al. (2006) (circles) at 1062 $U_{10}=30$ m/s (a) and $U_{10}=35$ m/s (b). In the bag breakup SGF, the wave-age parameter $\Omega=2.5$ 1063 (black curves) and $\Omega=3.5$ (grey curves); solid curves - Eq.(7), dashed curves - Eq. (9).