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## 2 **The choice of droplet size probability distribution function for** 3 **oil spill modeling is not trivial**

4

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18

### 19 **ABSTRACT:**

20 The droplet size distribution (DSD) formed by gas-saturated oil jets is one of the most important  
21 characteristics of the flow to understand and model the fate of uncontrolled deep-sea oil spills. The  
22 shape of the DSD, generally modeled as a theoretical lognormal, Rosin-Rammler or non-  
23 fundamental distribution function, defines the size and the mass volume range of the droplets. Yet,  
24 the fundamental DSD shape has received much less attention than the volume median size (d50)  
25 and range of the DSD during ten years of research following the *Deepwater Horizon (DWH)*  
26 blowout. To better understand the importance of the *distribution function* of the droplet size we  
27 compare the oil rising time, surface oil mass, and sedimented and beached masses for different  
28 DSDs derived from the DWH literature in idealized and applied conditions, while keeping d50  
29 constant. We highlight substantial differences, showing that the probability distribution function of  
30 the DSD for far-field modeling is, regardless of the d50, consequential for oil spill response.

1

31

32 **KEYWORDS:** Droplet size distribution; DSD; Oil spill; Oil spill model; Oil model; Deepwater  
33 Horizon; Blowout; d50; Probability Distribution Function

34

35 **MAIN TEXT:**

36 The droplets sizes are key characteristics to predict the transport and fate of uncontrolled deep-sea  
37 oil spills. The 2010 Deepwater Horizon (DWH) blowout released an estimated five million barrels  
38 of live oil into the northern Gulf of Mexico (GoM), impacting the beaches and seafloor (Nixon et al.  
39 2016, Romero et al. 2017). Studies had previously focused on the volume median size of oil  
40 droplets (d50) to understand its effect on emulsion stability (Tambe and Sharma 1994) and,  
41 immediately after the DWH, on the timing and amount of oil surfacing (Ryerson et al. 2012). Ten  
42 years later, there is still an ongoing debate on the correct d50 of the DWH oil droplets, since the d50  
43 estimated at the time of the spill was a decisive factor for the unprecedented use of sub-sea  
44 dispersant injection as a response strategy (Paris et al. 2018). Droplet size distribution (DSD)  
45 estimates have thus been the focus of numerous analytical models, such as dynamical models (Zhao  
46 et al. 2014, 2016, Gros et al., 2017) or equilibrium models with modified Weber number (Johansen  
47 et al., 2013, Li et al. 2017) and of laboratory work under normal atmospheric conditions and  
48 temperature using the Ohmsett tank (Zhao et al. 2016) and under high-pressure and cold-  
49 temperature using sapphire cell and jets experiments with gas-saturated petroleum (Aman et al.,  
50 2015, Malone et al. 2018). Improvement to the understanding of DSD in the jet plume and  
51 subsurface layer was set as a top priority to inform oil spill models (Malone et al. 2018; Lehr and  
52 Socolofsky 2020). However, regardless of the d50, the importance of the fundamental shape, that is  
53 the probability distribution function (PDF) of the DSD, on the rise of the oil from deep-sea blowout  
54 and on its transport has been, so far, overlooked; our study fills this gap.

55

56 While analytical methods predicted a large range of droplet sizes (up to 2.5 mm, Gros et al. 2017;  
57 e.g. Figure 1a), scaled-up laboratory experiments based on turbulent dissipation rates (TDR, Aman  
58 et al. 2005; Pesch et al. 2020a) estimated at the Macondo riser-pipe found smaller droplets (<500  
59  $\mu\text{m}$ , e.g. Figure 1b), similar to lognormal DSD acquired during the few dive operations in the deep

60 plume (1100 m) and in deeper waters at 1400 m (Li et al. 2015, Spaulding et al., 2015). Regardless  
61 of the estimated median droplet size, there is so far no consistency on the PDF of the oil DSD.  
62 Although the most commonly assumed PDFs such as lognormal (LN) and Rosin-Rammler (RR) are  
63 fairly similar in shape, their distributions' edges drastically differ (Figure 1). Here, we quantify the  
64 difference in proportion of oil mass rising at the surface, sedimented on the seafloor, and beached  
65 on the shoreline focusing on the importance of the *distribution function* of the droplet size rather  
66 than the d50. We generated two hypothetical cases in which oil droplets ranged from 1 to 2400  $\mu\text{m}$   
67 (case 1, d50 = 900  $\mu\text{m}$ ) and from 1 to 500  $\mu\text{m}$  (case 2, d50 = 117  $\mu\text{m}$ ), derived from two published  
68 studies which used different droplet models, Gros et al. (2017) and Aman et al. (2015), respectively.  
69 We kept the d50 and the droplet size ranges similar among cases, which thus only differed by their  
70 PDF (Figure 1a,c). For both cases, one dynamic probability distribution function from the  
71 simulation model VDROD-J obtained from Gros et al. (2017) was used as baseline, and two  
72 theoretical distribution functions, the lognormal and Rosin-Rammler distributions, were fitted to the  
73 DSDs from VDROD-J using the R package *fitdistrplus* (Delignette-Muller and Dutang, 2015). The  
74 standard deviation and scale parameters were extracted from the lognormal and Rosin-Rammler  
75 fits, and used to generate the new lognormal and Rosin-Rammler DSDs. The DSD parameters are:  
76 d50 set to 900  $\mu\text{m}$  and 117  $\mu\text{m}$ , log(SD) set to 0.3920 and 0.4297 for the lognormal, and shape set  
77 to 3.5010 and 3.3110 for the case 1 and case 2, respectively).

78  
79 In a first approach, considering idealized conditions, we estimated the maximum potential effect  
80 of the choice of DSD on the rising speed and surfacing time of the oil droplets. We applied an  
81 integrated formulation of buoyant velocity from Zheng and Yapa (2000). The critical diameter  
82  $d_c$ , beyond which Stokes' law does not apply (i.e., Reynolds number  $> 1$ ), which is defined as:

$$d_c = \frac{9.52\mu^{2/3}}{(g\rho\Delta\rho)^{1/3}}$$

83  
84 where  $\mu$  is the dynamic viscosity of the ambient fluid,  $g$  the gravitational acceleration  
85 ( $g=9.8 \text{ m s}^{-2}$ ),  $\rho$  is the ambient fluid density and  $\Delta\rho$  is the absolute difference of density between  
86 oil and seawater. If the spherical droplet diameter is smaller than  $d_c$ , then the terminal velocity  
87 ( $U_T$ ) of the droplet is:

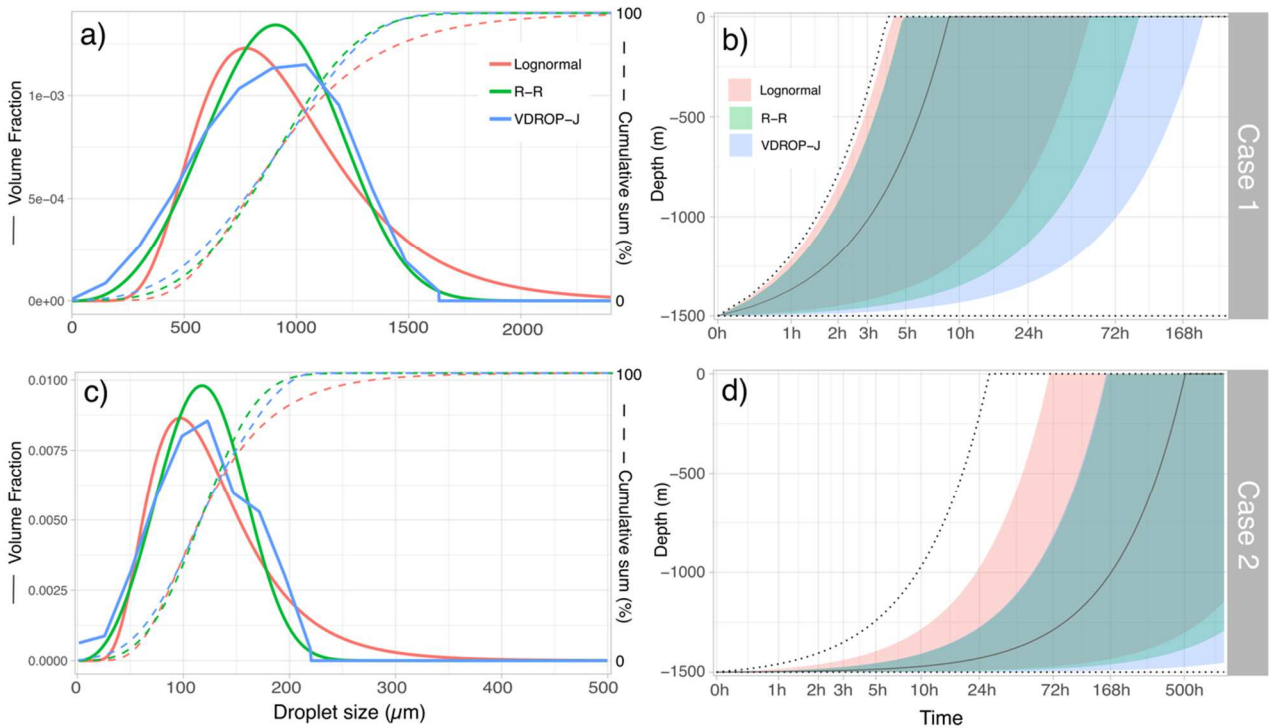
$$U_T = \frac{gd^2\Delta\rho}{18\mu}$$

88

89 When the droplet spherical diameter is larger than  $d_c$ , the terminal velocity of droplets is  
 90 approximated as:

$$U_T = \left( \frac{8gd\Delta\rho}{3\rho} \right)^{1/2}$$

91



92

93 **Figure 1. Distribution, cumulative sum and timing of surfacing of different analytical,**  
 94 **Lognormal and Rosin-Rammler (R-R) droplet size distributions (DSD) fitted on two cases**  
 95 **with different  $d_{50}$  and size ranges (case 1, size range and  $d_{50}$  from Gros et al. 2017 and case 2**  
 96 **from Aman et al. 2015). a)** Probability distribution functions from VDRO-P-J (blue) and the fitted  
 97 lognormal (red) and R-R (green) theoretical distribution laws for the hypothetical case 1 (solid  
 98 lines) and cumulative sums highlighting that all share the same  $d_{50}$  (dashed lines). **b)** Oil droplets'  
 99 vertical distribution for the various DSDs when released at the approximate depth (1500 m) and  
 100 temperature (4°C) of the Macondo well for the case 1 (upper depth: quantile 99%, solid lines:  
 101 median, lower depth: quantile 1%) over time (x-axis is log1p-transformed). The dashed lines show  
 102 the minimum and maximum depth of the oil droplets. **c)** and **d)** Same as a) and b) but for case 2.  
 103 The median droplet depths are similar among PDFs because they have the same  $d_{50}$ .

104

105 Differences in the rise velocities of the bulk of the oil were comparable, as expected from the  
106 design of the simulations which focused on the tails of the PDFs rather than the d50. However,  
107 solely based on the differences between the PDF of the DSD, the rising time of the largest  
108 droplets (quantile 99%) differed by up to 17.7% (45 min) for case 1 (Lognormal vs. VDROD-J;  
109 Figure 1b), and by as much as 127.1% (88 h) for case 2 (Figure 1d). While for the smallest  
110 droplets (quantile 1%), rising time would vary by up to 325.2% (167.5 h) for the case 1 (Figure  
111 1b), and by over 5-fold for the case 2 (Figure 1d). The estimated rising velocities of the different  
112 quantiles are provided in the Supplementary Information (Table S1). The surfacing time of the  
113 lognormal and Rosin-Rammler DSDs from case 1 with a d50 of 900  $\mu\text{m}$  (minimum surfacing  
114 time of 4 h, 8.75 h for 50% and 51.55 h for 99% of the oil) matches the estimates without sub-  
115 sea dispersant injection (SSDI) at the Macondo wellhead from Testa et al. (2016) and Gros et al.  
116 (2017) that suggest that all the oil reached the surface within 7 h for the largest droplets. In our  
117 idealized conditions with the same d50, the lower tail of the VDROD-J DSD requires an extra  
118 125.25 h to surfacing as compared to the Rosin-Rammler DSD and 167.5 h compared to the  
119 lognormal one (Figure 1b). For the case 2, with a d50 of 117  $\mu\text{m}$ , the relative anomalies of  
120 surfacing time are higher among the different PDFs due to the wider range of surfacing time  
121 compared to case 1. This long surfacing time results from the small d50 and the omission of  
122 internal degassing process, whereby the multiphase droplet ascent accelerates as its gas-phase  
123 expands with decreasing hydrostatic pressure (Pesch et al. 2018, 2020b). Notably, the lognormal  
124 distribution differs the most from the Rosin-Rammler and VDROD-J PDFs (Figure 1b,d). Our  
125 idealized conditions results are also in line with the predominating importance of the d50 for the  
126 median surfacing time of oil droplets, with longer surfacing time for smaller d50s (Paris et al.  
127 2012; Socolofsky et al. 2019), yet they still show that the choice of distribution function has  
128 notable influence.

129

130 We then applied the same approach of comparing different outputs by only varying the PDF in a  
131 more realistic application to the DWH oil spill. We used a four-dimension Lagrangian  
132 framework, the Connectivity Modeling System (CMS; Paris et al. 2013), in which the DSDs are  
133 deployed in simulations configured with the DWH initial conditions as described in Perlin et al.

134 (2020). In short, the CMS oil application performs Lagrangian particle tracking of oil droplets  
135 released in the far-field at the trap height, approximately 300 m above the wellhead (Socolofsky  
136 et al. 2011). Particle transport calculations take into account 3D ocean currents, wind drift,  
137 temperature, salinity, multi-fraction droplet density, droplet size evolution due to partitioning  
138 and biodegradation, and surface oil evaporation (Paris et al. 2012; Le Henaff et al. 2012; Lindo-  
139 Atichati et al. 2016; Perlin et al. 2020). The 4th order Runge-Kutta integration scheme forms the  
140 basis for particle advection in the model, while a random walk model represents sub-grid scale  
141 turbulent diffusion (Paris et al. 2013). In our simulations, the vertical and horizontal diffusivities  
142 coefficients were based on the empirical approximations from Paris et al. (2012) and set to  $10^{-5}$   
143  $\text{m}^2 \text{s}^{-1}$  and  $1 \text{m}^2 \text{s}^{-1}$ . Computations of the vertical terminal velocity of a droplet are based on its  
144 density and size, its Reynolds number, as well as on other ambient conditions such as water  
145 temperature, salinity, density, and kinematic viscosity (Zheng et al. 2003, Paris et al. 2012).  
146 Fluid flow includes a release of 3000 oil droplets at the trap height every 2 hours for 87 days  
147 until July 15, 2010—the day of successful containment of the spill using a capping stack. The  
148 release location is  $28.736^\circ\text{N}$ ,  $88.365^\circ \text{W}$  at 1222 m, i.e., the estimated height of oil and gas  
149 separation 300 m above the wellhead (Socolofsky et al. 2011). The oil droplets were tracked for  
150 total of 167 days from the initial blowout date, and the surface oil mass (in the layer 0-1 m) and  
151 the sedimented and beached oil mass were computed for each  $0.1^\circ \times 0.1^\circ$  spatial bin over the  
152 entire span of the oil spill. Here, oil concentrations are estimated by post-processing, and the six  
153 scenarios of DSD presented in Figure 1 were carried out using two model simulations: the first  
154 with oil droplets ranging from 1 to 2400  $\mu\text{m}$  (case 1) and the second with oil droplets ranging  
155 from 1 to 500  $\mu\text{m}$  (case 2). We then computed differences in the sedimented and beached oil  
156 mass and of the surface oil mass between the three scenarios of DSD for each simulation.

157

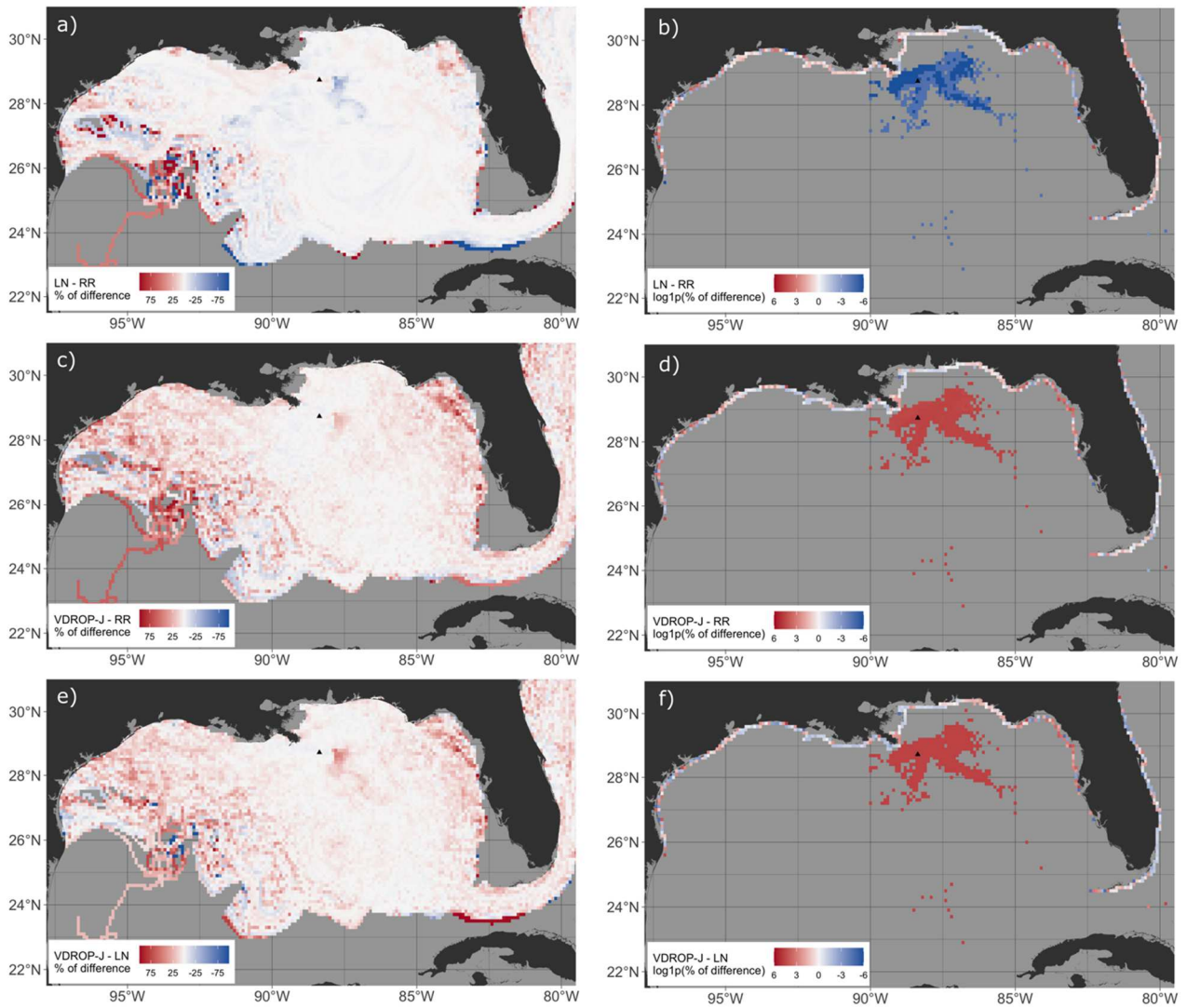
158 As expected, the differences are once again highly sensitive to the  $d_{50}$  and the size range of the  
159 oil droplet modeled. The total combined mass of oil that beached and sedimented was indeed  
160 higher by 45.29% (187,996 tons versus 129,394 tons), 43.83% (185,595 tons versus 129,038  
161 tons) and 50.68% (195,052 tons versus 129,444 tons) with the smaller range of oil droplet sizes  
162 and  $d_{50}$  (case 2) compared to the larger ones (case 1) for the lognormal, Rosin-Rammler and

163 VDROD-J DSDs, respectively. Yet, the choice of the DSD's PDF played an important role  
164 within each case. When comparing the proportion of oil mass that beached over the oil mass that  
165 sedimented, the vast majority of the oil beached in case 1 (i.e. large  $d_{50}$ ) and the choice of PDF  
166 had a relatively limited influence on these outputs: only  $3.2 \cdot 10^{-5}\%$  of the oil sedimented over the  
167 oil that beached with the lognormal distribution, 0.1% with the Rosin-Rammler, and 1.10% with  
168 the VDROD-J distribution used here. Yet the ratios of beached versus sedimented oil mass  
169 estimates for this case are far below the  $21 \pm 10\%$  sedimentation rate quantified for the DWH oil  
170 spill (Joye et al. 2016, Romero et al. 2017). For case 2 of the smaller oil droplets size range, this  
171 ratio was reversed and the differences were closer to empirical values: the oil sedimented 2.08  
172 times more than it beached with a lognormal DSD (126,978 tons sedimented vs. 61,018 tons  
173 beached), 2.11 times more with a Rosin-Rammler (126,094 tons vs. 59,501 tons), and 2.32 times  
174 more with the DSD modeled from VDROD-J (136,318 tons vs. 58,735 tons). The DSD with the  
175 larger  $d_{50}$  therefore predicts that almost no oil sedimented on the seafloor while most oil reached  
176 the surface and then drifted to the coast, with a total mass of beached oil twice as higher  
177 compared to the scenario with the smaller  $d_{50}$ . This difference also reflects high surface  
178 evaporation rates (de Gouw et al. 2011), affecting mostly the largest droplets since they rise  
179 faster. Interestingly, the total oil mass from case 2 that sedimented is on the same order of  
180 magnitude than the oil that beached in case 1, highlighting that the predictions of oil mass  
181 beached versus sedimented is critically important for evaluation response's tradeoff (Murawski  
182 et al. 2019).

183

184 The proportions of beached versus sedimented oil mass on the seafloor are markedly different  
185 between cases and *distribution functions* of the DSDs, but the spatial distribution of these  
186 differences is perhaps more informative. With the larger droplets (case 1), the major differences  
187 in surface oil concentration occur close to the blowout, 50-100 km to the ESE of the well, and at  
188 the edge of the distribution in the western Gulf of Mexico, over the West Florida shelf (Figure 2,  
189 left panels). The lognormal DSD leads to lower surface oil mass near the well and higher oil  
190 mass in the West Bay of Louisiana compared to the Rosin-Rammler and VDROD-J PDFs  
191 (Figure 2 and Fig. S2, left panels). The DSDs based on Rosin-Rammler and VDROD-J provide

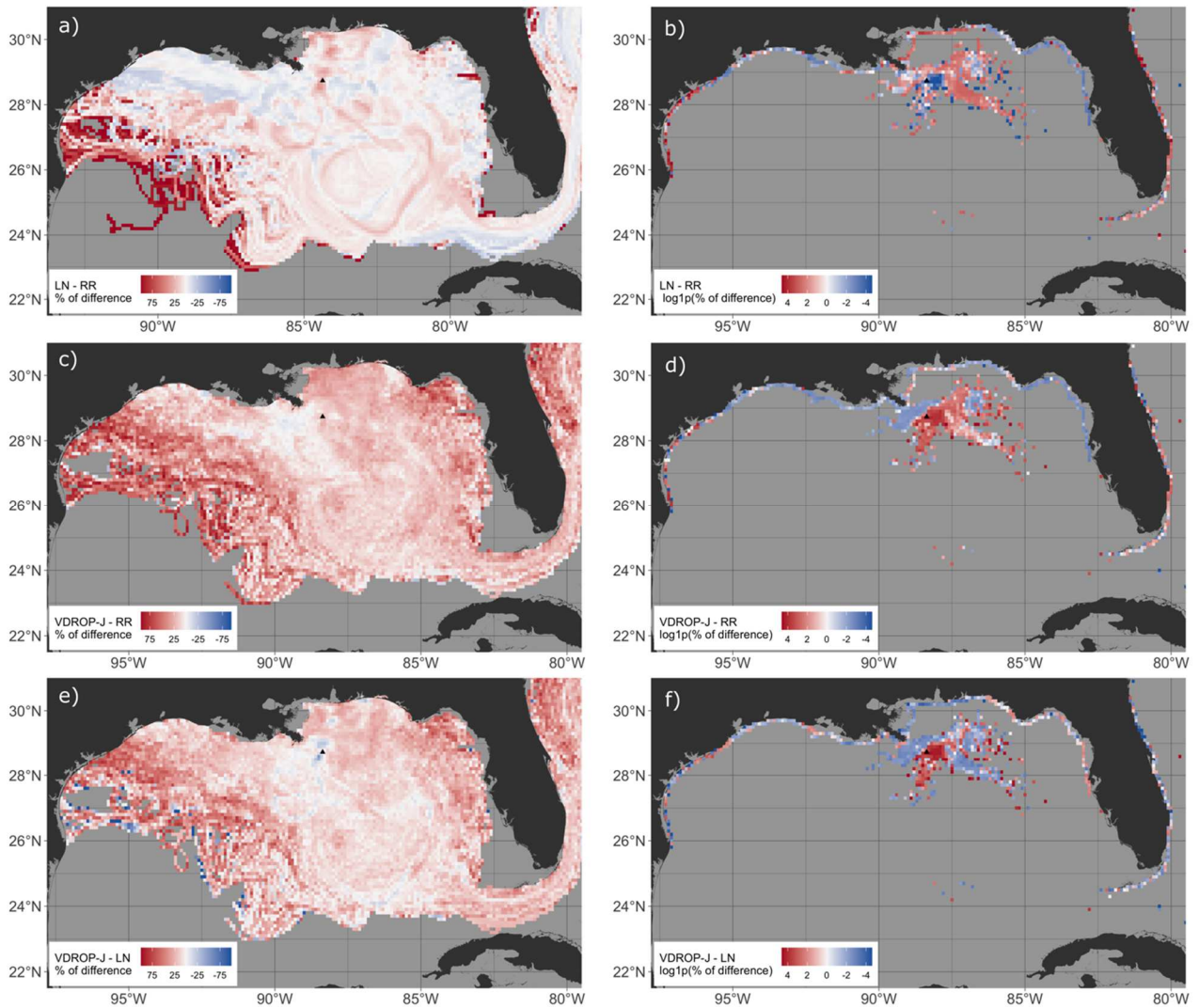
192 very similar results for this size range (i.e. 1-2400  $\mu\text{m}$ ), with VDROF-J showing the highest  
193 surface concentrations overall, in particular over the West Florida Shelf and western Gulf of  
194 Mexico (Figure 2 and Fig. S2). Considering a Rosin-Rammler distribution instead of a  
195 lognormal DSD increases the sedimentation near the blowout location, and also towards the  
196 West Florida slope while it reduces the beached oil along Louisiana, Mississippi and Alabama  
197 coastline (Figure 2b and Fig. S2b). Using DSD modeled from VDROF-J increases the  
198 sedimentation of oil near the well relatively to the lognormal and Rosin-Rammler DSD, while  
199 beaching increases in the western Gulf of Mexico and along the Florida Panhandle, with local  
200 differences of beached oil mass of up to 641 tons per  $0.1^\circ \times 0.1^\circ$  bin (*ca.* 5.9 tons/ $\text{km}^2$ ; Fig. S2).  
201 While the overall patterns were fairly similar between the VDROF-J and Rosin-Rammler  
202 distributions, the beaching was lower along the coast of Louisiana and Mississippi and East  
203 Florida compared to the lognormal DSD in both cases, except east of the Louisiana Peninsula  
204 (Figure 2b,f and Fig. S2b,f).  
205



206  
 207 **Figure 2. (a) Maps of surface (0-1 m) oil mass differences between lognormal (LN) and**  
 208 **Rosin-Rammler (RR), (c) VDROJ and RR, (e) VDROJ and LN, and (b) sedimented oil**  
 209 **mass differences between LN and RR, (d) VDROJ and RR, and (f) VDROJ and LN for**  
 210 **oil droplet sizes in the range 1-2400  $\mu\text{m}$  (case 1) for the three DSD presented on Fig. 1a.**

211  
 212 The differences generated by the choice of DSD functions are more marked when considering  
 213 smaller oil droplets ( $< 500 \mu\text{m}$ ; case 2). While the proportions of differences are lower than for  
 214 case 1 *per se* (generally  $\leq 100\%$ ), the differences in the total mass of oil are considerable and  
 215 surprising for the same  $d_{50}$ : up to over 400 tons of surface oil and  $>4,000$  tons of sedimented oil  
 216 per  $0.1^\circ \times 0.1^\circ$  bins, respectively, which represents a difference of *ca.* 38.5 tons of oil per  $\text{km}^2$   
 217 (Fig. S3). Spatially, the Rosin-Rammler DSD predicts the lowest surface oil amount north of the  
 218 Macondo well (Figure 3a,c). Here again, the largest relative differences occur at the edges of the  
 219 distribution, for each of the three DSDs. One notable difference is also found around the

220 Macondo well area, where the lognormal distribution predicts higher surface oil mass compared  
221 to VDROP-J and Rosin-Rammler DSDs immediately above and up to 60 km towards the North  
222 and South (Figure 3a,e). The masses of beached and sedimented oil resulting from a lognormal  
223 DSD (61,019 tons and 126,989 tons, respectively) are comparable to a Rosin-Rammler  
224 distribution (59,501 tons and 126,094 tons, respectively) but the spatial distribution patterns are  
225 notably different: the lognormal DSD leads to increased beached oil mass in the western Gulf,  
226 along the Louisiana, Mississippi, Alabama and westernmost Florida coasts, and in the domain's  
227 edges (Figure 3b), and higher sedimentation in the Macondo area expect within 50 km to the east  
228 of the well is lower with the latter (Figure 3b). Noteworthy differences also emerged with the  
229 VDROP-J that increased sedimentation south and west of the well compared to both the  
230 lognormal and Rosin-Rammler DSDs (Figure 3d,f and Fig. S3d,f). More oil beached along the  
231 west coasts of the Gulf compared to the Rosin-Rammler, and both the west and east Gulf coasts  
232 compared to the lognormal, while less oil reached the coasts of Louisiana and Mississippi  
233 (Figure 3f and Fig. S3f). Overall, the results show that the area near the Macondo well and the  
234 coastline around the Mississippi delta—the areas most impacted by the oil spill (Nixon et al.  
235 2016)—are the areas where most differences emerge based on the DSD considered.



236  
 237 **Figure 3. (a) Maps of surface (0-1 m) oil mass differences between lognormal (LN) and**  
 238 **Rosin-Rammler (RR), (c) VDROP-J and RR, (e) VDROP-J and LN, and (b) sedimented oil**  
 239 **mass differences between LN and RR, (d) VDROP-J and RR, and (f) VDROP-J and LN for**  
 240 **oil droplet sizes in the range 1-500  $\mu\text{m}$  (case 2) for the three DSD presented on Fig. 1b.**

241  
 242 This study focuses on the critical choice of the DSD's PDF for modeling the transport and fate of  
 243 uncontrolled deep-sea oil spills. To determine optimal oil spill responses and access tradeoffs,  
 244 different scenarios are typically weighted, including considering the application of surface  
 245 and/or subsea chemical dispersants (Xu et al. 2019, Murawski et al. 2019). Here, we demonstrate  
 246 that, even with similar  $d_{50}$ , the surfacing time (Figure 1b,d), the distance travelled by droplets,  
 247 and the patterns of sedimented and beached oil (Figure 2 & 3) will vary based on the two PDFs  
 248 most frequently used for modeling the DSD during deeps oil spills, lognormal and Rosin-  
 249 Rammler (Wang and Calabrese 1986, Spaulding et al. 2015, NASEM 2019). The larger

250 divergences in patterns of surface, sedimented, and beached oil mass and ratios occur between  
251 the theoretical DSDs and the modeled non-fundamental DSD from VDROF-J (Figures 1b,d,2 &  
252 3). Therefore, in the absence of robust observations of the DSD in the water column, sensitivity  
253 and uncertainty analyses should thus consider not only different d50, but also both of the most  
254 characteristic probability distribution functions of the DSD for far-field modeling.

255

256 Differences in oil transport and fate are found between all three distributions (i.e., lognormal,  
257 Rosin-Rammler, and non-fundamental VDROF-J). Our results suggest that for both d50  
258 considered in cases 1 and 2, the lognormal DSD qualitatively matches better the observed  
259 patterns of sedimented oil mass, with higher residual hydrocarbons in the sediment to the west  
260 and north of the Macondo well and on the coasts and wetlands of Louisiana, Mississippi and  
261 Alabama, and less landfall on the West Florida Shelf (Le Hénaff et al. 2012; Boufadel et al.  
262 2014; Nixon et al. 2016; Romero et al. 2017). A better fit with the lognormal distribution has  
263 also been suggested by laboratory (Malone et al., 2018) and field observations (Spaulding et al.,  
264 2015). In conclusion, the choice of DSD function, regardless of the d50, is not trivial as it has  
265 large consequences on the distribution of oil concentration in the water column, sediment and  
266 shoreline. For the DWH, major differences occur at the edge of the oil distribution, which is of  
267 remarkable importance since toxic oil concentrations were present in these regions, yet invisible  
268 to satellite imagery (Berenshtein et al. 2020). Given the rapid advances in oil spill research  
269 following DWH, extending hindcast modeling approaches to consider a range of characteristic  
270 DSDs functions (d50 and PDFs) are needed to ultimately help predict the distribution of oil mass  
271 and evaluating the response tradeoffs in any future deep-sea spill and response measures  
272 (Malone et al. 2020, Murawski et al. 2019).

273

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