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# Heavy and light inertial particle aggregates in homogeneous isotropic turbulence: A study on breakup and stress statistics

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## Abstract

The breakup of inertial, solid aggregates in an incompressible, homogeneous and isotropic three-dimensional turbulent flow is studied by means of a direct numerical simulation, and by a Lagrangian tracking of the aggregates at varying Stokes number and fluid-to-particle density ratio. Within the point-particle approximation of the Maxey-Riley-Gatignol equations of motion, we analyse the statistics of the time series of shear and drag stresses, which are here both deemed as responsible for particle breakup. We observe that, regardless of the Stokes number, the shear stresses produced by the turbulent velocity gradients similarly impact the breakup statistics of inertial and neutrally buoyant aggregates, and dictate the breakup rate of loose aggregates. When the density ratio is different from unity, drag stresses become dominant and are seen to be able to cause to breakup of also the most resistant aggregates. A transition from a shear-dominated to a drag-dominated breakup regime is observed. The present work aims at assessing the role of shear and drag stresses on aggregate breakup and to compute breakup rates to be possibly used in population balance models.

*Keywords:* turbulent breakup, DNS, inertial aggregates, shear stress, drag stress, breakup rate

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## 1. Introduction

Breakup of particles dispersed in a fluid flow is found at the core of many natural and engineering processes. For instance, in aquatic systems, the breakup of plastic waste governs the rate of microparticle production and, as such, plays a key role on the microplastics rate of release in the ocean (Garvey et al., 2020; Poulain et al., 2018; Brouzet et al., 2021). In some pharmaceutical applications, active particles are in need to be reduced in size before administration can take place (Capecelatro et al., 2022; Sabia et al., 2022; Vasquez Giuliano et al., 2022), and in polymer compounding processes, controlled breakup and redistribution

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8 of filler agglomerates is used to produce composites with enhanced mechanical and/or thermal properties  
9 (Frungieri et al., 2020b, 2022).

10 The breakup of dispersed particles is determined by a number of phenomena that challenges simple  
11 modelling approaches (Bäbler et al., 2008). The origin of this complexity relies on few main features: the  
12 first is the multi-scale nature of the problem, with relevant spatial scales ranging from the micron size of the  
13 particles to possibly hundreds of meters (the integral scale of the flow); the second is associated to the way  
14 aggregates spatially sample the flow, and the third depends on the complex interplay between fluid-induced  
15 stresses and inter-particle cohesive forces, which eventually governs the aggregate breakup dynamics.

16 Depending on the application of interest and degree of insight needed, different methods can be deemed as  
17 suitable to study breakup, and they can be mainly differentiated on the basis of the treatment of the dispersed  
18 and dispersing phase and in the mechanism taken into account to predict breakup (e.g. viscous shear, turbulent  
19 fluctuations, wall impact, drag or rotary stress (Breuer & Khalifa, 2019)). On the scale of the aggregate,  
20 detailed predictions can be obtained by first principle structural mechanics (Zaccone et al., 2009; Conchúir &  
21 Zaccone, 2013; Jiang et al., 2020) or by Stokesian dynamics (Brady & Bossis, 1988), with the latter that, when  
22 coupled with models for the inter-particle interactions, is able to fully characterize the breakup occurrence  
23 in terms of critical stress and fragment size distribution. By such an approach, for instance, Harada et al.  
24 (2006) studied the effect of the internal connectivity on the aggregate breakup. Similarly, Harshe & Lattuada  
25 (2012) computed breakup rates and fragment size distribution in linear flows, and Frungieri & Vanni (2021)  
26 studied the particle size distribution and the morphological evolution of a population of colloidal particles  
27 at varying shear stress intensity and physico-chemical properties (Frungieri & Vanni, 2017; Frungieri et al.,  
28 2020a; Vasquez Giuliano et al., 2023).

29 On the macro-scale (i.e., the scale of the equipment), where flow field heterogeneities and boundary layer  
30 phenomena affect the evolution of the dispersed phase, so called Eulerian-Eulerian approaches, especially  
31 when coupled with population balance models (Marchisio et al., 2006), are of particular interest, as they can  
32 be conveniently used to promptly compute the breakup dynamics and the particle size distribution. By such  
33 an approach a number of systems of practical relevance have been investigated, such as traditional (Lebaz  
34 et al., 2021) and Pickering emulsions (Frungieri & Briesen, 2023), bubbly flows (Syed et al., 2018; Zhang  
35 et al., 2021; Maluta et al., 2021; Lehnigk et al., 2022) and particle synthesis processes (Shiea et al., 2022;  
36 Schikarski et al., 2022). However, despite the wide range of applications, such approaches still rely on  
37 empirical correlations, generally assuming a single phenomenon (e.g. viscous shear, turbulent fluctuations,  
38 surface instability) to be responsible for breakup, with the overall robustness of the approach still often in  
39 need to be checked against dedicated experimental campaigns.

40 More recently, approaches aimed at linking small scale and large scale phenomena have emerged, most  
41 of which adopt an Eulerian-Lagrangian simulation strategy. By such an approach, it is possible to study

42 complex flow fields and treat in a more detailed way the dynamics of the dispersed phase, especially when the  
43 back-reaction of the particles on the flow is relevant. In this framework, Chen & Li (2020), considering an  
44 homogeneous isotropic turbulent flow, computed breakup rates in the early stage of an agglomeration process  
45 between adhesive particles, and single events such as restructuring and breakup by turbulent stresses were  
46 also studied (Ruan et al., 2020; Yao & Capecelatro, 2021). However, due to the high computational burden,  
47 this approach is limited to short simulated physical time, low level of turbulence (small turbulence Reynolds  
48 number  $Re_\lambda$ ) and aggregates made by a small number of primary particles compared to what is typically  
49 observed in experiments (Saha et al., 2016).

50 Turbulence affects particle motion in a distinctive manner, in particular in the case of inertial particles  
51 (Brandt & Coletti, 2022). Inertia arises when particles have a finite size, and/or a density mismatch with the  
52 suspending medium. Because of inertia, particles show complex behaviours that notably affect their spatial  
53 distribution (Wang & Maxey, 1993; Bec et al., 2007) and their relative velocity and acceleration statistics  
54 (Falkovich & Pumir, 2007; Bec et al., 2011; Scatamacchia et al., 2012). In particular, strong inhomogeneities  
55 in the particles spatial distribution emerge, an effect that is maximal when the Stokes number is of order unity,  
56 and becomes negligible in the limits of both small and large inertia. Moreover, in the case of large inertia,  
57 heavy particles move almost independently of the fluid, hence they may collide with a large relative velocity.  
58 These events – dubbed *caustics* – can cause a substantial increase in the collision rate (Pumir & Wilkinson,  
59 2016).

60 An attempt to understand how the properties of turbulence affect breakup was undertaken by Guseva &  
61 Feudel (2017) by evolving a population of particles of variable size in a synthetic turbulent flow. However,  
62 such a simulation method does not account for turbulence intermittency which is responsible for the generation  
63 of intense hydrodynamic stresses able to break also the strongest aggregates (Bäbler et al., 2015). In this  
64 context, Bäbler et al. (2012) computed the breakup rate of tracer-like aggregates at varying internal strength in  
65 a homogeneous isotropic turbulent flow, assuming particles to break under the action of the turbulent viscous  
66 dissipation only. Similarly, De Bona et al. (2014), by combining a DNS of the turbulent flow with a Discrete  
67 Element Method based on Stokesian dynamics, estimated the rate of breakup of aggregates addressing at the  
68 same time size and distribution of the formed fragments.

69 In this work, we study the fragmentation of inertial heavy and light aggregates in a turbulent flow  
70 by combining a direct numerical simulation of the turbulence with a Lagrangian tracking of the particles,  
71 performed within the point-particle approximation of the Maxey-Riley-Gatignol equations of motion. Likewise  
72 to our previous work (Bäbler et al., 2012), we neglect the internal structure of the aggregates, the hydrodynamic  
73 interactions between them (Zahnnow et al., 2011) and the accumulation of stresses on their structure (Marchioli  
74 & Soldati, 2015), and we assume breakup to occur in brittle manner whenever the local instantaneous  
75 hydrodynamical stresses acting on the aggregate exceeds a critical value (Bäbler et al., 2012; Breuer &

76 Khalifa, 2019). Differently from our previous work (Bäbler et al., 2015), in which aggregates were tracked  
77 as they were tracers, in this work we consider the particle inertia and we track their motion using a minimal  
78 formulation of the Maxey-Riley-Gatignol equation of particle motion, keeping into account inertia, drag stress  
79 and added mass effects. We focus in particular on the role of the Stokes number and of the particle buoyancy  
80 on both shear and drag stress statistics, with the aim of assessing separately their role on the rate of aggregate  
81 breakup. We study the breakup occurrence at varying aggregate strength.

82 The paper is organised as follows: in Section 2, we introduce the equation of motion for the inertial  
83 aggregates and the turbulent flow and we provide details about their numerical integration; in Section 2.2,  
84 we present different approaches to measure the breakup rate. Results are discussed in Section 3, which are  
85 followed by the concluding remarks in Section 4.

## 86 2. Methodology

87 The aggregates are assumed to be spherical assemblies of small unbreakable primary particles, bond to  
88 each other by adhesive colloidal forces. Aggregates, depending on the growth mechanism (due to velocity  
89 gradients, Brownian motion, external magnetic or electric field), restructuring effects and synthesis conditions,  
90 can be expected to have variable fractal features, and may or not display a spherical symmetry. However,  
91 in this work we restrict ourselves to the analysis of spherical bodies. This is also motivated by the fact  
92 that equivalent radii such as the hydrodynamic radius (that can be inferred from light scattering techniques),  
93 the Stokes radius (from settling analysis), and the gyration radius (determined by equaling the aggregate  
94 moment of inertia with those of a sphere) are often used for characterizing aggregates. We consider a dilute  
95 suspension of aggregates, which have no feedback on the flow in which they are suspended, and which have  
96 no hydrodynamic interactions between them. When addressing aggregate breakup in dilute suspensions, it is  
97 common to neglect the hydrodynamic interactions, i.e., the mutual flow disturbance due to the presence of  
98 the other aggregates, and the collision phenomena. Both, flow disturbances and collisions can in principle  
99 contribute to the overall breakup kinetics. However, even if few studies have addressed these phenomena  
100 (Dizaji et al., 2019; Frungieri & Vanni, 2021; Chen & Li, 2020; Perrone et al., 2023), the extent to which they  
101 affect the fragmentation dynamics in an ensemble of aggregates is yet to be assessed.

102 Aggregates have sizes smaller or comparable to the Kolmogorov scale of the flow  $\eta = (\nu^3/\langle\varepsilon\rangle)^{1/4}$ , where  
103  $\nu$  is the kinematic viscosity of the fluid and  $\langle\varepsilon\rangle$  is the mean rate of energy dissipation. Both the particle  
104 Reynolds number, defined as  $Re_p = 2R v_p/\nu$ , where  $v_p$  is a typical particle velocity and  $R$  the particle radius,  
105 and the particle Reynolds number based on the relative particle-fluid velocity,  $2R|v_p - u_f|/\nu$ , are small.

106 Aggregate trajectories are obtained by evolving a minimal formulation of the original Maxey-Riley-  
107 Gatignol equations of motion (Maxey & Riley, 1983; Gatignol, 1983) in which the pressure force, the added

108 mass and the Stokes drag are kept into account, and which has been frequently used to describe the motion  
 109 of small, rigid, spherical particles in unsteady flows (Bec et al., 2010). Such equations read as:

$$\frac{d\mathbf{X}}{dt} = \mathbf{V}(\mathbf{X}, t), \quad (1)$$

$$\frac{d\mathbf{V}}{dt} = \beta \frac{D\mathbf{u}(\mathbf{X}, t)}{Dt} + \frac{\mathbf{u}(\mathbf{X}, t) - \mathbf{V}(\mathbf{X}, t)}{\tau_p}, \quad (2)$$

110 where  $\mathbf{V}$  is the particle velocity,  $\mathbf{X}$  is the particle position and  $\mathbf{u}$  is the undisturbed fluid velocity at the  
 111 particle position. It is apparent that only two dimensionless parameters govern the aggregate motion: the  
 112 density ratio  $\beta$  defined as  $\beta = \frac{3\rho_f}{\rho_f + 2\rho_p}$  where  $\rho_p$  and  $\rho_f$  represent the particle and fluid density, respectively,  
 113 and the Stokes number  $St = \tau_p/\tau_\eta$ , where the aggregate relaxation time is defined as  $\tau_p = R^2/(3\beta\nu)$ , being  
 114  $R$  the aggregate radius. The Kolmogorov time scale of the flow, entering the definition of the Stokes number,  
 115 is  $\tau_\eta = (\nu/\langle\varepsilon\rangle)^{1/2}$ . From the definition of  $\beta$ , it is clear that neutrally buoyant particles have  $\beta = 1$ , extremely  
 116 light particles have  $\beta \rightarrow 3$ , whereas heavy particles have  $\beta \rightarrow 0$ . We track the motion of particles at varying  
 117 inertia, by changing both their buoyancy parameter and their Stokes number as schematically illustrated in  
 118 Fig. 1. We focus our analysis on the role of drag and shear stress with the aim of assessing separately their role  
 119 on the rate of aggregate breakup in turbulence. Further effects can be in principle taken into account, such as  
 120 Faxén and Basset forces, the correction of the drag coefficient at varying particle size and slip velocity, or a  
 121 2-way coupling can be used. However, for the purpose of the present paper, we deem as suitable the minimal  
 122 formulation of the equation of particle motion reported in Eq. (2) which allows us to keep the parameters space  
 123 small, being formed by  $\beta$  and  $St$  only, and to limit the computational burden of the simulations, which can  
 124 become large in particular when history effects are taken into account (Olivieri et al., 2014). Considerations  
 125 about the full point-particle formulation of the equation of motion can be found in the work of Maxey (1987),  
 126 Horwitz & Mani (2018) and Volk et al. (2008).

127 The fluid velocity  $\mathbf{u}$  is evolved according to the incompressible Navier-Stokes (NS) equations reading as:

$$\frac{\partial\mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla\mathbf{u} = -\frac{\nabla p}{\rho_f} + \nu\nabla^2\mathbf{u} + \mathbf{F}, \quad \nabla \cdot \mathbf{u} = 0. \quad (3)$$

128 A steady, statistically homogeneous and isotropic turbulent flow is obtained by adding to the NS equations  
 129 a forcing term  $\mathbf{F}$  injecting energy in the first low-wave number shells and keeping constant their spectral  
 130 content (Bec et al., 2010). The kinematic viscosity is chosen in such a way that the Kolmogorov length  
 131 scale equals the grid spacing  $\eta \simeq \delta x$ . By doing so, a good resolution of the small-scale velocity fluctuations  
 132 is obtained. At the steady state, the energy input balances the mean kinetic energy dissipation such that  
 133  $\langle\mathbf{F} \cdot \mathbf{u}\rangle \simeq \langle\varepsilon\rangle$ . The Navier Stokes equations are solved on a  $512^3$  cubic grid with periodic boundary conditions  
 134 and a Taylor-Reynolds number  $Re_\lambda \simeq 185$ . The equations of fluid motion are integrated until a statistically  
 135 steady state is reached. Then, particles are released with homogeneously distributed initial positions and

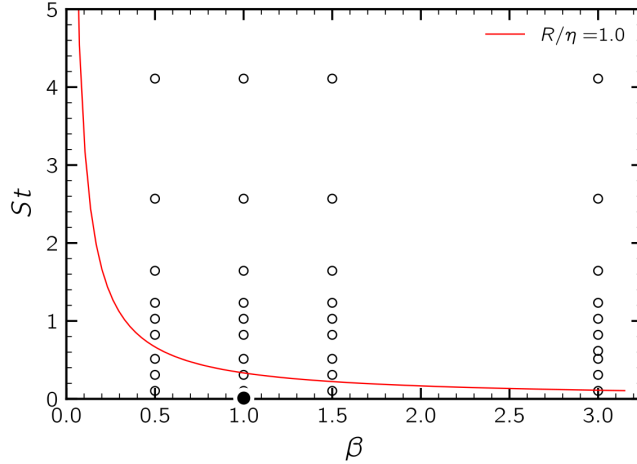


Figure 1: Parameters space of the particles used in our numerical experiments. The red line represents the locus of constant  $R/\eta$ . We analyzed 128k trajectories per each single  $(\beta, St)$  choice. Tracers are represented by the black dot. Trajectories were evolved using a second-order Adam-Bashforth time-stepping.

Table 1: Parameters of the DNS simulation. Microscale Reynolds number  $Re_\lambda$ , root-mean-square velocity  $u_{rms}$ , mean energy dissipation  $\varepsilon$ , kinematic viscosity  $\nu$ , Kolmogorov scale  $\eta = (\nu^3/\langle\varepsilon\rangle)^{1/4}$ , integral scale  $L$ , Eulerian large-eddy turnover time  $T_E = L/u_{rms}$ , Kolmogorov timescale  $\tau_\eta = (\nu/\langle\varepsilon\rangle)^{1/2}$ , grid spacing  $\Delta x$ , number of grid points  $N$ , simulation time  $t_s$ .

$Re_\lambda$	$u_{rms}$	$\varepsilon$	$\nu$	$\eta$	$L$	$T_E$	$\tau_\eta$	$\Delta x$	$N^3$	$t_s$
185	1.4	0.94	0.00205	0.010	$\pi$	2.2	0.047	0.012	$512^3$	13.2

136 velocities equal to the local fluid velocity. The equation of fluid and particle motion are then advanced in  
 137 parallel with a time step equal to  $1/100\tau_\eta$ . The choice of a small time-resolution allows us to finely resolve  
 138 the particle motion. After releasing particles, a transient in the particle dynamics follows, lasting for about  
 139 2-3 large-scale eddy turnover times before Lagrangian stationary statistics are reached. Only after this stage,  
 140 the particle trajectories are stored. For each particle class, we track  $128 \cdot 10^3$  trajectories and dump the data  
 141 every  $1/10\tau_\eta$ .

142 In Table 1 the main characteristics of the flow and the numerical details of the simulations are reported.  
 143 Further numerical details on both the Eulerian and Lagrangian approaches can be found in the work by Bec  
 144 et al. (2010).

### 145 2.1. Hydrodynamic stresses

146 We assume aggregates to be brittle objects that instantaneously respond to the external stress and that  
 147 undergo breakup as soon as their critical resistance is exceeded by the total fluid dynamic stress acting on

148 them.

149 Following Kusters (1991) and Breuer & Khalifa (2019), two hydrodynamic stresses are deemed responsible  
 150 for the breakup of the aggregate, namely the shear stress  $\sigma_\varepsilon$ , due to the fluid velocity gradients at the particle  
 151 position, and the drag stress  $\sigma_{St}$ , due to the slip velocity between the aggregate and the underlying flow. The  
 152 shear stress along the aggregate trajectory  $\mathbf{X}$  is calculated using (Kusters, 1991):

$$\sigma_\varepsilon(\mathbf{X}, t) = \mu \sqrt{\frac{2}{15} \frac{\varepsilon(\mathbf{X})}{\nu}} \quad (4)$$

153 where  $\mu = \nu \rho_f$  is the dynamic viscosity of the flow and  $\varepsilon = 2\nu e_{ij}e_{ij}$  is the local energy dissipation  
 154 rate, with  $e_{ij} = 1/2(\nabla_j u_i + \nabla_i u_j)$  denoting the symmetric part of the velocity gradient tensor  $\nabla_j u_i$ . The  
 155 prefactor in Eq. (4), i.e.  $\sqrt{2/15}$ , is derived assuming isotropy and homogeneity of the turbulent flow (Kusters,  
 156 1991). These assumptions become invalid in strongly non-homogenous flows, for instance, in a wall bounded  
 157 boundary layer flow where the local velocity gradient is dominated by the (anisotropic) mean gradient. In  
 158 such a case the prefactor in Eq. (4) is expected to become larger. However, the square-root dependency on  
 159 the local energy dissipation rate remains valid also in such cases.

Likewise, the drag stress along the aggregate trajectory is calculated as:

$$\sigma_{St}(\mathbf{X}, t) = \mu \frac{3|\mathbf{u}(\mathbf{X}) - \mathbf{V}(\mathbf{X})|}{2R} \quad (5)$$

160 where we have assumed, as above, that the stress is isotropic and that the aggregate has a spherical shape (see  
 161 also Breuer & Khalifa (2019) for a discussion). Therefore the drag stress acting on an aggregate can be simply  
 162 computed as the ratio between the drag force  $6\pi\mu R|\mathbf{u} - \mathbf{V}|$  and the surface area  $4\pi R^2$ . Following Kusters  
 163 (1991), the two stresses are added up linearly  $\sigma_{tot} = \sigma_{St} + \sigma_\varepsilon$ , thus assuming that the two stresses propagate  
 164 instantaneously across the aggregate bond network and that their load on the structure can be superimposed.

## 165 2.2. Breakup rate measurements

166 Our work is aimed at evaluating particle breakup rates at varying inertia. In our model, an individual  
 167 aggregate undergoes breakup instantaneously when the total hydrodynamic stress acting on it exceeds a  
 168 critical threshold value  $\sigma_{cr}$ , representing the aggregate mechanical strength.

169 Figure 2 illustrates the simple modeling framework we use. The aggregate is released at a random time  
 170  $t_0$  and it samples the flow until it experiences for the first time a hydrodynamic stress exceeding its resistance  
 171  $\sigma_{cr}$  (this, in the schematics of Fig. 2, occurs at  $t_1$ ). The time-lag between release at  $t_0$  and breakup at  $t_1$   
 172 identifies the exit time  $\tau_{\sigma_{cr}}$ . The breakup rate is therefore calculated as the inverse of the mean exit time, i.e.,  
 173 as:

$$f_{\sigma_{cr}} \equiv \frac{1}{\langle \tau_{\sigma_{cr}} \rangle} \quad (6)$$

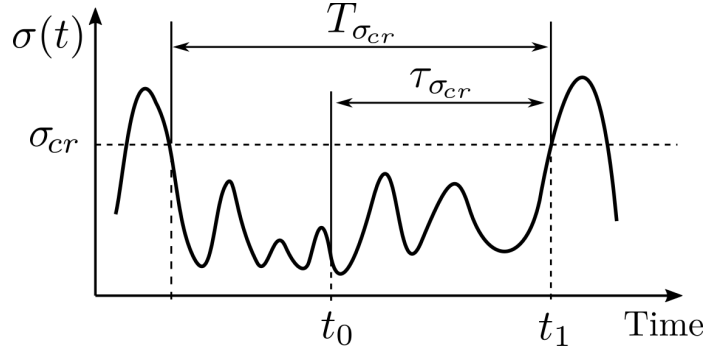


Figure 2: Schematic of the total hydrodynamic stress along a particle trajectory. The particle is released at time  $t_0$  and breaks up at time  $t_1$ . The time-lags  $\tau_{\sigma_{cr}}$  and  $T_{\sigma_{cr}}$  are the exit time and the diving time, respectively.

174 where  $\langle \cdot \rangle$  denotes the ensemble average over the Lagrangian trajectories. Notice that within this modelling  
 175 framework, situations where the hydrodynamic stress exceeds the critical stress already at the point of release  
 176 are ignored when computing the average in Eq. (6). The reason for this is that breakup events that occur right  
 177 at the point of release would be governed by the frequency of aggregate release and not by the timescale of  
 178 the turbulent fluctuations.

179 The picture presented in Fig. 2 can be used to identify a second time scale. For this, we notice that the  
 180 hydrodynamic stress experienced by the aggregate during its lifetime is part of the Lagrangian time series of  
 181 a particle with the same kinematic characteristics (Stokes number and density parameter) as the aggregate.  
 182 Since this Lagrangian time series exists throughout the lifetime of the flow, the hydrodynamic stress can  
 183 also be traced backwards in time, as depicted in Fig. 2. Hence, by considering the Lagrangian time series  
 184 of which the aggregate released at  $t_0$  sampled the segment between  $t_0$  and  $t_1$ , we can identify a new time  
 185 scale, called diving time  $T_{\sigma_{cr}}$ , as the time-lag between the last down-crossing of  $\sigma_{cr}$  before  $t_0$  and the first  
 186 up-crossing of  $\sigma_{cr}$  after  $t_0$ . This time scale  $T_{\sigma_{cr}}$  allows us for deriving a few additional relationships to  
 187 express or approximate the breakup rate. At first, it can be shown that for a statistically stationary flow the  
 188 breakup rate defined in Eq. (6) can also be expressed through the variance of the diving time as (Bäbler et al.,  
 189 2012):

$$f_{\sigma_{cr}} = \frac{2\langle T_{\sigma_{cr}} \rangle}{\langle T_{\sigma_{cr}}^2 \rangle} \quad (7)$$

Moreover, we can define a proxy-breakup rate by using the diving time instead of the exit time. The  
 proxy-breakup rate simply reads as:

$$\tilde{f}_{\sigma_{cr}} = \frac{1}{\langle T_{\sigma_{cr}} \rangle} \quad (8)$$

190 For a stationary stochastic process, the above can be calculated by the frequency of upward-crossing of

191 the threshold  $\sigma_{cr}$  divided by the fraction of time the trajectory stays below the threshold. The former can  
 192 be expressed by means of the Rice theorem, whereas the latter follows from the probability density function  
 193 (PDF) of  $\sigma_{tot}(t)$  (Lindgren, 2019). By doing so, the proxy-breakup rate can be computed as:

$$\tilde{f}_{\sigma_{cr}} = \frac{\int_0^{\infty} \dot{\sigma}_{tot} p_2(\sigma_{cr}, \dot{\sigma}_{tot}) d\dot{\sigma}_{tot}}{\int_0^{\sigma_{cr}} p(\sigma_{tot}) d\sigma_{tot}} \quad (9)$$

194 where  $p_2(\sigma_{tot}, \dot{\sigma}_{tot})$  is the joint PDF of the total stress and of its time derivative, and where  $p(\sigma_{tot})$  is the  
 195 PDF of  $\sigma_{tot}$ . Approximating the breakup rate using the Rice formula was first proposed by Loginov (1986).  
 196 Accordingly, Eq. (9) is referred to as Loginov's approximation.

197 To independently estimate the breakup rate for large threshold values (and thus to validate the Loginov's  
 198 formula) we make use of another routine. This consist of releasing a large number of aggregates at the  
 199 beginning of the simulation and monitoring the decay of their number concentration as breakup events occur.  
 200 From the temporal evolution of the number of aggregates  $N(t)$  we can estimate the breakup rate assuming a  
 201 first-order process as:

$$\frac{dN}{dt} = -\hat{f}_{\sigma_{cr}} N \quad (10)$$

202 On the basis of Eq. (10), the proxy breakup rate can be determined from the slope of a plot of  $\ln(N/N_0)$   
 203 versus  $t$ , with  $N_0$  being the number of released aggregates.

204 For large values of the threshold  $\sigma_{cr}$ , the proxy-breakup rate based on the diving time (Eq. (8)) approaches  
 205 the exact breakup rate based on the exit time (Eq. (6)):

$$\langle \tau_{\sigma_{cr}} \rangle \sim \langle T_{\sigma_{cr}} \rangle \quad \text{for } \sigma_{cr} \gg \langle \sigma_{tot} \rangle \quad (11)$$

206 To understand this, let us consider a long timeseries of length  $t_L$ , with  $t_L$  being much larger than the large  
 207 eddy turnover time  $T_E$ . Along this time series, let us assume that the threshold  $\sigma_{cr}$  is crossed  $N$ -times in  
 208 the upward direction. If the threshold is very large, the total time the timeseries spends above  $\sigma_{cr}$  is small  
 209 and the mean diving time follows simply as  $\langle T \rangle = t_L/N$ . If the threshold is large, the timespans between two  
 210 consecutive up-crossings is also large compared to the large eddy turnover time  $T_E$ , and diving events can be  
 211 assumed to be distributed randomly along the trajectory. For the exit times, next to the  $N$  upward-crossing  
 212 events along the trajectory, there is also the release at a random time  $t_0$ , such that in total  $N + 1$  events occur  
 213 along the trajectory. Thus, the mean exit time follows as  $\langle \tau \rangle = t_L/(N + 1)$ . For a stationary flow, the length  
 214  $t_L$  and the number of upward-crossings  $N$  are large such that  $t_L/N \approx t_L/(N + 1)$ , thus finally rationalizing  
 215 the result of Eq. (11).

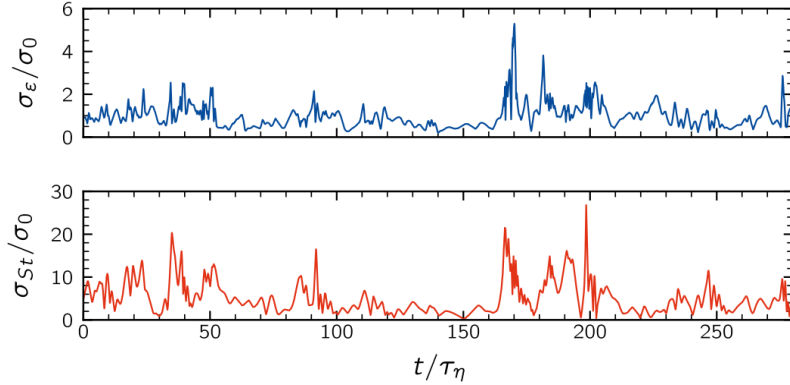


Figure 3: Time series of the shear stress  $\sigma_\varepsilon$  (top panel) and drag stress  $\sigma_{St}$  (bottom panel) for a sample aggregate with Stokes number  $St = 1.64$  and buoyancy parameter  $\beta = 3$ . The  $x$ -axis is normalized by the Kolmogorov time scale of the flow. The  $y$ -axis is normalized by the average shear stress  $\sigma_0$  experienced by tracer particles ( $\beta = 1$ ,  $St = 0$ ).

### 216 3. Results and discussion

#### 217 3.1. Hydrodynamic stress statistics

218 To study the statistics of the hydrodynamic stresses experienced by inertial aggregates, we consider them  
 219 as infinitely strong and let them move in the flow field without undergoing breakup. Figure 3 shows the time  
 220 series of the shear stress  $\sigma_\varepsilon$  and of the drag stress  $\sigma_{St}$ , recorded along the trajectory of a heavy aggregate with  
 221 inertia values  $\beta = 0.5$  and  $St = 1.64$ . We observe that both stresses exhibit strong fluctuations, each with its  
 222 own dynamics. Indeed, the first is determined by the way the particle sample the different regions of the flow,  
 223 whereas the second comes from the fluid-particle slip velocity. For the chosen set of inertia parameters, we  
 224 also observe that the drag stress dominates over the shear stress along the whole particle trajectory.

225 Figure 4 reports the probability density function of the shear stress for two buoyancy parameters ( $\beta = 0.5$   
 226 and  $\beta = 3.0$ , corresponding to heavy and light particles, respectively) and for the smallest and largest Stokes  
 227 values investigated in this work. The figure also reports the distribution of the shear stress experienced  
 228 by tracer particles, for which  $\beta = 1$  and  $St \rightarrow 0$ . It is apparent that for all combinations of  $\beta$  and  $St$ ,  
 229 the distributions are similar and tend to overlap. This overlap implies that, regardless of their inertia, the  
 230 aggregates sample the shear field in almost the same manner as tracer particles. The small differences in the  
 231 shear stress distributions, visible in particular on the right tail, reflect the uneven sampling of the shear field  
 232 due to the segregation effects of inertial particles, which will be shortly addressed in the following.

233 Figure 5 shows the PDFs of the drag stress and of the total stress ( $\sigma_{tot} = \sigma_\varepsilon + \sigma_{St}$ ) for different Stokes  
 234 values for heavy (top row) and light particles (bottom row). The left panels (panel (a) and (b)) show the  
 235 PDF of the drag stress with the horizontal axis made dimensionless using the average shear stress for tracers  
 236 (denoted by  $\sigma_0$ ), which, as seen, is close to the average value of all the other particle class. From this

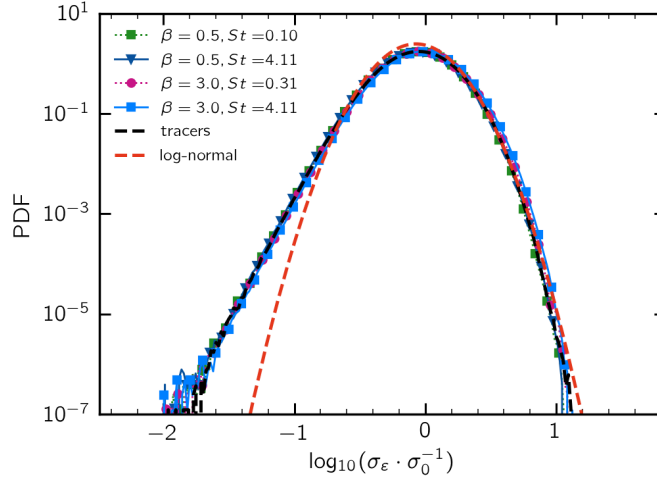


Figure 4: Probability density function of the shear stress  $\sigma_\varepsilon$  at varying particle inertia. The red dotted line represents a log-normal distribution  $P(\sigma_0, 0.50\sigma_0)$ , with  $\sigma_0$  being the average  $\sigma_\varepsilon$  experienced by tracer particles.

237 representation, it is apparent that the distributions move towards larger stress values, implying that the mean  
 238 drag stress increases as the Stokes number grows. However, for very large Stokes numbers, extreme drag  
 239 events become less frequent. This is particularly apparent in Fig. 5b where for the largest Stokes number  
 240 (black triangles) the right tail of the PDF exhibits a faster drop-off compared to the smaller Stokes number  
 241 distributions. To further explore this observation, in the middle panels of Fig. 5 (panel (c) and (d)) we plot  
 242 the PDF of the drag stress normalized by its mean value. This representation leads to a collapse of the  
 243 left tails of the PDFs and makes it more apparent the faster drop-off of the right tail that occurs at large  
 244 Stokes number. From this, it can be inferred that at increasing Stokes number particles filter out extreme drag  
 245 stress events. Furthermore, we also note that the left tail of the drag stress distribution is well described by  
 246 a  $\chi^2$  distribution with 3 degrees of freedom. We consider this an outcome of the Gaussian dynamics of the  
 247 smallest stress contributions, that occur in the smooth regions of the flow and that are proportional to the first  
 248 moment of the distribution of the drag stress.

249 The right panels of Fig. 5 (panel (e) and (f)) show the PDF of the total stress together with the shear stress  
 250 for tracers (dashed black line), which, as seen in Fig. 4, is a good approximation for the shear stress of all  
 251 particle families. For the  $\beta \neq 1$  particles, it is observed that the total stress, that is the summation of the shear  
 252 and drag stress, is substantially larger than the shear stress alone, an effect that becomes more apparent as  
 253 the particle inertia increases. The filtering behavior of highly inertial light and heavy particles, which makes  
 254 extreme drag events less likely, is also here observed.

255 The mean drag stress is further explored in Fig. 6. Panel (a) shows the mean drag stress for different  $\beta$   
 256 parameters plotted versus the Stokes number (the  $\sigma_0$  used to normalize the vertical axis in Fig. 6a is the same

257 of Fig. 5). The plot also shows the mean shear stress of tracers (dashed line) that is approximately equal to  
 258 the mean shear stress of all the other particle families. It is apparent that, as soon as deviations from neutral  
 259 buoyancy occur ( $\beta \neq 1$ ), particles experience significant drag stresses, which become largely dominating  
 260 over the shear stress at increasing Stokes. To rationalize this observation, we recall that the drag stress is  
 261 proportional to the slip velocity  $|\mathbf{u} - \mathbf{V}|$  (Eq. (5)) and we analyze the mean slip velocity as a function of both  
 262 the Stokes number and the buoyancy parameter in Fig. 6b. At small Stokes number, we observe that the mean  
 263 slip velocity scales linearly for all the investigated buoyancy parameters, except for neutrally buoyant particles.  
 264 This linear scaling is in agreement with the Maxey approximation for weak inertial particles (Maxey, 1987).  
 265 Indeed, starting from Eq. (2), in the limit of  $St \rightarrow 0$ , the slip velocity becomes (Daitche, 2015):

$$\mathbf{u} - \mathbf{V} \equiv \tau_p (d\mathbf{V}/dt - \beta D\mathbf{u}/Dt) \simeq \tau_p (1 - \beta) a_f, \quad (12)$$

266 where we have assumed that in the limit of small Stokes numbers the aggregate acceleration and the fluid  
 267 acceleration along the aggregate trajectory are equal, i.e.,  $d\mathbf{V}(\mathbf{X})/dt \simeq D\mathbf{u}(\mathbf{X})/Dt \simeq a_f(\mathbf{X})$ . Hence, in the  
 268 weak inertia limit,  $\langle |\mathbf{u} - \mathbf{V}| \rangle \propto St$ , as shown by the black dotted line (Boffetta et al., 2007). As the Stokes  
 269 number further increases, the increase in  $\langle |\mathbf{u} - \mathbf{V}| \rangle$  flattens, and it is expected to reach a plateau for very large  
 270 Stokes number, at which  $\mathbf{u}$  and  $\mathbf{V}$  become decorrelated. To relate these findings to the behavior of  $\langle \sigma_{St} \rangle$  seen  
 271 in Fig. 6a, we recall that the aggregate radius appearing in the definition of Eq. (5) goes as  $R \sim St^{1/2}$ . Hence,  
 272 the behavior of  $\langle \sigma_{St} \rangle$ , for the  $\beta \neq 1$  particles, is governed by the slip velocity, that, as seen, grows linearly at  
 273 small  $St$  numbers and flattens down as the particle radius further increases, that (for a fixed  $\beta$ ) happens with  
 274  $St^{1/2}$ .

275 The case of neutrally buoyant particles ( $\beta = 1$ ) requires a separate treatment. For these particles, the  
 276 drag stress is relatively small and the stress experienced is mostly due to shear, except for the large Stokes  
 277 number of our dataset, i.e.,  $St > 4$  (see Fig. 6a). This is also shown in the inset in Fig. 6b where  $\langle |\mathbf{u} - \mathbf{V}| \rangle$   
 278 is plotted versus  $\beta$  for Stokes fixed at  $St \approx 1$ . A clear minimum can be observed at  $\beta = 1$ , whereas large  
 279 slip velocities are seen as soon as deviations from neutral buoyancy occur. A small slip velocity for neutrally  
 280 buoyant particles compared to buoyant particles was also observed by Daitche (2015).

To close the discussion about the stress statistics experienced by aggregates in HIT, let us shortly address  
 segregation effects, i.e. the preferential clustering of particles in certain regions of the flow. Preferential  
 concentration of inertial particles is a well studied phenomena, generally quantified analyzing the pair  
 probability distribution or a fractal correlation dimension. (Calzavarini et al., 2008a,b; Bec et al., 2005;  
 Daitche, 2015). However, when it comes to breakup, it is also relevant to understand what flow structures are  
 preferentially sampled by the aggregates. To quantify this, we make use of a mixing index parameter  $\lambda$  (Yang  
 & Manas-Zloczower, 1992):

$$\lambda = \frac{\sqrt{II_{\mathbf{E}^\infty}}}{\sqrt{II_{\mathbf{E}^\infty} + \sqrt{II_{\mathbf{Q}^\infty}}}} \quad (13)$$

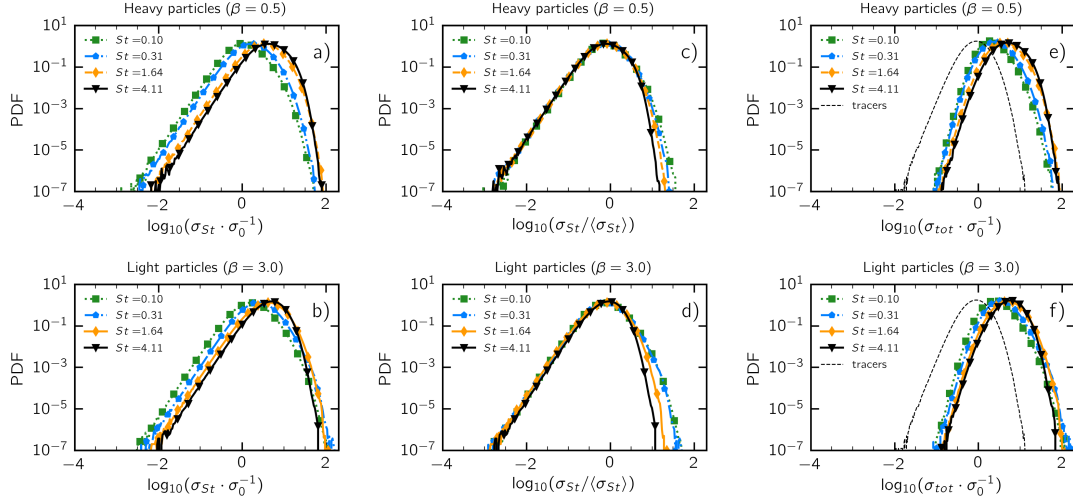


Figure 5: Probability density function of the fluid dynamic stresses for heavy and light particles at varying Stokes number. Panel a) and b) report the PDFs of the drag stress, normalized by the average shear stress experienced by tracers  $\sigma_0$ . In panel c) and d) the same PDFs are rescaled by the average values of the drag stress of each particle family. Panel e) and f) report the total stress distributions. In these two panels, the black dotted line is the PDF of the shear stress experienced by tracers.

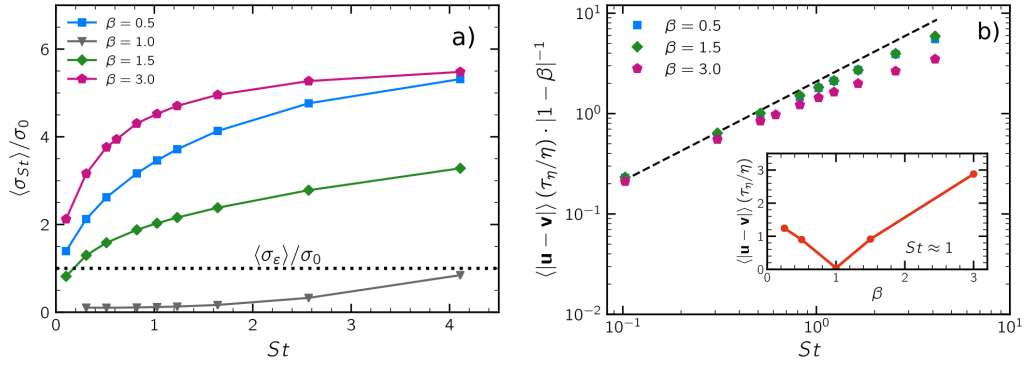


Figure 6: a) Drag stress for various  $\beta$  values as a function of  $St$ . The dotted line reports the average  $\sigma_{\mathcal{E}}$  experienced by tracers. b) Log-log plot of the fluid-particle slip velocity as a function of the Stokes number, for various  $\beta$  values. The slip velocity is made dimensionless by the Kolmogorov time  $\eta$  and length scale  $\tau_\pi$ . The black dashed lines show the theoretical scaling in the limit of small Stokes number  $\langle |\mathbf{u} - \mathbf{v}| \rangle \propto St|1 - \beta|$ . In the inset the slip velocity is reported as a function of the buoyancy parameter at fixed  $St \approx 1$ .

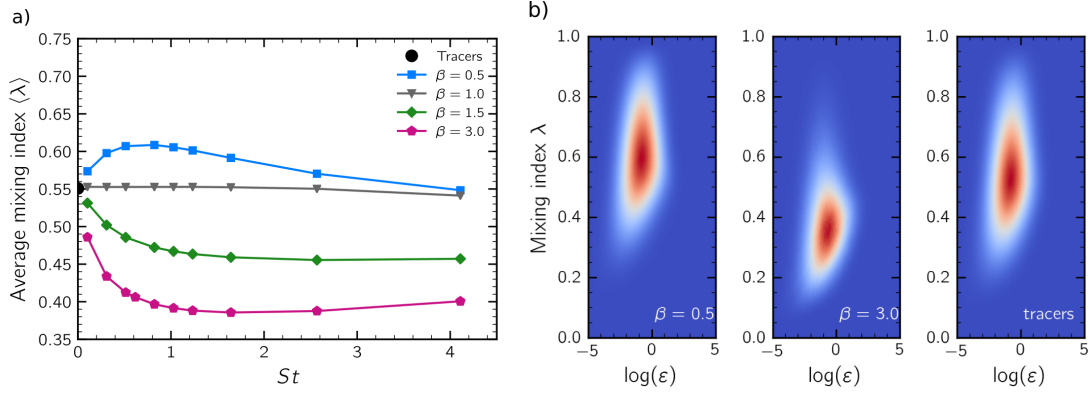


Figure 7: a) Average mixing index seen by aggregates as a function of both  $St$  and  $\beta$ . The black circle indicates the tracer behavior. b) Bivariate distribution function of the turbulent dissipation rate and mixing index for heavy aggregates ( $\beta = 0.5$ ), light aggregates ( $\beta = 3$ ) and tracers. Stokes number is approximately equal to 1 for both the light and the heavy aggregate class.

281 where  $II_{\mathbf{E}^\infty}$  is the second invariant of the rate-of-strain tensor  $\mathbf{E}^\infty$  and  $II_{\mathbf{\Omega}^\infty}$  is the second invariant of the  
282 vorticity tensor  $\mathbf{\Omega}^\infty = 0.5 (\nabla \mathbf{u}^\infty - \nabla \mathbf{u}^{\infty, T})$ . This parameter has a 0–1 range, with 0 indicating a pure rotational  
283 motion, and 0.5 and 1 indicating pure shear and pure elongational flow, respectively. Figure 7a reports the  
284 average mixing index  $\lambda$  seen by the aggregates for different Stokes numbers and buoyancy parameter  $\beta$ . The  
285 black dot at  $\lambda = 0.55$ ,  $St = 0$  indicates the behavior of tracers. The plot makes apparent that, regardless of the  
286  $\beta$  parameter, small Stokes aggregates behave very similarly to tracer aggregates with an average mixing index  
287 close to 0.55. As the Stokes number increases, ~~qualitative differences can be observed~~: neutrally buoyant  
288 aggregates ( $\beta = 1$ ) keep behaving as tracer aggregates with the average  $\lambda$  almost constantly equal to 0.55. On  
289 the contrary, the heavy aggregates ( $\beta = 0.5$ ) present a maximum in  $\langle \lambda \rangle$  for  $St \approx 1$  and converge to the tracers  
290 behaviour again as Stokes increases. Light aggregates ( $\beta = 1.5$  and  $\beta = 3$ ) have a minimum  $\langle \lambda \rangle$  at  $St \approx 1$ ,  
291 which is kept almost constantly up to  $St = 4$ . This feature can be explained by the fact that at increasing  
292 relaxation times the particle motion is no longer correlated to the topological feature of the flow. These  
293 behaviors confirm what already pointed out by other researchers (Calzavarini et al., 2008b,a), who observed  
294 that light aggregates preferentially sample the vortical regions of the flow (i.e., lower  $\lambda$  regions), whereas  
295 heavy aggregates are ejected from vortical regions and tend to sample the higher strain regions (larger  $\lambda$ ). This  
296 phenomenon, referred to as *turbulence induced segregation*, is here observed to be particularly important  
297 at  $St \approx 1$ , in line with what reported by Bec et al. (2005). In Figure 7b for  $St \approx 1$ , we report the joint  
298 probability distribution functions of the mixing index and turbulent dissipation rate  $\epsilon$ , which finally make  
299 visually apparent the preferential sampling of the  $\lambda$ -space at varying buoyancy.

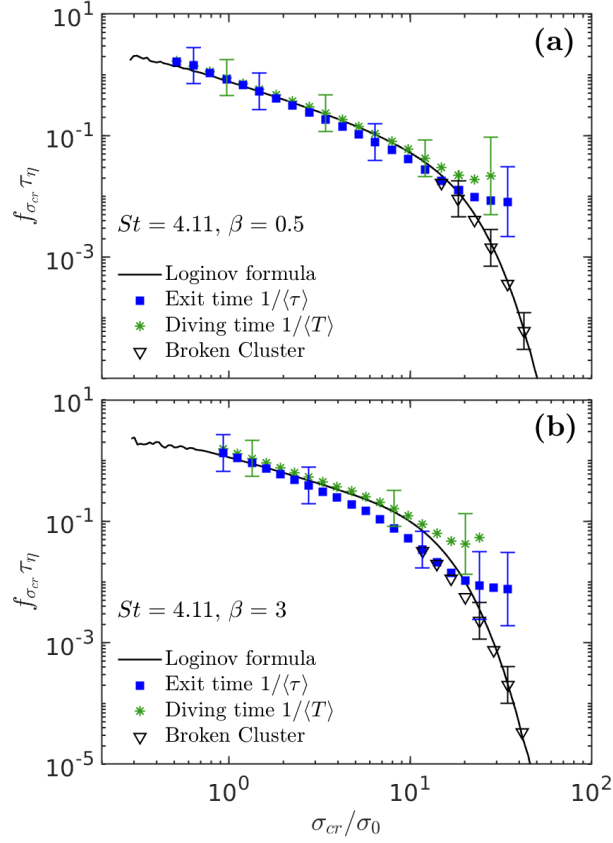


Figure 8: Breakup rate versus the threshold stress for aggregates with  $St = 4.11$  and (a)  $\beta = 0.5$  and (b)  $\beta = 3$ . Breakup rates are measured by the exact expression based on the exit time (square symbols) and based on the approximations obtained by using the diving time (asterisk symbols), the concentration decay (triangles) and the Loginov's formula (lines). The horizontal axis is normalized by the mean shear stress  $\sigma_0$ , whereas the vertical axis is normalized by the Kolmogorov time scale  $\tau_{\eta}$ . For the sake of clarity, error bars are shown for few data points only.

### 3.2. Breakup statistics

We compute breakup rates by assuming that aggregates are brittle objects that break instantaneously as soon as they experience for the first time a total hydrodynamic stress exceeding their critical strength  $\sigma_{cr}$ . We make use of the different approaches outlined in Section 2.2 and in Fig. 8 we report the breakup rate as a function of the threshold critical stress for two particle families with the same Stokes number ( $St = 4.11$ ) and different buoyancy parameters, namely  $\beta = 0.5$  (panel a) and  $\beta = 3$  (panel b). The threshold stress on the horizontal axis is normalized by the mean shear stress  $\sigma_0$  and the breakup rate is normalized by the Kolmogorov time scale  $\tau_\eta$ . Although the data shown refers to particle families on the outskirts of our dataset (see Fig. 1), the graph reflects the general trend of the breakup rate and allows us for discussing its characteristics and the different approaches used for measuring it.

The total stress experienced by an aggregate is calculated from the three components of the particle-fluid slip velocity and from the spatial derivatives of the fluid velocity, that are both quantities that fluctuate along a trajectory. Thus, for a fixed critical strength of the aggregate, the breakup rate have to be considered as an outcome of a combination of random variables. For small threshold values of the critical stress, this combination leads to a breakup rate following a power-law behavior: in this regime, in fact, breakup events are controlled by hydrodynamic stresses that are lower or close to the mean stress and that occur in the smooth regions of the flow, where the stress statistics are Gaussian distributed. As the strength of the aggregates increases, the curve of the breakup rate has a sudden super-exponential drop-off. This regime is controlled by the rare turbulent events vigorous enough to break the strong aggregates and whose occurrence is controlled by turbulent intermittency.

Regarding the different approaches for measuring the breakup rate, it is observed that for small threshold values the exact breakup rate based on the exit time (square symbols) is very close to the proxy-breakup rate based on the diving time, that in Fig. 8 is shown by both its discrete measurement (asterisk symbols) and its analytical extension provided by the Loginov's formula (line). We also note that for large thresholds values, for which breakup is governed by the rare and intense turbulent events, there are not enough statistics to measure the mean exit and mean diving time with confidence. In other words, the length of exit and diving events becomes comparable or larger than the run-time of our simulation. This causes the data for the breakup rate based on the exit time and on the diving time to level off or even to increase. In comparison, the Loginov's formula (solid line) keeps on decaying, thus giving the more realistic picture.

To understand the similarity between the breakup rate computed by exit times and the proxy-breakup rate based on the diving time, we analyze their PDFs in some detail. Figure 9a shows the PDF of the diving time for the same particle family as shown in Fig. 8a and for two threshold values. We observe that the PDF has a sharp decrease for small diving times followed by a well developed exponential tail. The presence of the exponential tail implies that the realization of diving events whose duration exceeds the correlation time of the

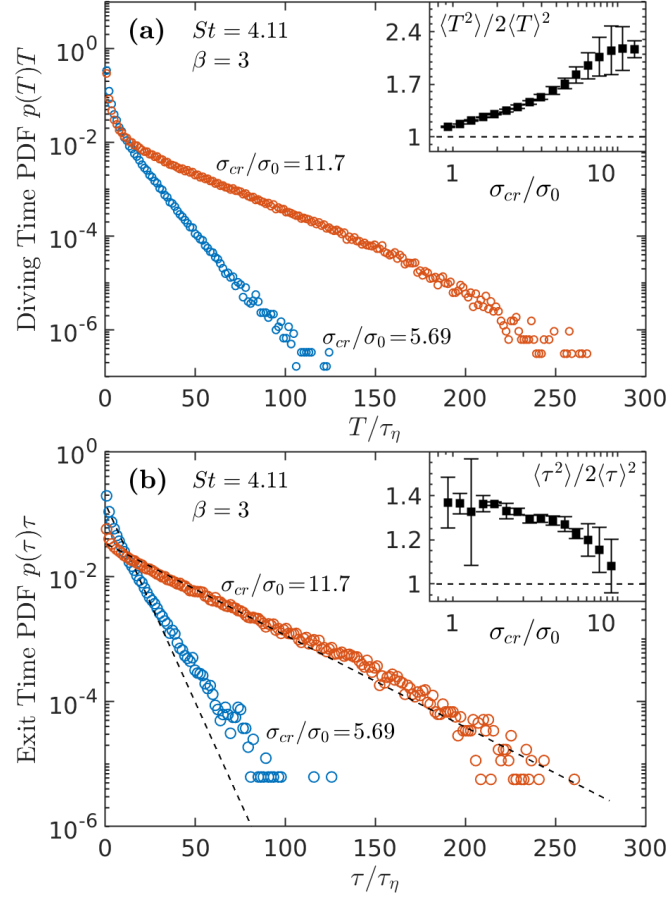


Figure 9: Probability density function for aggregates with  $St = 4.11$  and  $\beta = 3$  of (a) the diving time and (b) the exit time at two critical stress values. The dashed lines in (b) show exponential distributions with an expected value equal to the mean exit time for the given thresholds. In both plots, the inset shows the normalized second order moment of the distributions as a function of the critical stress.

334 hydrodynamic stress can be described as a Poisson process, where individual diving events are independent  
 335 of each other. On the contrary, the deviations for the shorter diving times reflect the correlation of short-lived  
 336 turbulent fluctuations, which often come with multiple spikes in the stress intensity, as visible in Fig. 3. To  
 337 quantify the deviation from an exponential distribution we report in the inset of Fig. 9a the moment ratio  
 338  $\langle T^2 \rangle / (2\langle T \rangle)$ . For an exponential distribution, this moment ratio has a value of unity. As it can be seen, the  
 339 moment ratio of the diving time PDF is close to one for weak aggregates and it increases as  $\sigma_{cr}$  grows.

340 The deviations from the exponential distribution for short diving events, that are particularly pronounced  
 341 for the larger value of the threshold shown in Fig. 9a, result in the diving time being different from the exit  
 342 time. An exactly exponentially distributed diving time would lead to an equivalence of the mean diving time  
 343 and the mean exit time, i.e.  $\langle T \rangle = \langle \tau \rangle$  (this follows from using  $p(T) \sim e^{-t/\langle T \rangle}$  in Eq. (7)).

344 The first order process given by Eq. (10) assumes that the events that cause breakup (i.e. the intense  
 345 fluctuations in the hydrodynamic stress) are independent from each other. Moreover, the dynamics underlying  
 346 Eq. (10) is equivalent to an exit time that has an exponential distribution. This is explored in Fig. 9b that shows  
 347 the PDF of the exit time for two threshold values. It is seen that the exit time is approximately exponentially  
 348 distributed, with small deviations at short exit times, and at small threshold values for large exit times. To  
 349 assess the deviations from an exponential distribution in the inset of Fig. 9b the moment ratio of the exit time,  
 350 i.e.  $\langle \tau^2 \rangle / (2\langle \tau \rangle)$ , is shown as a function of the threshold stress. For an exponential distribution, this has a value  
 351 of unity. As it can be seen, at increasing threshold stress, the normalized second order moment approaches  
 352 one, implying that the PDF of the exit time approaches an exponential distributions and hence, the dynamics  
 353 proposed by Eq. (10) become more accurate. This latter observation is also confirmed by the breakup rate  
 354 in Fig. 8. Here, the open triangle symbols show the estimate based on Eq. (10), from which we see that, for  
 355 intermediate threshold values, for which both the exit time measurement and the concentration decay can be  
 356 measured with confidence, we find a good agreement between the two. Moreover, for large threshold values  
 357 the estimate based on the concentration decay (open triangles) is in agreement with the Loginov's formula  
 358 (lines), a result which is in line with Eq. (11). Based on these findings, in the following, we will report the  
 359 breakup rate in terms of the exact expression based on the exit time for small and intermediate thresholds,  
 360 that is for breakup rates  $f_{\sigma_{cr}} \tau_{\eta} > 0.01$ , whereas for large thresholds, we will report the estimate from the  
 361 decaying particle concentration of Eq. (10).

### 362 3.3. Breakup rate of inertial aggregates

363 Figure 10 shows the breakup rate as a function of the threshold stress for neutrally buoyant aggregates  
 364 with varying Stokes number. The data shows a clear change of behavior of the breakup rate. We explain this  
 365 as the consequence of a transition from a shear-dominated (at small  $St$  values) to a drag-dominated breakup  
 366 regime (at larger  $St$  values). Shear stresses are relatively weak and, accordingly, only weak aggregates are

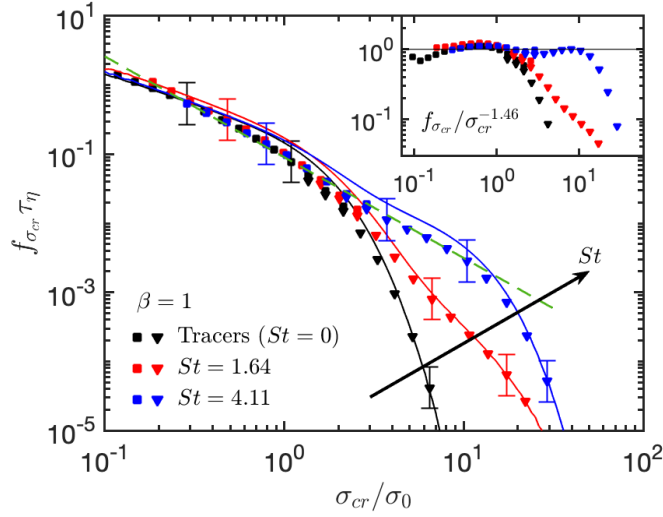


Figure 10: Breakup rate as a function of the threshold stress for  $\beta = 1$  and three different Stokes numbers measured by the exact expression based on the exit time (Eq. (6), square symbols) and on the broken cluster approach (Eq. (10), triangles). Solid lines report the breakup rate measured by the Loginov's formula (Eq. (9)). The dashed line shows the fitted power-law expression  $f_{\sigma_{cr}} \sim \sigma_{cr}^{-k}$ , with  $k = 1.46 \pm 0.04$ . The inset shows the breakup rate compensated by the power-law expression.

367 broken down by shear. This is the reason why an early drop-off of the breakup rate occurs for tracer aggregate.  
 368 For instance, this makes a tracer aggregate of strength  $\sigma_{cr} \sim 10 \sigma_0$  already unlikely to be broken down. As  
 369 the Stokes number increases, the slip velocity grows and gives origin to an additional drag stress acting on  
 370 the aggregate. This makes it possible to induce the breakup of even the stiffest aggregates, such that in this  
 371 case an inertial aggregate with strength  $10 \sigma_0$  has a substantial breakup frequency, as made apparent by the  
 372 two non-tracer curves in Fig. 10.

373 It is interesting to notice that despite the additional contribution of the drag stress, the power-law part of  
 374 the breakup rate is not affected by inertia, i.e., for small threshold stresses, the breakup rates for neutrally  
 375 buoyant aggregates at varying Stokes and for tracers collapse to a single master curve following a power-law of  
 376 the form  $f_{\sigma_{cr}} \tau_{\eta} = 0.090 (\sigma_{cr} / \sigma_0)^{-k}$ , where  $k = 1.46 \pm 0.04$ , as shown by the green dashed curve in Fig. 10.  
 377 The inset of Fig. 10 shows the breakup rate compensated by this power-law expression, making apparent the  
 378 quality of the power-law master curve. However, a rationalization of the value of  $k$  is still lacking. This will  
 379 be subject of future studies.

380 The breakup rate for heavy and light inertial aggregates is shown in Fig. 11. For these aggregates, the  
 381 drag stress is dominant and causes an increase of the breakup rate at already small Stokes numbers. As the  
 382 Stokes number further increases, the breakup rate for both heavy and light aggregates saturates. For heavy  
 383 particles ( $\beta = 0.5$ , Fig. 11a) the breakup rate reaches a maximum. This maximum, made apparent by the  
 384 overlap of the curves for  $St = 1.64$  and  $St = 4.11$  in Fig. 11a, follows again a power-law for the small threshold

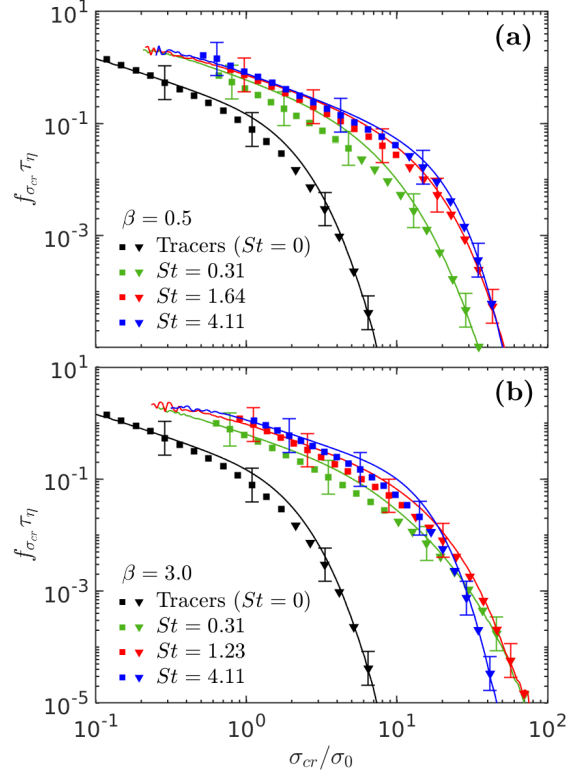


Figure 11: Breakup rate for varying aggregate critical strength. Data refer to heavy aggregates with  $\beta = 0.5$  (a) and light aggregates with  $\beta = 3.0$  (b).

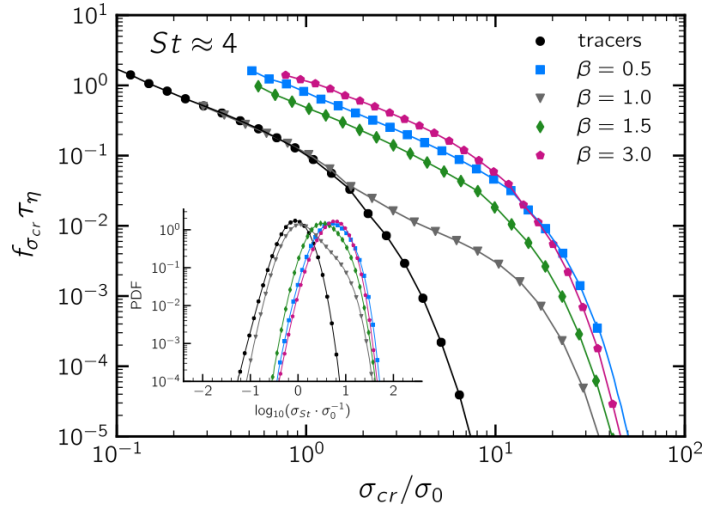


Figure 12: Breakup rate as a function of the threshold stress for  $St = 4$  particles and four buoyancy parameters, measured by the exact expression based on the exit time for  $f_{\sigma_{cr} \tau_{\eta}} > 0.1$  and on the broken cluster approach for  $f_{\sigma_{cr} \tau_{\eta}} < 0.1$ . The inset shows the PDF of the total stress.

385 values and has a sharp super-exponential cut-off at larger thresholds. For light particles ( $\beta = 3$ , Fig. 11b) the  
386 overlap at large Stokes numbers does not hold and we even observe a reduction in the breakup rate for large  
387 threshold stresses and large Stokes numbers. This, made apparent by the sharp drop-off at  $St = 4.11$ , well  
388 agrees with the PDFs of the total stress reported in Fig. 5f. Careful inspection of the PDFs had revealed in  
389 fact a slightly wider right tail at intermediate Stokes numbers (i.e.  $St = 1.64$ ) compared to the largest Stokes  
390 number studied in our work ( $St = 4.11$ ). This implies that the most intense burst of the total stress are more  
391 frequent for intermediate Stokes numbers than for large Stokes numbers, i.e., light particles with large Stokes  
392 number exhibit a mild filtering effect of the largest drag stresses, that causes the sharper cut-off of the breakup  
393 rate for  $St = 4.11$  visible in Fig. 11b.

394 Finally, we analyze the statistics of the breakup rate at fixed  $St$  and variable buoyancy parameters. Figure 12  
395 reports the breakup rate together with the corresponding PDFs of the total stress (see inset of Figure 12).  
396 Analyzing both the stress and breakup statistics, we observe that the breakup rate for light particles ( $\beta = 3$ ) is  
397 larger than the one of the heavy particles over nearly the whole range of critical stresses, except for very large  
398 values of  $\sigma_{cr}$  where we observe that the breakup rate of heavy particles exceeds the one of light particles.  
399 This observations hints to a situation where light particles moving in the flow filter out large fluctuations in  
400 the drag stresses, leading eventually to a reduced breakup rate compared to heavy particles. This effect is  
401 significant for a Stokes value  $St \approx 4$ , and it is not apparent at the smaller Stokes numbers of our dataset,  
402 where extremely light particles ( $\beta = 3$ ) have large breakup rates compared to both heavy and moderately light  
403 particles for any value of the critical stress.

#### 404 **4. Conclusions**

405 In this work we have studied the stress and breakup statistics of inertial aggregates in homogeneous  
406 isotropic turbulence, at varying fluid-to-particle density ratio and Stokes number. We have solved the flow  
407 dynamics by a direct numerical simulation and tracked the particles trajectories by evolving a minimal  
408 formulation of the Maxey-Riley-Gatignol equation. We have deemed both shear stress and drag stress as  
409 responsible for the particle breakup and we have assumed breakup to occur in a brittle manner, i.e., aggregates  
410 break instantaneously when they experience a fluid dynamic stress larger than their mechanical resistance.  
411 We have devised and tested different approaches for measuring the breakup frequency, discussing in detail  
412 their theoretical foundations and limitations.

413 Two distinct breakup regimes exist, depending on the aggregate mechanical strength. Loose aggregates  
414 have large breakup frequencies and are broken down in the smooth regions of the flow, where the stresses are  
415 Gaussian distributed, thus making the breakup frequency to follow a power-law behaviour with the aggregate  
416 strength. Conversely, strong aggregates have lower breakup rates and are broken down by the burst of the  
417 hydrodynamic stresses, which are dictated by the turbulent intermittency.

418 Results have also shown that inertial effects have a major role in determining breakup rates. When  
419 inertial effects are limited (i.e., for neutrally buoyant particles with small Stokes number), particles behave  
420 very similarly to tracers: they similarly sample the flow topology and their breakup is mostly dictated by the  
421 shear stress statistics. However, as soon as deviations from neutral buoyancy occur, inertial effects become  
422 dominant even at small Stokes numbers and have been seen to be able to cause the breakup of even the most  
423 resistant aggregates. A quantitative characterization of the flow topology seen by particles has also been  
424 conducted, and seen to agree with previously reported data for turbulence induced segregation.

425 To conclude, we point out that in the minimal formulation used in this work, some ingredients of the full  
426 equation of particle motion are not considered. This allowed us to keep the parameters space small. History  
427 forces, a possible correction for the drag coefficient and the curvature of the velocity field at the particle  
428 scale may be relevant, in particular when considering neutrally buoyant particles. However, the addition of  
429 these effects in the equation of particle motion has to be considered together with the substantial increase of  
430 numerical complexity and growth of the parameters space.

431 The role of the internal structure of the aggregates was disregarded, and aggregates were considered to be  
432 spherical assemblies of small contacting particles, treated as point-particles, with no specific modeling of their  
433 internal connectivity and colloidal particle-particle interactions, and a single internal parameter (the critical  
434 strength) was considered for evaluating the breakup occurrence. While this approach has been frequently  
435 used and allows us to infer stress statistics in a rather simple way, it misses to address the effect that the  
436 aggregate fractal features and restructuring mode can have on the breakup behavior of the aggregates. To take  
437 into account these, more complex models addressing simultaneously the dynamics of the turbulent stress (e.g.  
438 by DNS) and the aggregate response (e.g. by discrete element methods/Stokesian dynamics) are required.

439 Our investigation puts the basis for further developments in the measure and modeling of breakup in  
440 turbulence. Laboratory and numerical investigations aimed at assessing the outcome of breakup events in  
441 terms of the fragment size distribution would complement our findings, and provide the full information  
442 needed for calibrating macroscopic population balance models addressing breakup in homogeneous solid-  
443 liquid turbulent flows.

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450 **References**

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