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Dispersion of finite-size particles probing inhomogeneous and anisotropic turbulence

C.A. Mériaux^{a,*}, Miguel A. C. Teixeira^b, Joe J. Monaghan^c, Raymond Cohen^d, Paul Cleary^d

^a*School of Earth, Atmosphere and Environment, Monash University, Clayton, Australia*

^b*Department of Meteorology, University of Reading, Reading, UK*

^c*School of Physics and Astronomy, Monash University, Clayton, Australia*

^d*CSIRO Data61, Commonwealth Scientific and Industrial Research Organisation, Clayton, Australia*

Abstract

A series of 8 laboratory experiments was used to investigate the dynamics of a few almost neutrally-buoyant finite-size particles in the entire volume of a rectangular tank open to air and filled with water. Stirring was achieved by a cylinder executing a two-dimensional periodic Lissajoux figure. The rate and direction of stirring by the cylinder was varied. The particle motions were analyzed using a tracking method developed for the experimental design. The Reynolds number associated with the large-scale stirring motion was in a turbulent range of $[5, 693 - 11, 649]$ across all experiments. The absence of stirring in the direction of the cylinder axis, the constant interference of the cylinder with the eddies and the presence of walls and the free-surface resulted in a flow that was both inhomogeneous and anisotropic as recorded by the particle motion. Despite these unusual conditions, the single-particle dispersion across all experiments could be seen to follow a ballistic regime until

*Corresponding author

Email address: `catherine.meriaux@monash.edu` (C.A. Mériaux)

about two-fifths of the particle Lagrangian velocity auto-correlation time T_L . It was followed by a brief diffusive regime between T_L and $2.5T_L$, after which the presence of the boundaries prevented further dispersion. Such evolution is consistent with classic predictions for fluid tracer dispersion in homogeneous and isotropic turbulence. Particle-pair dispersion was more complex. Both the fixed time-averaged and length-scale-dependent particle-pair dispersion rates averaged across pairs showed the ballistic dispersion regime, whereas the subsequent diffusive regime was better borne out by the length-scale-dependent particle-pair dispersion. A super-diffusive Richardson regime was not unmistakably detected. Substantial variability was however found across the different pairs of particles, which was linked to differences in the decorrelation time of the velocity difference as a result of the inhomogeneity of the turbulence. For short initial separations, some particle pairs had a better separation of the time scales delimiting the ballistic and diffusive regimes and showed hints of a brief Richardson regime.

Keywords: Turbulence, Dispersion, Particle mixing, Experimental modelling

1. Introduction

In many contexts, from natural systems to industrial processing, the transport, dispersion or mixing of particulate matter in turbulent flows comes into play. In the oceans, understanding how the wind mixes an ever increasing number of floating plastic fragments down into the water is at the heart

6 of estimating how much plastic waste exists in our oceans. In industry, the
7 transformation of ingredients fed to a vessel and stirred by the motion of an
8 impeller is determined by judiciously choosing an impeller and its motion
9 that will create a homogeneous end-product so that it can pass its primary
10 quality control. In realistic contexts, turbulence is prone to be inhomoge-
11 neous and anisotropic.

12 Yet, although numerous applications have motivated intense research on
13 these topics, theoretical, numerical or experimental studies have mostly fo-
14 cused on the behaviour of fluid parcels within turbulent flows (e.g. Toschi and
15 Bodenschatz, 2009; Salazar and Collins, 2009; Balachandar and Eaton, 2010).
16 Theoretically, Kolmogorov (1941a,b) showed that, in three-dimensional (3D)
17 homogeneous and isotropic turbulence, energy cascades from the larger scales,
18 where energy is injected, down to a length scale η , at which dissipation by
19 molecular viscosity becomes important. In the Eulerian reference frame (i.e.
20 in terms of variables defined at points fixed in space), the energy spectrum
21 $E(k)$ as a function of wavenumber k follows $E(k) \sim \varepsilon^{2/3} k^{-5/3}$, where ε is
22 the energy dissipation rate, in the inertial range, which lies in between the
23 production and dissipation scales, $k_f < k < k_\eta$, where k_f is the forcing
24 wavenumber and $k_\eta \sim 1/\eta$ is the dissipation wavenumber. In the same
25 range, the equivalent to the -5/3 law can be expressed in physical space by
26 velocity structure functions of order 2, satisfying $C_2(l) = \Lambda C_K \varepsilon^{2/3} l^{2/3}$, where
27 l is the spatial separation, C_K is a universal constant found to be equal to
28 2.01 for homogeneous and isotropic turbulence and Λ is a constant equal
29 to 1 in the case of a longitudinal structure function and 4/3 in the case of
30 a transverse structure function (Sreenivasan, 1995). For $k > k_\eta$, viscosity

31 becomes important, and $E(k)$ rapidly decays. In the Lagrangian reference
 32 frame (i.e. in terms of time-dependent variables following particles originat-
 33 ing at position \mathbf{y} and with velocity $\mathbf{U}(\mathbf{x}(\mathbf{y}, t), t)$), properties of homogeneous
 34 and isotropic turbulence are characterized by the velocity structure functions
 35 $D(\tau)$ representing the variance of temporal increments of any velocity com-
 36 ponent U , $D(\tau) = \langle \delta U(\tau) \delta U(\tau) \rangle$, where $\delta U(\tau) = U(t + \tau) - U(t)$, and the
 37 velocity frequency spectrum $E(\omega)$, defined as the Fourier cosine transform
 38 of the velocity autocovariance $R(\tau) = \langle U(t)U(t + \tau) \rangle$. In the inertial range,
 39 $D(\tau)$ is predicted to scale as $D(\tau) = C_0 \varepsilon \tau$ and $E(\omega)$ as $E(\omega) = (C_0/\pi) \varepsilon \omega^{-2}$,
 40 and the constant C_0 has been found to equal 5 (Monin and Yaglom, 2013;
 41 Ouellette et al., 2006).

42 When particles are neutrally buoyant and small compared to the Kol-
 43 mogorov dissipative length scale η , they behave as tracers of the fluid motion
 44 by passively following the flow. The upper size limit d_p for tracer behaviour
 45 was determined to be $d_p = 5\eta$ (e.g. Qureshi et al., 2007; Volk et al., 2011).
 46 Dispersion of tracer-like particles in homogeneous and isotropic 3D turbu-
 47 lence differentiates single-particle dispersion, which is defined by the mean-
 48 square displacement of a particle from its initial position, from particle-pair
 49 dispersion or relative dispersion, which involves the mean-square separation
 50 of a pair of particles. In the dispersion of a single particle, also called Taylor
 51 dispersion, the mean-square displacement varies as t^2 for short times (ballistic
 52 regime) and is proportional to t in a long-time diffusion limit (Taylor, 1922;
 53 Einstein, 1956). The particle-pair or relative dispersion, however, has been
 54 described by three regimes (Batchelor, 1950; Richardson, 1926; Csanady,
 55 1973; Bourgoïn, 2015). In the inertial regime, where the initial separation

56 between two particles, $|\mathbf{S}| = S_0$, is greater than η , a ballistic regime is ex-
 57 pected, for which $\langle(|\mathbf{S}| - S_0)^2\rangle \propto t^2$ if $t \ll t_0 = (S_0^2/\varepsilon)^{1/3}$. The time t_0 is
 58 identified as the time for which the two fluid elements “recall” their initial
 59 relative velocity when moving in an eddy of size S_0 . When $t_0 \ll t \ll T_L$,
 60 where T_L is the Lagrangian velocity auto-correlation time, an intermediate
 61 super-diffusive regime, also named the Richardson regime, is expected, for
 62 which $\langle(|\mathbf{S}| - S_0)^2\rangle \propto t^3$. Physically, this is caused by the fact that the scale
 63 of the eddies contributing to relative dispersion, which in this phase lies in
 64 the inertial range, is proportional to the separation between the dispersing
 65 particles. Finally, when $t \gg T_L$, i.e. when the particle separation equals or
 66 exceeds the scale of the dominant, energy-containing eddies in the turbulence,
 67 the particles are expected to separate diffusively as $\langle(|\mathbf{S}| - S_0)^2\rangle \propto t$. An alter-
 68 native to the previous fixed time-averaged indicators of relative dispersion is
 69 a length-scale-dependent dispersion rate, which is defined through the finite-
 70 scale Lyapunov exponent (FSLE). Given the spatial separation δ between two
 71 particle trajectories and the mean time $\langle\tau(\delta)\rangle$ that δ takes to be amplified by
 72 a factor ρ , then the (Lagrangian) FSLE is defined as $\lambda(\delta) = \ln \rho / \langle\tau(\delta)\rangle$. Di-
 73 mensional arguments further establish that if $\langle|\mathbf{S}|^2\rangle \propto t^{2/\zeta}$, then $\lambda(\delta) \propto \delta^{-\zeta}$
 74 (Aurell et al., 1996; Boffetta et al., 2000). Boffetta and Sokolov (2002) showed
 75 that the advantage of averaging at a fixed scale separation, as opposed to at a
 76 fixed time, is that it removes crossover effects since all sampled particle pairs
 77 belong to the same scales and as a result they allow a better identification of
 78 the super-diffusive Richardson regime.

79 The experimental study of particle motion in turbulence has developed
 80 substantially in the last decade with the use of new optical (e.g. La Porta

81 et al., 2001) and acoustic (e.g. Mordant et al., 2004) tracking techniques.
 82 The synchronization of multiple fast cameras or ultrasonic/laser Doppler ve-
 83 locimetry allows fully resolving the 3D particle trajectories in turbulent flows,
 84 but the measurements are limited to time intervals of a few Kolmogorov
 85 times. In many experiments, the von Kármán apparatus is used (e.g. Zand-
 86 bergen and Dijkstra, 1987; Mordant et al., 2003; Gibert et al., 2010). This
 87 is a closed flow chamber filled with a carrier fluid and consisting of two-
 88 counter rotating disks generating the turbulence. Properties of turbulence
 89 are inferred from hot anemometry or tracer-like particles. The observation
 90 volume is commonly limited and selected relatively far from the disks to
 91 avoid anisotropy and inhomogeneity in the turbulence. In such ideal turbu-
 92 lent conditions, both laboratory experiments and numerical simulations have
 93 confirmed the theoretical predictions on the dispersion of tracers (Bourgoin,
 94 2015; Xia et al., 2019; Boffetta and Sokolov, 2002; Biferale et al., 2008; Bi-
 95 tane et al., 2012). In particular, these studies have shown that observation
 96 of the Richardson regime requires a significant scale separation between the
 97 different lengths, η , S_0 , L_i , where L_i is the integral length scale, a statisti-
 98 cally characteristic length related to the largest energy-containing eddies in
 99 the turbulence ($L_i \propto 1/k_f$).

100 Otherwise, experimental studies (Zimmermann et al., 2011; Fiabane et al.,
 101 2012; Qureshi et al., 2007; Bourgoin et al., 2011) have typically investigated
 102 the behaviour of particles in a size and density range of $d_p \sim [5 - 30]\eta$ and
 103 $\rho_p = [1 - 70]\rho_a$, where ρ_a is the density of the ambient fluid, respectively.
 104 Qureshi et al. (2007) and Bourgoin et al. (2011) especially showed that the
 105 inertia of finite-size particles primarily affects their acceleration, whereas

106 their Lagrangian velocity statistics are almost similar to those of tracers.
 107 With the exception of the studies by Klein et al. (2012) and Machicoane
 108 and Volk (2016), much less attention has been given to particles with $d_p \sim$
 109 $\mathcal{O}(100)\eta \sim \mathcal{O}(10^{-1})L_i$. The present study falls within this context.

110 Our experimental study was designed to examine the dispersion of large-
 111 sized particles (compared to fluid tracers) in the entire volume of a rect-
 112 angular open tank filled with water, in which turbulence was generated by
 113 moving a cylinder of diameter $2R$ and length L very similar to the depth of
 114 the tank along a periodic Lissajoux figure. In this setting, the turbulence
 115 is neither homogeneous nor isotropic, as boundary-layers at the walls and
 116 the free-surface are part of the volume of study, the cylinder is constantly
 117 interfering with the turbulent vortices, and the stirring is two-dimensional
 118 (2D), as no forcing is imposed in the direction along the axis of the cylinder.
 119 Particles were slightly negatively buoyant spheroids and their concentration
 120 in the fluid carrier was low. Their size was $d_p \sim R$. Consequently, particles
 121 were only to respond to eddies of size $\geq d_p$ such as those produced by the
 122 cylinder and its wake, while being unaffected by any eddy of size $< d_p$.

123 Following the works of Qureshi et al. (2007) and Bourgoin et al. (2011),
 124 we assumed that the Lagrangian particle velocity statistics were essentially
 125 similar to those of tracers. In other words, we assumed that the velocity-
 126 based properties of the turbulence could be inferred from the velocities of the
 127 finite-size particles. The energy dissipation rate ε was thus derived from the
 128 particle motion. We found both *a priori* and *a posteriori* that this assumption
 129 was sensible. *A priori*, values of $\langle U^2 \rangle$ and of ε derived from the inertial
 130 range of spectra probed by the particles were estimated not to differ by

131 more than 20% from the corresponding exact fluid properties (see section
132 2.4). *A posteriori*, the regions in which the different dispersion regimes are
133 displayed are within the temporal and spatial limits derived using the energy
134 dissipation rate (see section 6.2). We stress however that the statistics of the
135 experiments relies on a few particles in a bounded domain and is obtained
136 from sampling in time via particle tracking.

137 The same stirring system and moving bodies were previously studied in
138 a two-dimensional context with Smooth Particle Hydrodynamics (SPH) nu-
139 merical models (Valizadeh and Monaghan, 2015; Monaghan, 2017; Monaghan
140 and Mériaux, 2018a,b). In the presence of bodies, not all but many of the
141 properties of the fluid could be estimated from the dynamics of those bodies.
142 For instance, the velocity auto-correlation times for the bodies and the fluid
143 were found to be similar.

144 The structure of this paper is as follows. The laboratory experiments
145 are described in §2, whereas the methodology of analysis is presented in §3.
146 Since the experimental setup has never been described before, section §4
147 details the particle dynamics, from which we establish the inhomogeneity
148 and anisotropy of the turbulence at the particle scale; Turbulence statistical
149 properties are further inferred in section §5. Analysis of particle dispersion
150 follows in section §6, and conclusions are gathered in §7.

151 **2. Laboratory experiments**

152 *2.1. Experimental setup*

153 The laboratory experiments were conducted in an Acrylic tank, $D = 0.3$
154 m long (x direction), $W = 0.3$ m wide (y direction), and 0.5 m high that

155 was filled with tap water up to a height $H = 0.3$ m (z direction). The tank
 156 itself was inserted into a metal wire frame secured to the experimental bench,
 157 at the top of which were fixed two electric actuator ball screw drives (SMC
 158 Pneumatics) mounted one over the other at right angles (Figure 1). These
 159 actuators were driven by two motors (Model AM8023 from BECKHOFF
 160 Automation), which were controlled by the TwinCAT software (BECKHOFF
 161 Automation). The two actuators were responsible for moving a cylinder in
 162 the tank in both the horizontal (y) and vertical (z) directions. The cylinder,
 163 which was hollow but capped at both ends, was hanging by a rigid rod of
 164 adjustable length from one of the actuators. Its centre was initially positioned
 165 at the tank mid-width, at a height of 0.15 m. The cylinder had a radius
 166 $R = 0.02$ m, and a length $L = 0.298$ m, so it was only 2 mm shorter than the
 167 length of the tank D . It had been coated with a black film for visualisation
 168 purposes.

169 2.2. Turbulence forcing

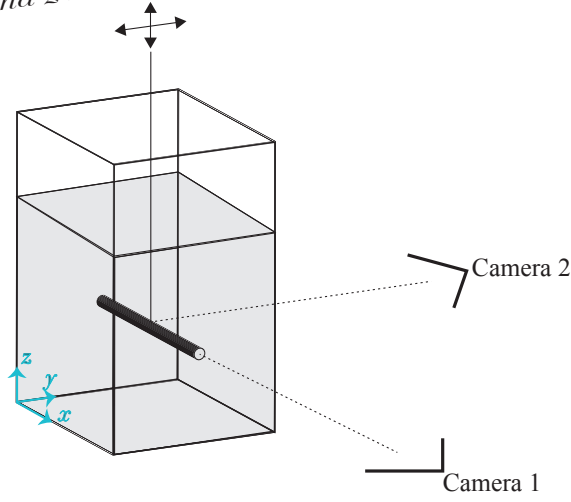
170 Turbulence was generated in the water by forcing the cylinder to follow
 171 a cyclic Lissajous loop defined by

$$172 \qquad y_c = y_c(0) + A \sin(2\pi t/T), \qquad (1)$$

$$173 \qquad z_c = z_c(0) \pm A \sin(4\pi t/T), \qquad (2)$$

174 where $y_c(0)$ and $z_c(0)$ are the initial y_c and z_c positions of the cylinder (see
 175 Figure 1). The amplitude A was fixed at 0.075 m, and the forcing period T
 176 varied within the range $T = 1.75\text{--}3$ s. The motion started initially either go-
 177 ing down to the right as shown in Figure 1 or going up to the left (reverse). We

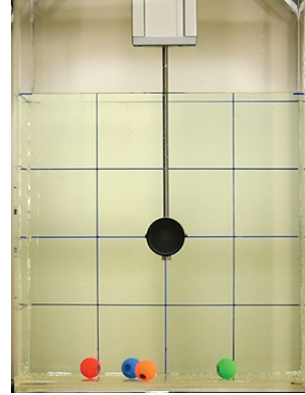
y and z motion driven by motors



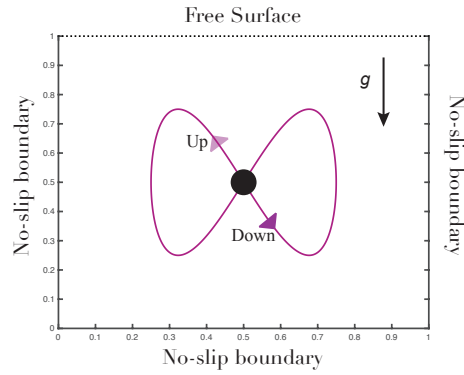
Spheroids



View from Camera 1



Cylinder motion



View from Camera 2

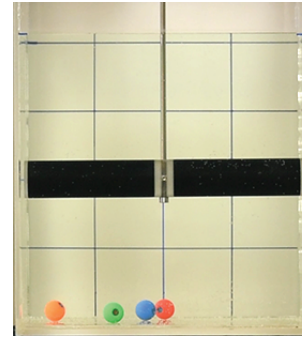


Figure 1: Experimental setup and coordinate system used, the Lissajous figure executed by the cylinder, and the finite-size particles used in the experiments. Note that the x direction is along the cylinder, and the y direction is horizontally across the cylinder. The z direction is vertical. The cylinder motion executing a Lissajous figure starts either going down to the right or up to the left. The finite-size particles used in the experiments 1-10 are slightly oblate spheroids.

178 identify those two initial directions of motion by down and up in Table 1. The
 179 velocity magnitude of the cylinder is given by $u_c = \sqrt{(dy_c/dt)^2 + (dz_c/dt)^2}$.
 180 In the two directions of motion, the absolute maxima of the cylinder vertical
 181 velocity are located at mid-height in the tank at points given in dimension-
 182 less units by $(y_c/W, z_c/H) = (0.25, 0.5), (0.5, 0.5)$, and $(0.75, 0.5)$ cm, and
 183 occur over a complete cycle at the times $2\pi t/T = 0, \pi/2, \pi, 3\pi/2$, and 2π
 184 (see Figure 1).

185 *2.3. Finite-size particles*

186 The finite-size particles, shown in Figure 1, were built from hollow plastic
 187 beads of different colours and are slightly oblate spheroids with an equatorial
 188 diameter $d_p = 2.21$ cm only 12% longer than the distance from pole to pole
 189 along the symmetry axis. Particles were filled with a single fishing weight
 190 and plasticine in order to be quasi-neutrally buoyant. The average density of
 191 the particles was 1015 ± 10 kg m⁻³ giving an excess of density of the particles
 192 relative to the ambient water of 1.7%.

193 *2.4. Experimental runs*

194 We report on eight experiments, which differed by the stirring period
 195 and direction of motion as detailed in Table 1. Across all experiments, the
 196 temperature of the water was $T_w = (20.9 \pm 1.1)$ °C. Changes of the exper-
 197 imental conditions due the temperature change could be neglected as they
 198 were equivalent to a change of less than 0.1% in water density ρ_a , less than
 199 7% in water dynamic viscosity μ_a and less than 1% in surface tension (see
 200 Vargaftik et al., 1983).

Experiment Id.	T_w °C	T s	Initial motion	N_c s	t_s
4	20.9	2.5	Down	128	51
5	22.4	3	Down	118	72
6	21.6	3	Down	102	78
7	21.6	2	Down	100	86
8	21.6	1.75	Down	121	86
9	19.45	3	Down	110	74
10	19.84	3	Up	85	56
11	19.84	1.75	Up	99	96

Table 1: Conditions for each run. N_c refers to the total number of collisions between the particles and the cylinder over 100 cycles of its motion, which was manually counted by systematically inspecting all the video recordings. t_s is the time at which the transient motion ends as defined in Appendix A.

201 As we changed the stirring rate, the first normal mode of sloshing, at
 202 which a single peak and trough of a free surface wave oscillated between the
 203 y vertical walls of the tank, was observed with a period of $T = 1.75$ s, but
 204 only when the motion of the stirrer was initially going down to the right.
 205 When the motion of the stirrer was reversed, initially going up to the left, we
 206 did not detect any sloshing mode, which points to a different interaction with
 207 the free surface in the two directions of stirring. The observed sloshing period
 208 was also larger than the predicted sloshing period of a fluid in a rectangular
 209 tank (Ibrahim, 2005), estimated as

$$210 \quad T_n^s = \frac{2\pi}{\sqrt{\frac{ng\pi}{W} \tanh\left(\frac{n\pi H}{W}\right)}}, \quad (3)$$

211 where n is the mode number. When $n = 1$, $T_1^s = 0.6211$ s. This difference
 212 is likely due to the presence of the cylinder in the fluid, which acts as an
 213 obstacle.

214 Eight experiments were performed using the four particles previously de-
 215 scribed. We did not use any tracers to follow the fluid. The experiments were
 216 characterized by a set of dimensionless numbers and characteristic length and
 217 time scales, which are given in Table 2, with the underlying assumption that
 218 the impact of the particles on the velocity fluctuations in the fluid was small.
 219 In this regard, at least two dimensionless numbers have been found to be
 220 important for assessing the effect of particles on turbulence intensity: the
 221 volume fraction of particles in the fluid ϕ_v and the ratio of the particle size
 222 to the integral length scale of the turbulence d_p/L_i (see for instance Balachan-
 223 dar and Eaton (2010) and Gore and Crowe (1989)). In our experiments, the

224 volume fraction of particles in the fluid was

$$225 \quad \phi_v = \frac{v_p}{V_f} = 4 \times \frac{\pi}{6} \left[\frac{d_p^3}{(DWH - \pi R^2 L)} \right], \quad (4)$$

226 where V_f , which coincides with the volume of measurement, is the total
 227 volume $D \times W \times H$ minus the volume of the cylinder $\pi R^2 L$ and v_p is the
 228 volume of the four particles

$$229 \quad v_p = 4 \times \left(\frac{\pi}{6} d_p^3 \right). \quad (5)$$

230 ϕ_v was 8.4×10^{-4} in the experiments, which is small. Such a volume fraction
 231 is, for example, below the threshold value of 1.4×10^{-3} , at which neutrally-
 232 buoyant Taylor-size spherical particles were shown to reduce by 15% the
 233 turbulent kinetic energy of the fluid (Bellani et al., 2012). Apart from ϕ_v , in
 234 our experiments, $d_p/L_i = 0.18 - 0.21$, which falls in the range where Gore
 235 and Crowe (1989) found that particles cause an increase in turbulence by
 236 not more than 20%. So, regardless of whether there is a slight increase or
 237 a decrease in turbulence due to the particles, the values of ϕ_v and d_p/L_i in
 238 our study imply that the modulation of the turbulence due to the particles
 239 should be limited.

240 Additionally, our study shares dynamic similarities with the studies by
 241 Bellani et al. (2012) ($\phi_v \ll 1$; $d_p/L_i = 0.11$), Qureshi et al. (2007) ($\phi_v \ll 1$;
 242 $d_p/L_i = 0.02 - 0.10$) and Bourgoïn et al. (2011) ($\phi_v \ll 1$; $d_p/L_i = 0.04 - 0.12$),
 243 which all showed little impact of the particles on the velocity fluctuations in
 244 the fluid. It however departs from these studies by its ratio of the particle size
 245 to the Kolmogorov scale d_p/η , which is of order $\mathcal{O}(100)$ and therefore greater
 246 than the ratios used in the previous studies, $d_p/\eta = [7, 30]$. In our study, the
 247 departure of the acceleration variance of particles from that of tracers in the

248 inertial range $\mathcal{R} = \langle a^2 \rangle_{particle} / \langle a^2 \rangle_{fluid}$ is estimated to be in the interval [0.16-
 249 0.22], following equations 2 and 3 of Qureshi et al. (2007), which predict $\mathcal{R} \sim$
 250 $6(d_p/\eta)^{-2/3}$. The impact of particle inertia on the acceleration is therefore
 251 substantial (80%). This impact on the acceleration does not however imply
 252 a substantial impact of the particles on the velocity fluctuations in the fluid,
 253 as shown previously by Qureshi et al. (2007) and Bourgoïn et al. (2011) when
 254 $d_p/\eta = [7, 30]$. The time integration by which the velocity is obtained from
 255 the acceleration acts like a low-pass filter ($\Phi(\omega) = \omega^2 E(\omega)$, where $\Phi(\omega)$ is
 256 the acceleration frequency spectrum), making the velocity be dominated by
 257 lower frequencies, less affected by inertia, compared to the acceleration. In
 258 practice, the relation between the velocity variance of the particles $\langle u^2 \rangle$ and
 259 the energy spectrum $E(k)$ can be defined as for tracers, but integrated over a
 260 narrower range of wavelengths that excludes scales smaller than the particle
 261 size, that is $(3/2)\langle u^2 \rangle = \int_{2\pi/L_i}^{2\pi/d_p} E(k) dk$. This truncation of the spectrum of
 262 the turbulence reflects the fact that particles do not respond to scales of fluid
 263 motion smaller than their own size. Calculations using the model spectrum
 264 adopted by Teixeira and Belcher (2000) for a range of Reynolds numbers of
 265 the order of magnitude of those used in the experiments, $Re = [10^2 - 10^4]$,
 266 actually indicate that this truncation does not lead to an underestimation of
 267 the velocity variance of the fluid motion by more than about 20%. Similarly,
 268 the estimate of ε captured by the particles from the slope of the inertial range
 269 is expected to be even more accurate, since there is a factor of ~ 5 between
 270 the integral length scale L_i and the scale of the particles d_p , which allows
 271 a sufficient window of motions in the inertial range to be well resolved by
 272 the particles. However, the inertial range detected in this way is necessarily

273 relatively narrow, as will be confirmed later. These arguments allow us to
 274 extend the domain of validity of the conclusions drawn by Bourgoïn et al.
 275 (2011) and Qureshi et al. (2007) for $d_p/\eta \sim 30$ to $d_p/\eta \sim \mathcal{O}(100)$.

276 The experiments were characterized by a set of dimensionless numbers
 277 and characteristic length and time scales, which are given in Table 2. These
 278 dimensionless numbers and scales depend on the energy dissipation rate ε ,
 279 whose estimate will be thoroughly detailed in section 5.

Experiment Id.	U' cm s ⁻¹	ε cm ² s ⁻³	L_i cm	τ_e s	Re	Re_λ	λ mm	η μ m	τ_η ms
4	7.0	31.0 ± 2.5	11.0	1.6	7682	339	4.9	133.8	18.0
5	5.6	17.1 ± 1.6	10.2	1.8	5693	292	5.2	155.3	24.2
6	5.7	15.4 ± 1.4	12.1	2.1	6942	323	5.5	159.8	25.5
7	8.6	55.9 ± 4.3	11.3	1.3	9733	382	4.4	115.7	13.4
8	10.3	108.3 ± 7.6	10.2	1.0	10586	398	3.9	96.9	9.6
9	5.8	16.5 ± 1.5	11.5	2.0	6636	316	5.5	157.0	24.6
10	5.2	11.8 ± 1.3	11.9	2.3	6206	306	5.9	170.9	29.1
11	9.6	73.4 ± 5.9	12.1	1.3	11649	418	4.3	108.1	11.7

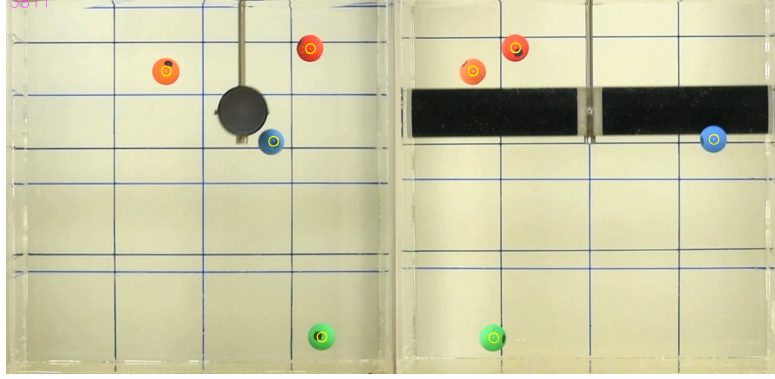
Table 2: Experimental scales and dimensionless numbers. $U' = \sqrt{(\langle U_x \rangle^2 + \langle U_y \rangle^2 + \langle U_z \rangle^2)/3}$ is the particle velocity averaged over components. The energy dissipation ε represents an average of the estimates from the Lagrangian velocity structure function, the energy spectrum and the longitudinal structure function. The integral length scale is given by $L_i = U'^3/\varepsilon$ and the eddy turn-over time by $\tau_e = L_i/U'$. The Reynolds number is estimated as $Re = L_i U'/\nu$ and the Reynolds number based on the Taylor micro-scale λ is estimated as $R_\lambda = \lambda U'/\nu$ with $\lambda = \sqrt{15 U'^2 \nu / \varepsilon}$. The Kolmogorov length and time scales are respectively $\eta = (\nu^3/\varepsilon)^{1/4}$ and $\tau_\eta = (\nu/\varepsilon)^{1/2}$.

280 3. Analysis methodology

281 3.1. Particle tracking

282 The experiments were recorded during 300 s from two sides by cameras
283 in video mode providing one plane view across the cylinder axis, and another
284 along the cylinder axis. As we varied the period of the cylinder motion from
285 3 s to 1.75 s, the recording covered a minimum of 100 cycles (18,000 frames)
286 to a maximum of 171 cycles, implying that the number of samples slightly
287 differs across the 11 experiments when calculating statistical quantities. The
288 videos were produced at a resolution of 1920×1080 pixels and at a number of
289 frames per second $n_f=59.94$ frames/s. Note that at this sampling rate we did
290 not expect to resolve the dissipative turbulence range as $T/n_f \sim \mathcal{O}(10)\tau_\eta$,
291 where τ_η is the Kolmogorov time scale (see Table 2). The two recordings were
292 first synchronized using the frame at which the cylinder started to move. A
293 camera calibration was performed using the landmarks of grids that had been
294 drawn on the sides of the tank to measure the 3D coordinates of the particles.
295 Particles and cylinder were tracked based on their coloured pixels, and we
296 followed the centre of the finite-size spheres as shown in Figure 2a. The set
297 of centre particle positions over time $(X(t), Y(t), Z(t))$ defined the particle
298 trajectory as shown Figure 2b. We did not track the particle orientations.
299 To compute the velocity of the spheres, a monotonic cubic spline was fitted
300 to the (X, Y, Z) particle positions for the purpose of applying a first-order
301 differentiation.

a)



b)

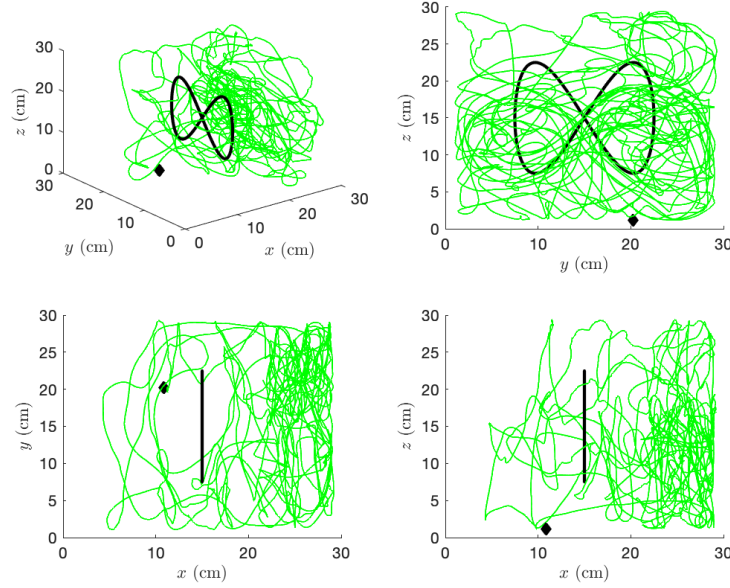


Figure 2: a) Three-dimensional tracking of the particles from the two side views taken by the cameras. The background lines are the grids used for 3D geo-referencing and camera calibration. The yellow circles mark the centre of the finite-size particles identified by particle tracking. b) Reconstructed trajectory. The example shows that of the green particle in experiment 5 seen in different views, including looking parallel and perpendicular to the axis of the cylinder and from the top. The black line represents the path of the cylinder in each plane (y, z) , (x, y) and (x, z) . For visibility reasons, the cylinder position along the x direction is simply shown at the centre $x = 15$ cm. The black diamond indicates the initial position of the particle.

3.2. Statistical analysis

Statistical analysis was used to interpret the data. We first checked the equivalence of the 4 particles, and assessed the role of the collisions and transient behaviour of the particles. Appendix A and Appendix B give full details on the existence of a transient period and its duration, and on the lack of impact of the collisions on the particle velocity statistics. As a result, in data sets containing positions and velocities, the data corresponding to the transient were removed; velocities of the resulting data sets were not filtered for the collisions; and data sets of the four particles were assimilated into a single set for analysis, such as for estimating Probability Density Functions (PDF). Besides, spatial statistical analyses were performed after subdividing the entire (undisturbed) volume of fluid $V = H \times D \times W = (30 \text{ cm})^3$ into 15^3 cells of dimensions $V_c = (2 \text{ cm})^3$, i.e. the cubic volume occupied by a particle. In a cell (i, j, k) of central position (x_i, y_j, z_k) , we evaluated the count of particles $N(i, j, k)$, and the velocity $\mathbf{U}(i, j, k)$. We note that all the cells close to a boundary will be statistically different from interior cells because the finite-size of the particles implies that their centres are at a distance of at least 1 cm from the lateral walls or bottom of the tank. In other words, compared to an interior cell, only half a cell effectively contributes to the statistics when it is bounded by a tank wall, because in those cells the centre of a particle is constrained to take a position in only half of its volume (farther from the boundary).

324 4. Dynamics of the particles

325 4.1. Ensemble particle localization

326 An insight into the ensemble wandering of particles in the tank was first
 327 gained by analyzing the percentage of fluid volume that was never visited
 328 by the four particles over the duration of each experiment excluding the
 329 transient. Wandering of particles increases as the stirring rate increases, as
 330 shown in Figure 3. An exponential fit to the data further indicates that
 331 this study uses a range of stirring rates that achieves reasonable excursion of
 332 the particles. Increasing further the stirring rate would have increased the
 333 particle wandering but it was technically not possible due to the torque limit
 334 of the actuators.

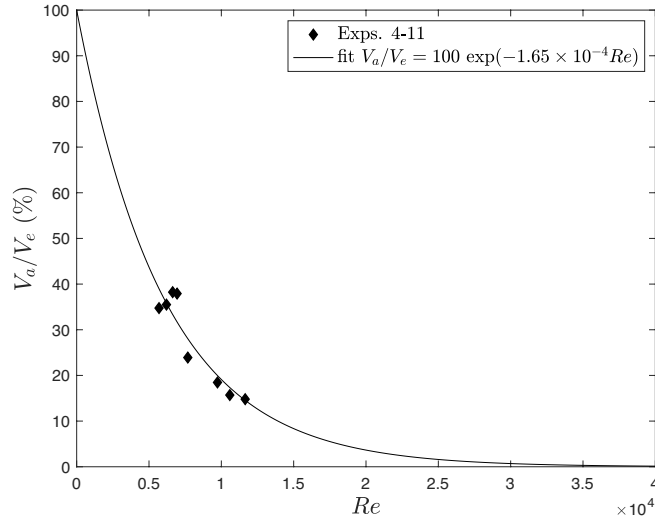


Figure 3: Dimensionless volume V_a/V_e (in %) never visited by any of the four particles as a function of Reynolds number Re for experiments 4–11. Note that V_a has been normalized here using the volume accessible to the particles $V_e \sim V(1 - d_p/W)^2$.

335 The increase in wandering with the stirring rate especially applies to the
 336 particle excursion in the z (vertical) direction. As shown in Figure 4, at low
 337 stirring rate, the PDFs of the Z particle coordinate are higher in the lower
 338 half of the tank regardless of the direction of stirring. At high stirring rate,
 339 however, there is much less vertical disparity between the two directions of
 340 cylinder motion. The increase in stirring velocity helps to counteract the
 341 slight negative buoyancy of the particles, whose presence in the upper half
 342 of the tank is facilitated by the more vigorous vortices.

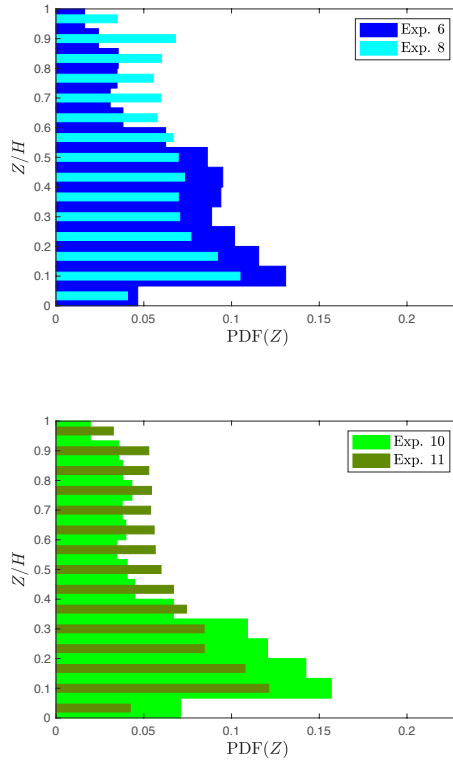


Figure 4: Probability Density Functions (PDFs) of particle dimensionless vertical coordinate Z/H in experiments 6 & 8, and 10 & 11. The stirring is weaker in experiments 6 & 10 than in experiments 8 & 11.

4.2. Inhomogeneous and anisotropic flow inferred from the particles

While the stirring is obviously anisotropic, the flow inferred the particle motion is also influenced by the anisotropy of the forcing. This is shown in Figure 5 by the distributions of the direction cosines of the particle velocity vectors calculated as $\text{cosine}(\alpha_i^j) = U_i^j / |\mathbf{U}^j|$, where i refers to components x , y and z and j runs from 1 to four times the number of frames recorded between the end of the transient and the end of the experiment. Regardless of the direction and intensity of stirring, the direction cosines are more uniformly distributed in the y and z directions, whereas in the x direction the distribution is non-uniform and peaks around zero, indicating a preferred direction of the velocity vectors normal to the x axis. However, the histograms in Figure 5 also show that the motion is far from being perfectly two-dimensional (which would correspond to a Dirac function $\delta(0)$ for $\text{cosine}(\alpha_x)$).

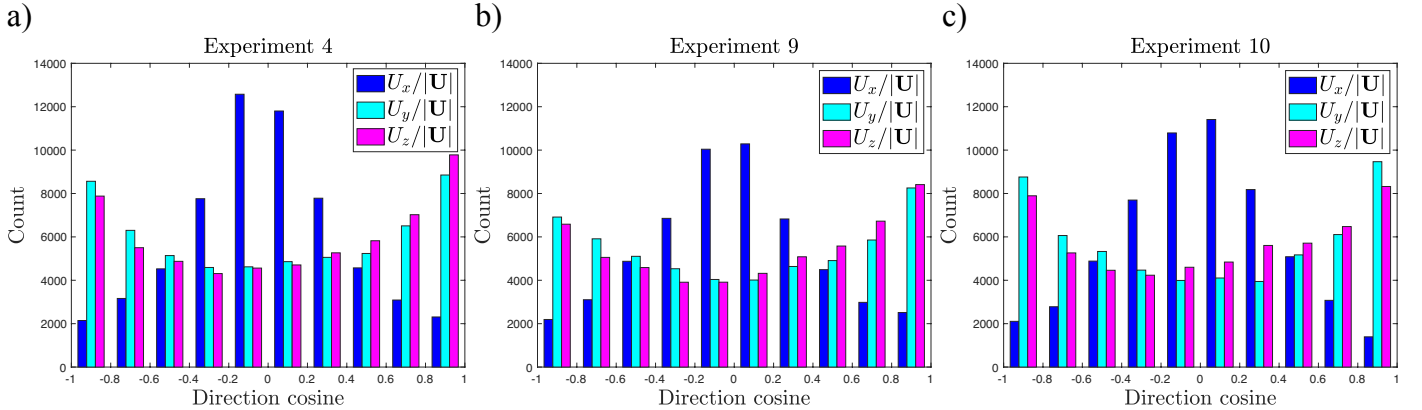


Figure 5: Raw histograms of the direction cosines of the particle velocity for experiments a) 4, b) 9 and c) 10. Experiments 4, 9 and 10 exemplify experiments at different directions and/or intensity of stirring.

Further insights into the inhomogeneity and anisotropy of the motion can be found by looking at the velocity fields at the particle scale, as shown in Figures 6 and 7 for two representative planes (y, z) and (x, y) and, for the two directions of stirring.

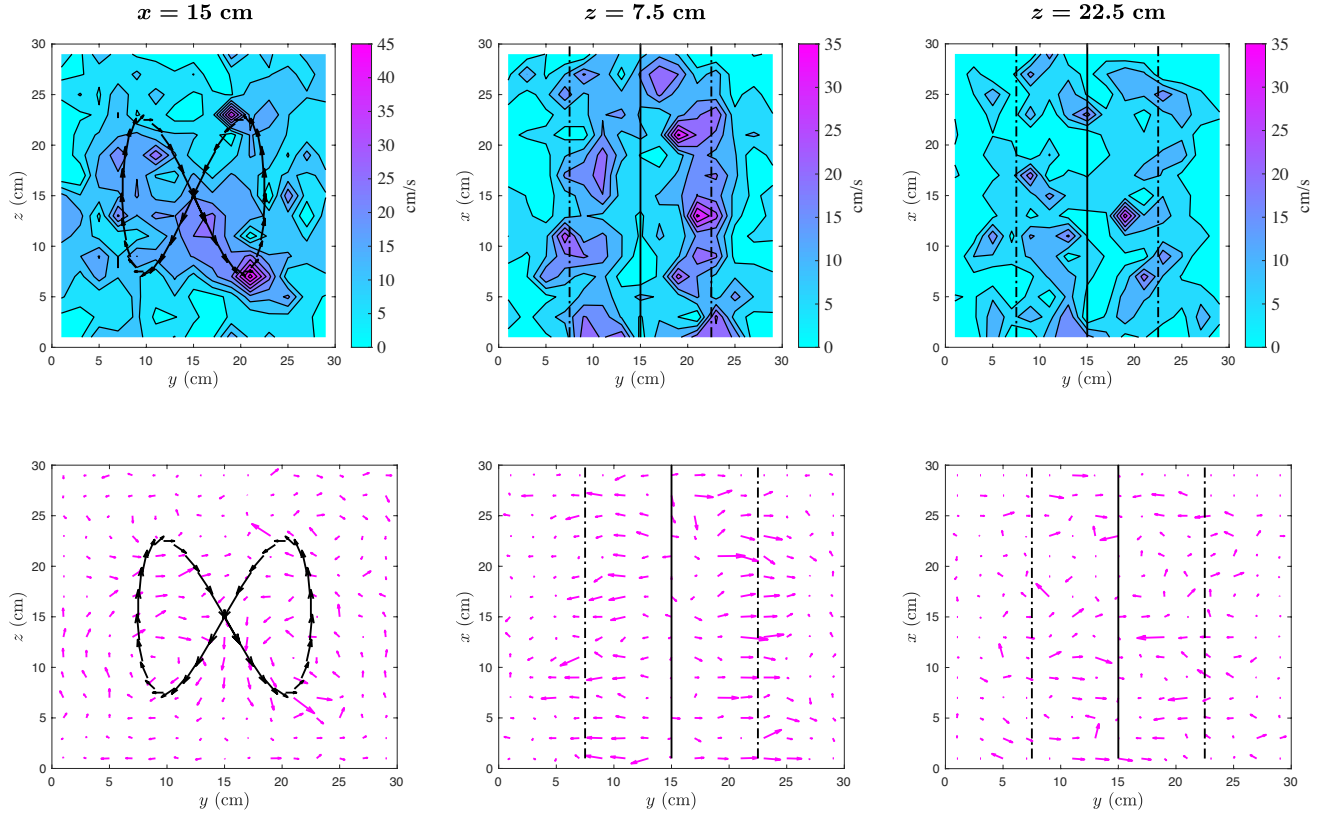


Figure 6: Contour maps of the flow speed (top frames) and corresponding velocity vector field (lower frames) in experiment 8. The black lines delimit the path or region reached by the cylinder. The colour scale indicates the magnitude of the flow speed. The size of the velocity vectors in the flow fields has been scaled for visualisation purposes.

Aside from preferential directions normal to the x axis, consistent the anisotropy discussed in Figure 5, the velocity fields in the interior of the

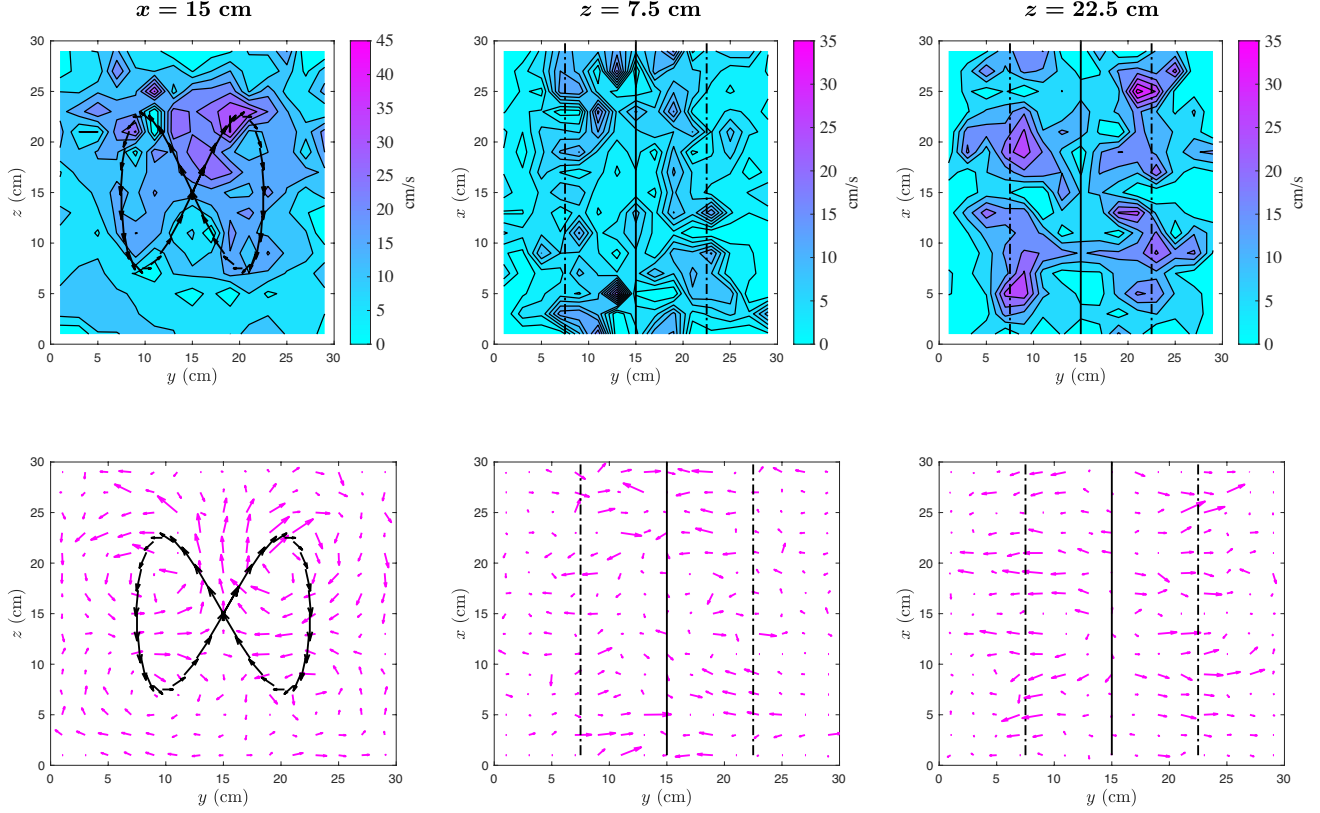


Figure 7: Contour maps of the flow speed (top frames) and corresponding velocity vector field (lower frames) in experiment 11. The black lines delimit the path or region reached by the cylinder. The colour scale indicates the magnitude of the flow speed. The size of the velocity vectors in the flow fields has been scaled for visualisation purposes.

362 measurement volume are quite inhomogeneous. They comprise areas of high
 363 velocity being essentially located along the cylinder path, which contrasts
 364 with areas of low velocity, especially close to boundaries (i.e. walls, and
 365 the free surface). However, differences in inhomogeneity exist between the
 366 two directions of stirring, especially in the vertical direction. In the case of
 367 stirring with initial downward cylinder motion, large-scale vortices are most

active in the lower half of the tank, whereas for stirring with initial upward cylinder motion, large-scale vortices are most active in the upper half of the tank, as shown by Figures 6 and 7. Interestingly, for stirring with initial upward cylinder motion, large-scale vortices also exist in the lower half of the tank, although they have a weaker intensity, but there are almost no vortices in the upper half of the tank in the case of stirring with initial downward cylinder motion.

4.3. *Individual trajectories*

While the global flow features seem to point to a reasonably efficient wandering of the particles in the tank, although constrained by the looping motion of the cylinder in the (y, z) plane perpendicular to the cylinder axis, more complex behaviour emerges when looking at the particles individually. In particular, substantial variability between different particles is found regarding their excursions in the x (i.e. along-axis) direction, in which the cylinder does not generate any direct forcing motion. This effect seems to prevent individual particles from crossing the whole domain along x , leading to particles being strikingly confined, as shown in Figure 2b. The observed particle confinement in the x direction is consistent with the anisotropic flow dominated by motion in the (y, z) plane, as shown in Figure 5, and is a manifestation of the conservation of the angular momentum perpendicular to that plane. An uneven localization along the x direction is observed regardless of the stirring rate and direction, as shown by the (y, z) plane-averaged residence times as a function of x in Figure 8. We will see that this contributes to a differentiation of the particle pairs during dispersion.

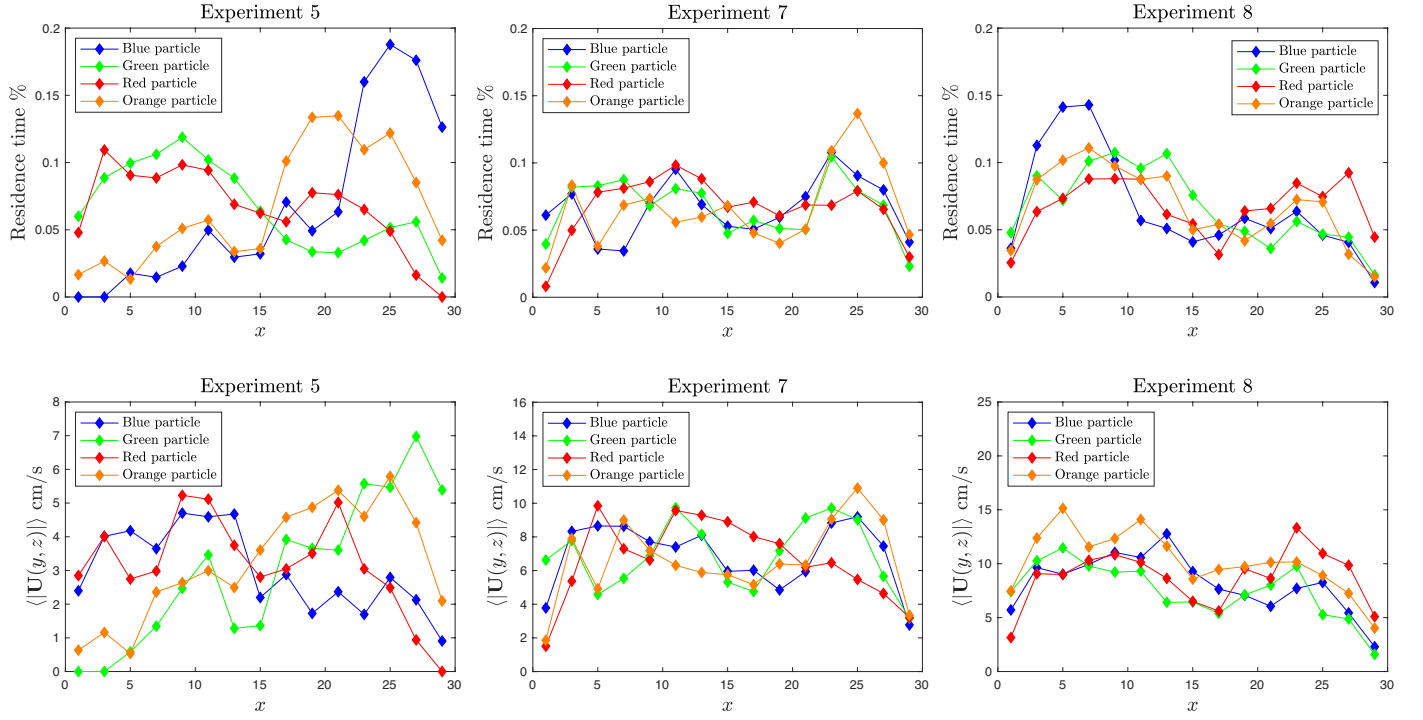


Figure 8: Top frames: $y - z$ plane-averaged residence time of the particles along the x direction, respectively, for experiments 5, 7 and 8. Bottom frames: corresponding $y - z$ plane-averaged velocity magnitude along the x direction in the same experiments. Note that the particle residence time anti-correlates well with the particle plane-averaged velocity: where the averaged velocities appear larger, the residence time is lower, and vice-versa. This is to be expected, since the residence time should scale inversely to the flow velocity.

392 5. Statistical properties

393 As was mentioned in the Introduction, the time t_0 delimiting the ballistic
 394 and super-diffusive regimes in 3D dispersion is defined in terms of the energy
 395 dissipation rate, ε . This requires obtaining an estimate of this quantity. To do
 396 so, we used both the Lagrangian and Eulerian frameworks. The Lagrangian
 397 velocity structure function D and frequency spectrum E are strictly tensors
 398 of order 2 because the anisotropy of the large-scale flow is also present in
 399 the smaller-scale fluctuations of the particles. These tensors are respectively
 400 defined as:

$$401 \quad D_{ij}(\tau) = \langle \delta U_i(\tau) \delta U_j(\tau) \rangle, \quad (6)$$

402 where $\delta U_i(\tau) = U_i(t + \tau) - U_i(t)$ (Monin and Yaglom, 2013; Mordant et al.,
 403 2003) and

$$404 \quad E_{ij}(\omega_k) = \frac{\delta t}{2\pi} \left[2 \left(\sum_{j=1}^{n-1} R_{ij}(j\delta t) \cos(\omega_k t_j) \right) + R_{ij}(0) + R_{ij}(n\delta t) \right], \quad (7)$$

405 taken at equal sampling intervals of size δt with $\omega_k = k\pi/n\delta t$ ($k = 0, \pm 1, \dots, \pm$
 406 n) and $R_{ij}(\tau) = \langle U_i(t) U_j(t + \tau) \rangle$ (Yeung and Pope, 1988). Here we only
 407 evaluated the D_{ii} and E_{ii} components of these tensors together with an es-
 408 timate of the trace of D defined as $\text{Tr}(D) = 1/3 \sum_i D_{ii}$. Alternatively, in
 409 the Eulerian framework, we estimated the dissipation rate from the second-
 410 order longitudinal structure function $C_2(l)$ assuming that the instantaneous
 411 velocity at a similar time t of two particles respectively with positions \mathbf{x} and
 412 $\mathbf{x} + \mathbf{l}$ coincides with the local Eulerian velocity field. $C_2(l)$ is thus defined by

$$413 \quad C_2(l) = \langle ([\mathbf{U}(\mathbf{x} + \mathbf{l}, t) - \mathbf{U}(\mathbf{x}, t)] \cdot \mathbf{l}/l)^2 \rangle, \quad (8)$$

414 where $\mathbf{U}(\mathbf{x}, t)$ and $\mathbf{U}(\mathbf{x} + \mathbf{l}, t)$ are the velocities of a pair of particles at time t
 415 and positions \mathbf{x} and $\mathbf{x} + \mathbf{l}$. Such an evaluation of $C_2(l)$ is similar to that used
 416 in Valizadeh and Monaghan (2012) with SPH tracer-like particles, but for
 417 particle separations $l > d_p$, where the particle velocities can be representative
 418 of the flow (Qureshi et al., 2007; Bourgoin et al., 2011). $C_2(l)$ was further
 419 averaged over the six pairs of particles.

420 Figure 9 shows the compensated Lagrangian velocity structure function
 421 $D_{ii}(\tau)/(C_0\tau)$, frequency spectrum $\pi E_{ii}(\omega)\omega^2/C_0$ and the second-order longi-
 422 tudinal structure function $(C_2(l)l^{-2/3}/C_K)^{3/2}$. All three statistical properties
 423 were compensated with the dimensional expression given by the classical
 424 Kolmogorov theory in the inertial range (see section 1). We used the scaling
 425 constants $C_0 = 5$ and $C_K = 2.01$, which have been associated with three-
 426 dimensional turbulence, including in anisotropic contexts (Ouellette et al.,
 427 2006). All three statistical quantities consistently show a plateau even if
 428 the plateau is better developed for the energy spectrum and the longitudi-
 429 nal structure function than for the Lagrangian velocity structure function.
 430 Values of the compensated functions at their plateaus were used to esti-
 431 mate ε for each experiment. In practice, the value of ε inferred from the
 432 compensated Lagrangian velocity structure function $D_{ii}(\tau)/(C_0\tau)$ was sim-
 433 ply taken as being an average of the maximum compensated values for the
 434 three velocity components at $\tau = 0.2 - 0.3$ as the structure function did not
 435 have a well-defined plateau. For the frequency spectrum $\pi E_{ii}(\omega)\omega^2/C_0$ and
 436 the second-order longitudinal structure function $(C_2(l)l^{-2/3}/C_K)^{3/2}$, which
 437 had better-defined plateaus, values of ε were calculated as an average of the
 438 means of the compensated values over the frequency range $w = [4, 10] \text{ s}^{-1}$

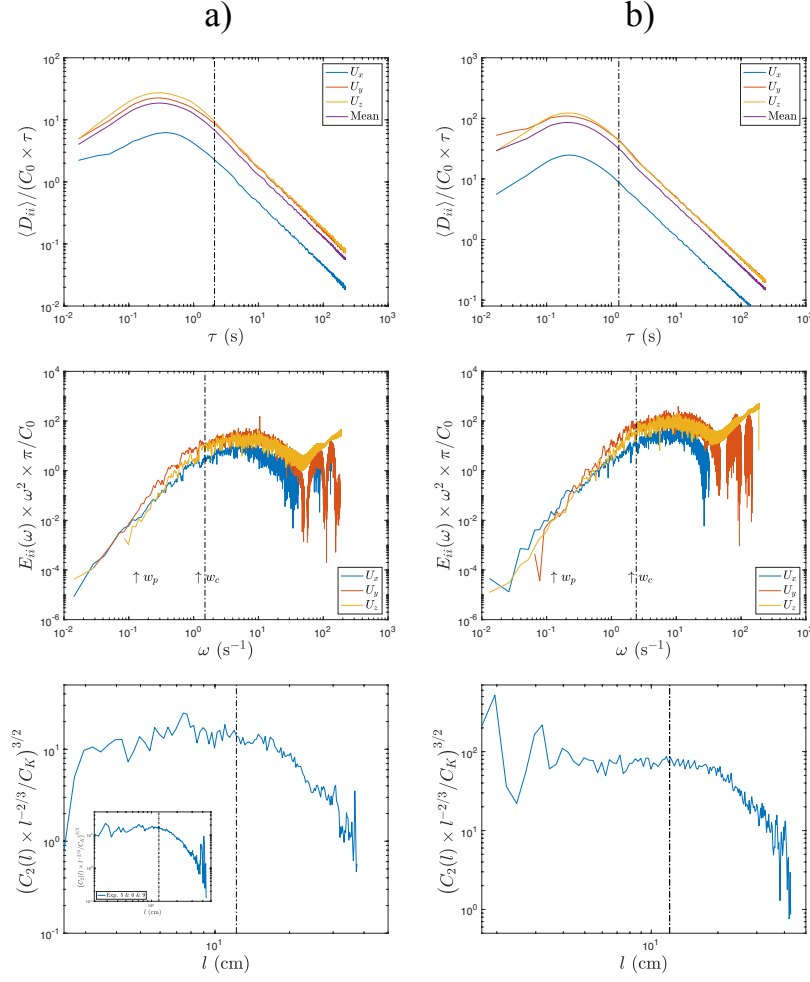


Figure 9: Compensated Lagrangian velocity structure function, frequency spectrum and second-order longitudinal structure function in a) experiment 6 and b) experiment 11. The values $w_c = \pi/T$ and $w_p = \pi/t_p$, where t_p is the particle relaxation time, are the frequency of the particles and cylinder, respectively. The inset in the figure for the longitudinal structure function $(C_2(l)l^{-2/3}/C_K)^{3/2}$ of experiment 6 is the same structure function but evaluated using the three experiments 5, 6 & 9, giving a smoother compensated function as the statistical sampling is increased. The vertical dashed lines indicate the limits $\tau = \tau_e$, $\omega = \pi/\tau_e$ and $l = L_i$. In experiment 6, ε derived from the three functions gives a mean value with standard error, $\varepsilon = (15.39 \pm 1.40) \text{ cm}^2\text{s}^{-3}$. In experiment 11, ε derived from the three functions gives a mean with standard error, $\varepsilon = (73.38 \pm 5.85) \text{ cm}^2\text{s}^{-3}$.

439 and length range $l = [5, 10]$ cm, respectively, for the three velocity compo-
 440 nents. We note that these averages for the Lagrangian velocity structure
 441 function, $1/3 \sum_i D_{ii}(\tau)$, and the frequency spectrum, $1/3 \sum_i E_{ii}(\tau)$, repre-
 442 sent the isotropic decomposition of D and E . Finally, as the standard errors
 443 of the mean of values of ε inferred from D , E and C_2 ranged only between 4
 444 and 6%, we obtained a final estimate of ε as the average of these three esti-
 445 mates. The resulting value of ε was then used to calculate the associated flow
 446 scales and dimensionless numbers (see Table 2). The time τ corresponding
 447 to the maximum of the compensated Lagrangian velocity structure function
 448 $D(\tau)$ is reasonably consistent with the values of the eddy turn-over time τ_e
 449 given in Table 2. Similarly, the lower limit ω of the plateaus of the frequency
 450 spectrum $E(\omega)$ reasonably agrees with the frequency $\omega_e = \pi/\tau_e$. In the spa-
 451 tial domain, the region of the function $C_2(l)$ between the lags $l = 4$ cm and
 452 $l = L_i = 10 - 12$ cm in Figure 9 also matches with what would be expected
 453 for an inertial range. The quoted lower limits of l are dictated by spatial
 454 resolution, and the upper limits coincide with the forcing length scales of
 455 $L_i \sim 12$ and 10 cm in experiments 6 and 8, respectively. We note that val-
 456 ues of L_i are naturally close to the size of the Lissajoux curve executed by
 457 the cylinder while stirring, that is $L_c = 2A = 15$ cm.

458 **6. Particle dispersion**

459 In the context of the inhomogeneous and anisotropic turbulence just de-
 460 scribed, we now examine both single-particle dispersion and particle-pair
 461 dispersion, also known as relative dispersion. Although the dispersion of
 462 particles must be three-dimensional, the flow has been shown to be strongly

anisotropic (and increasingly so as the stirring rate increases). So, the aim here is to check if large-sized particle dispersion in strongly inhomogeneous and anisotropic turbulence satisfies the same scaling laws as tracers in homogeneous and isotropic 3D turbulence.

6.1. Single-particle dispersion

Single-particle dispersion can be investigated by analysing the trajectory of a single particle, by calculating $\langle |\Delta(\tau)|^2 \rangle$, where $\Delta(\tau) = \mathbf{X}(t + \tau) - \mathbf{X}(t)$, where $\mathbf{X}(t)$ is the position of a particle at each time t along its trajectory and τ is the time lag. Figure 10a shows $\langle |\Delta(\tau)|^2 \rangle / Li^2$ as a function of τ/τ_e for each of the four particles in experiment 7. When $\tau/\tau_e \lesssim 0.25$, the ballistic dispersion regime holds, i.e. $\langle |\Delta(\tau)|^2 \rangle \propto \tau^2$, whereas when $\tau/\tau_e \gtrsim 0.6 - 0.7$, the mean-square displacement follows a diffusive regime ($\langle |\Delta(\tau)|^2 \rangle \propto \tau$) over a brief time interval of length approximately equal to τ_e , as shown in Figure 10c. The start of the diffusive regime coincides with the time required for the decay of the Lagrangian velocity auto-correlation of the particles $\langle U_i(t)U_i(t + \tau) \rangle / \langle U_i(t)^2 \rangle$, that is $T_L/\tau_e = 0.6$, as shown in Figure 10b. At $\tau/\tau_e \approx 2.5$, the mean-square displacement reaches a plateau. The brevity of the diffusive regime is due to the finite dimensions of the domain, which limit the particle's excursion at large times. Similar dimensionless curves of $\langle |\Delta(\tau)|^2 \rangle / Li^2$ and $\langle U_i(t)U_i(t + \tau) \rangle / \langle U_i(t)^2 \rangle$ as a function of τ/τ_e were displayed for all other experiments (not shown).

The dispersion regimes of Figure 10 may be also interpreted in terms of space instead of time. Thus $\tau/\tau_e \sim 0.25$ corresponds to the mean-square particle displacement $\langle |\Delta(\tau)|^2 \rangle / L_i^2 \sim 0.16$, which corresponds to a root-mean-square displacement $L_b/L_i = (\langle |\Delta(\tau)|^2 \rangle)^{1/2} / L_i \sim 0.4$ or in dimensional terms

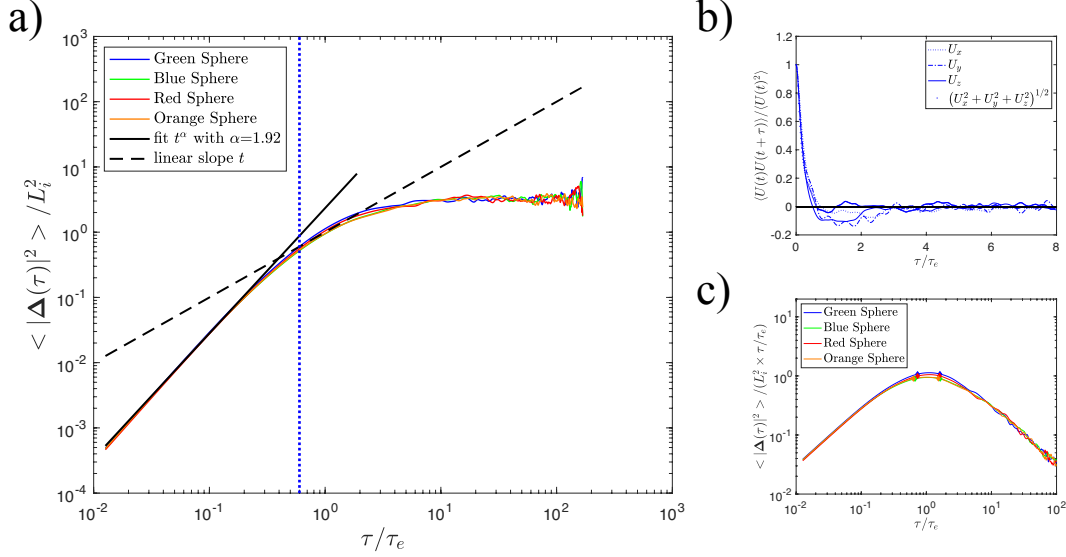


Figure 10: a) Mean square displacement relative to initial position as a function of time τ/τ_e along the trajectories of the 4 particles in experiment 7. The four particles have a similar behaviour. The black solid line represents a linear fit of the data (in logarithmic scales) up to $\tau/\tau_e = 0.25$, which gives a slope of 1.92, very close to the predicted slope of 2. The black dashed line represents a slope of 1, indicating a diffusive regime. b) Lagrangian velocity auto-correlation function. The Lagrangian velocity auto-correlation time $T_L/\tau_e \sim 0.6$ is shown by the vertical blue dotted line in a). c) Single-dispersion compensated for the diffusive regime. The diamonds delimit the intervals over which $\langle |\Delta(\tau)|^2 \rangle / (L_i^2 \times \tau/\tau_e)$ is equal to its maximum within a tolerance of 5%. This criterion is used to ascertain the presence of plateaus and hence the diffusive regime. The average width of such intervals for the four particles is $\Delta\tau/\tau_e = 0.9$. Here the diffusive regime is observed between $T_L/\tau_e \lesssim \tau/\tau_e \lesssim 2.5T_L/\tau_e$.

488 in experiment 7, $L_b = (\langle |\Delta(\tau)|^2 \rangle)^{1/2} \sim 4.5$ cm. Hence, in this latter experi-
 489 ment, the ballistic regime remains valid for particle displacements below \sim
 490 5 cm. On the other hand, $\tau/\tau_e = 0.6$ corresponds to $\langle |\Delta(\tau)|^2 \rangle / L_i^2 \sim 0.63$,
 491 or $L_d/L_i = (\langle |\Delta(\tau)|^2 \rangle)^{1/2} / L_i \sim 0.8$, or equivalently, in experiment 7, $L_d =$

492 $(\langle |\Delta(\tau)|^2 \rangle)^{1/2} \sim 9$ cm, which means that the diffusive regime will apply to
 493 particle displacements larger than 9 cm. Finally, the plateau reached by the
 494 dispersion curves ends when $\langle |\Delta(\tau)|^2 \rangle / L_i^2 \sim 1.8$, or $(\langle |\Delta(\tau)|^2 \rangle)^{1/2} / L_i \sim 1.34$,
 495 or equivalently, in experiment 7, $(\langle |\Delta(\tau)|^2 \rangle)^{1/2} \sim 15$ cm, which means that
 496 once the particles approach displacements around 15 cm the displacement is
 497 unable to on average increase further due to the limited dimensions of the
 498 domain.

499 Aside from showing the existence of the ballistic and diffusive regimes, the
 500 results of this analysis of single-particle dispersion are the above definition
 501 of T_L and the characteristic dimensionless lengths L_b and L_d delimiting the
 502 different dispersion regimes, which will be used in the next section in the
 503 interpretation of particle-pair dispersion.

504 6.2. Particle-pair dispersion

505 To analyse particle-pair dispersion, we used both the traditional way of
 506 looking at the relative dispersion as a function of time and the fixed length-
 507 scale method (FSLE). We will show in this section that the two methods
 508 are complementary. We first looked at the time evolution of the separation
 509 between particles i and j , $|\mathbf{S}_{ij}(t)| = |\mathbf{X}_i(t) - \mathbf{X}_j(t)|$, by calculating the mean-
 510 square relative distance $\langle (|\mathbf{S}_{ij}| - S_0)^2 \rangle$ of pair ij relative to the initial pair
 511 separation $|\mathbf{S}_{ij}(0)| = S_0$. The values of S_0 were carefully chosen so that they
 512 span characteristic lengths of the system within the interval $S_0 = [3, 11]$ cm.
 513 This allowed a calculation of statistically representative ensemble averages of
 514 the mean-square relative distance. However, the sampling was uneven: the
 515 number of ensemble members was found to roughly linearly increase with
 516 intermediate values of S_0 , be sometimes small at the lowest (< 3 cm) and

517 largest (> 30 cm) values of S_0 , and vary between pairs for the same S_0 , es-
 518 pecially for experiments with low stirring rates. For instance, in experiment
 519 5, the number of ensemble members averaged over the six pairs increased
 520 from 18 to 151 as S_0 increased from 3 to 11 cm, and varied between pairs at
 521 $S_0 \sim 3$ cm from zero (pair 4) to 62 (pair 5), whereas in experiment 11, the
 522 number of ensemble members averaged over the six pairs increased from 17
 523 to 172 for a similar set of values of S_0 , and varied between pairs at $S_0 \sim 3$ cm
 524 from 8 (pair 5) to 28 (pair 6). The fact that sampling varies between particle
 525 pairs likely results from the fact that the turbulence is neither isotropic nor
 526 homogeneous.

527 In all experiments, as exemplified in Figure 11 for experiments 5 and
 528 11, the mean-square relative distance $\langle (|\mathbf{S}| - S_0)^2 \rangle$ shows three main trends:
 529 in the interval $\tau/\tau_e < 0.25$, it evolves as t^2 , clearly following a ballistic
 530 regime; when $0.25 < \tau/\tau_e < 0.6$, it varies as t^β with variable β values,
 531 $1.0 < \beta < 2.3$, so that there is no indication of a super-diffusive regime;
 532 when $\tau/\tau_e \gg T_L/\tau_e = 0.6$, it finally grows more slowly (eventually becoming
 533 stationary) with short-period oscillations, reflecting the finite dimensions of
 534 the domain. The diffusive dispersion regime in the interval $\tau/\tau_e > 0.6$ is
 535 equivocal, partly for the same reasons related to particle confinement as in
 536 single-particle dispersion, but also partly because the statistics are noisier.

537 The lack of identification of an intermediate super-diffusive regime can
 538 be explained by the narrow time windows $[t_0, T_L]$ that are available for this
 539 regime to exist, as shown in Table 3 for experiments 5 and 11 and the initial
 540 separations S_0 considered in Figure 11. For the time $t_0 = (S_0^2/\varepsilon)^{1/3}$ marking
 541 the transition from the ballistic to the super-diffusive regime at a given ini-

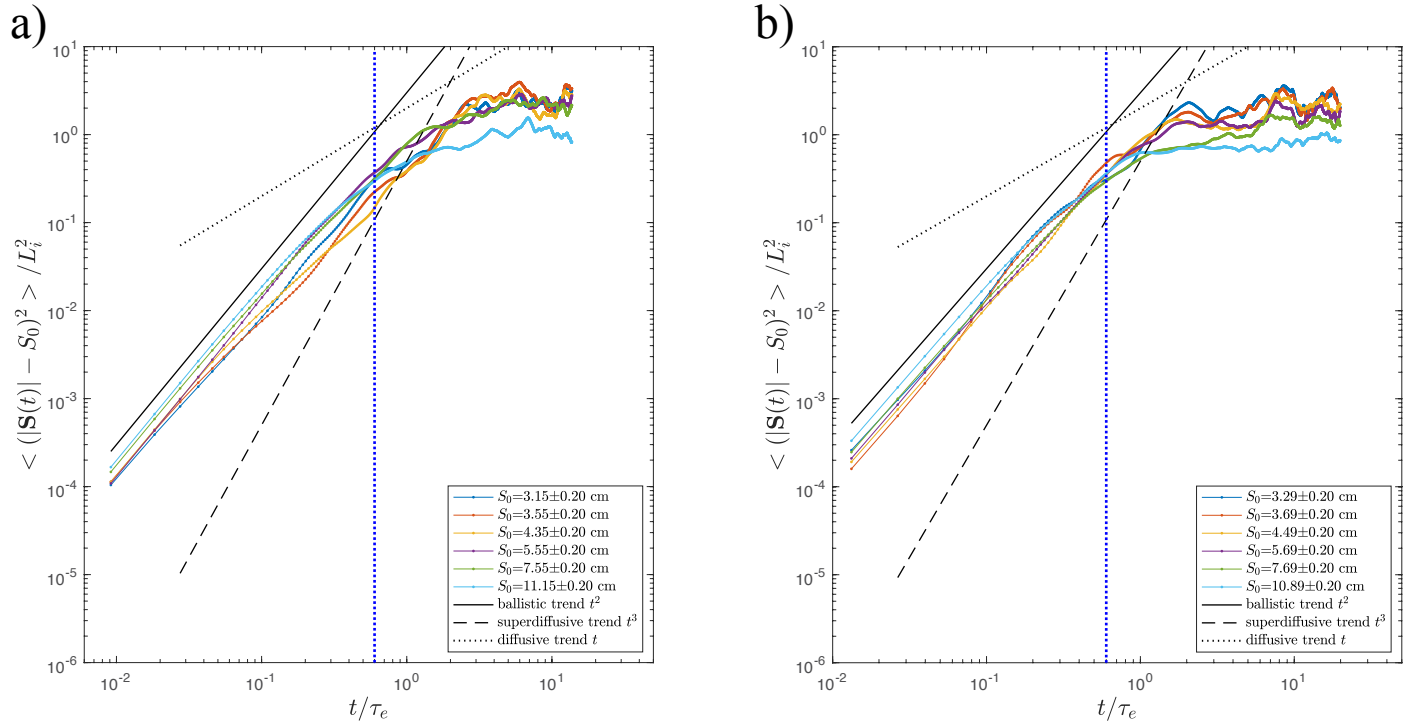


Figure 11: Mean square distance averaged over the six particle pairs for different initial separations S_0 as a function of time in a) experiment 5 and b) experiment 11. The black solid, dashed and dotted lines represent slopes of 2, 3, and 1, respectively, characterizing the ballistic, super-diffusive and diffusive regimes. The time $T_L/\tau_e = 0.6$ is shown by a vertical blue dotted line.

542 tial separation S_0 to be shorter and further separated from the Lagrangian
543 integral time T_L marking the start of the diffusive regime (see section 1),
544 an increase in ε and/or a smaller S_0 would be required. When $t_0 > T_L$,
545 the super-diffusive regime can not occur. The ratio of t_0 to T_L can also be
546 expressed as:

$$547 \quad \frac{t_0}{T_L} = \frac{(S_0^2/\varepsilon)^{1/3}}{0.6\tau_e} = \frac{(S_0^2/\varepsilon)^{1/3}}{0.6(L_i^2/\varepsilon)^{1/3}} = \frac{1}{0.6} \left(\frac{S_0}{L_i} \right)^{2/3}. \quad (9)$$

548 So, $t_0/T_L > 1$ is equivalent to $S_0/L_i > (0.6)^{3/2} = 0.47$ or $S_0 > 4.7 - 5.6$ cm
549 (see Table 2), which is a limit that is very close to that found for the end of
550 the ballistic regime in the single-particle dispersion, namely $L_b/L_i \sim 0.4$ or
551 $L_b \approx 4.5$ cm.

Exp. 5		Exp. 11	
S_0 (cm)	t_0/T_L	S_0 (cm)	t_0/T_L
3.15	0.76	3.29	0.70
3.55	0.83	3.69	0.75
4.35	0.95	4.49	0.86
5.55	1.11	5.60	1.00
7.55	1.36	7.69	1.23
11.15	1.77	10.89	1.55

Table 3: Estimates of t_0/T_L as a function of S_0 for experiments 5 and 11.

552 As an alternative to the analysis of fixed-time average of inter-particle dis-
553 tances over the ensemble of particle pairs, we computed the Finite-Scale Lya-
554 punov Exponent (FSLE). We thus calculated the function $\lambda(\delta) = \ln \rho / \langle \tau(\delta) \rangle$,

555 where δ is the spatial separation between two particle trajectories and $\langle\tau(\delta)\rangle$
 556 the mean time that δ takes to be amplified by a factor ρ . We took ρ as equal
 557 to $\sqrt{2}$ (Corrado et al., 2017). We ultimately averaged $\lambda(\delta)$ over the six par-
 558 ticle pairs. The results of the FSLE analysis are shown in Figure 12. In all
 559 experiments, two regimes ($\lambda(\delta) \propto \delta^{-\zeta}$) are consistently found: the ballistic
 560 separation ($\zeta \sim 1$) is present for $\delta < [0.77 - 0.99]L_i$; and the diffusive regime
 561 ($\zeta \sim 2$) for $[0.77 - 0.99]L_i < \delta < [1.2 - 1.7]L_i$. We note that the length
 562 scale interval for diffusion is consistent with that found in the single particle
 563 analysis of the dispersion regime. For instance, in experiment 7, the diffusion
 564 regime is found for separations between 9 cm and 16 cm, which agrees with
 565 the analysis of section 6.1. The fact that we find the transition to diffusive at
 566 $\delta \sim L_i$ shows that our estimates of the energy dissipation rate ε derived from
 567 the particles, and consequently of a number of scales derived from ε , such
 568 as L_i , are reliable. Finally, it is to be noted that, whereas the traditional
 569 approach shows the ballistic regime with much less noise, the FSLE analysis
 570 shows the diffusive regime much more clearly.

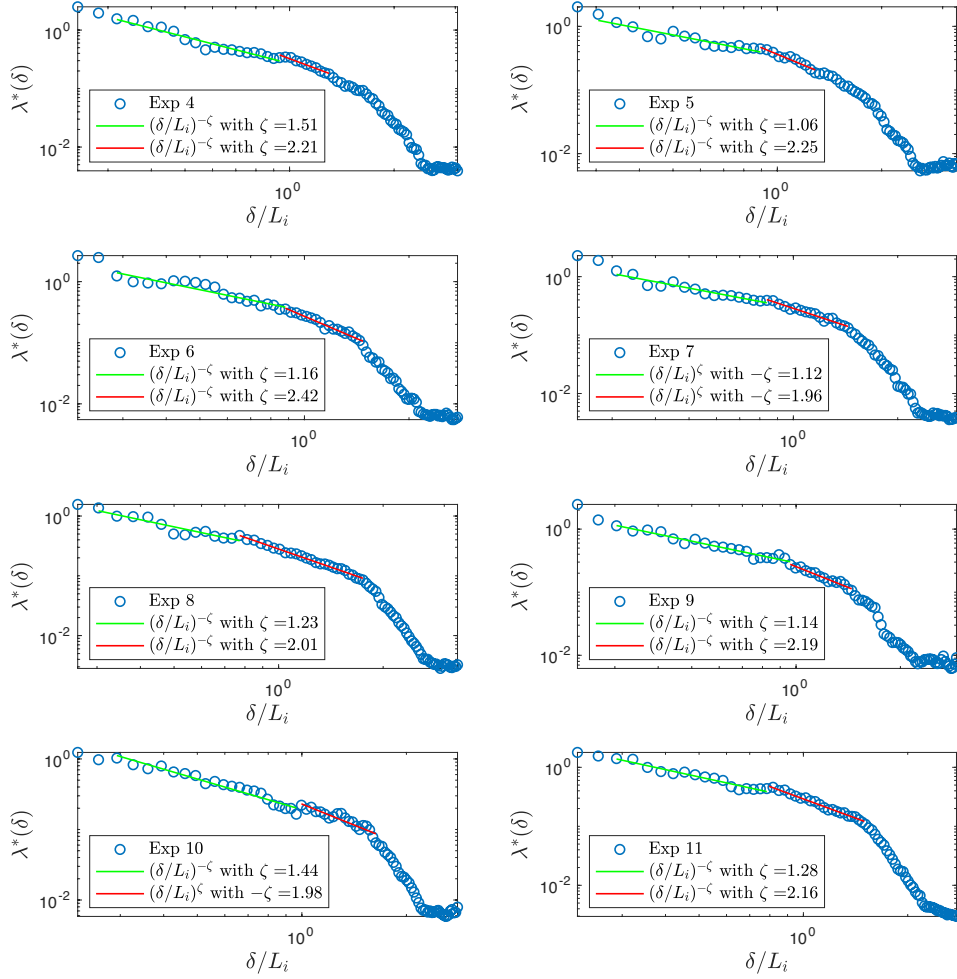


Figure 12: Lagrangian FSLE $\lambda(\delta) = \ln \rho / (\langle \tau(\delta) \rangle / \tau_e)$ as a function of δ/L_i for all experiments. The FSLE scaling exponent $\delta^{-\zeta}$ corresponds to: ballistic separation ($\zeta = 1$), and diffusive regime ($\zeta = 2$).

Whereas two dispersion regimes were identified when the statistics were
 averaged over the six particle pairs, the dispersion between single pairs had
 more variability, especially at low stirring rate, as shown in Figure 13. For
 instance, in experiment 5, pair 2 separated as $t^{2.9}$ in the interval $0.13 <$
 $\tau/\tau_e < 0.6$, whereas pair 6 separated as $t^{1.9}$ in the interval $0.01 < \tau/\tau_e < 0.6$.
 Similarly, in experiment 11, pair 3 separated as $t^{3.05}$ in the interval $0.2 <$
 $\tau/\tau_e < 0.6$, whereas pair 2 separated as $t^{1.9}$ in the interval $0.01 < \tau/\tau_e <$
 0.6 . Individually, particle pairs could thus seemingly exhibit a super-diffusive
 behaviour extended outside of the expected time window $t_0 < \tau < T_L$ (but
 overlapping with it). This variability affecting different dispersion pairs can
 be related to the variability of the Lagrangian correlation time of velocity
 differences (relative velocity between two particles of a pair) between the
 pairs separated by $S_0 \sim 3$ cm, as shown in Figure 14. For instance, in
 experiment 5, the velocity difference of pair 2 loses its memory of the initial
 separation at $\tau/\tau_e = 0.13$, five times more rapidly than for pair 6. This
 indicates not only that the ballistic regime ended earlier for pair 2 than
 for pair 6 but also that it ended earlier than the theoretical time $t_0/\tau_e =$
 $t_0/T_L \times T_L/\tau_e = 0.76 \times 0.6 = 0.46$. In practice, this corresponds to a better
 separation between t_0 and T_L , possibly allowing the Richardson regime to
 exist in this case. Similarly, in experiment 11, the velocity difference of pair 3
 decorrelated at $\tau/\tau_e = 0.19$ instead of $t_0/\tau_e = 0.42$ and earlier by a factor of 3
 than for pair 2. It is tempting to attribute the difference in the decorrelation
 time of the velocity difference between pairs to the inhomogeneity of the
 turbulence, although it is rather intricate to identify why it would affect the
 pairs differently. Nevertheless, for $S_0 \sim 3$ cm, histograms of the particle

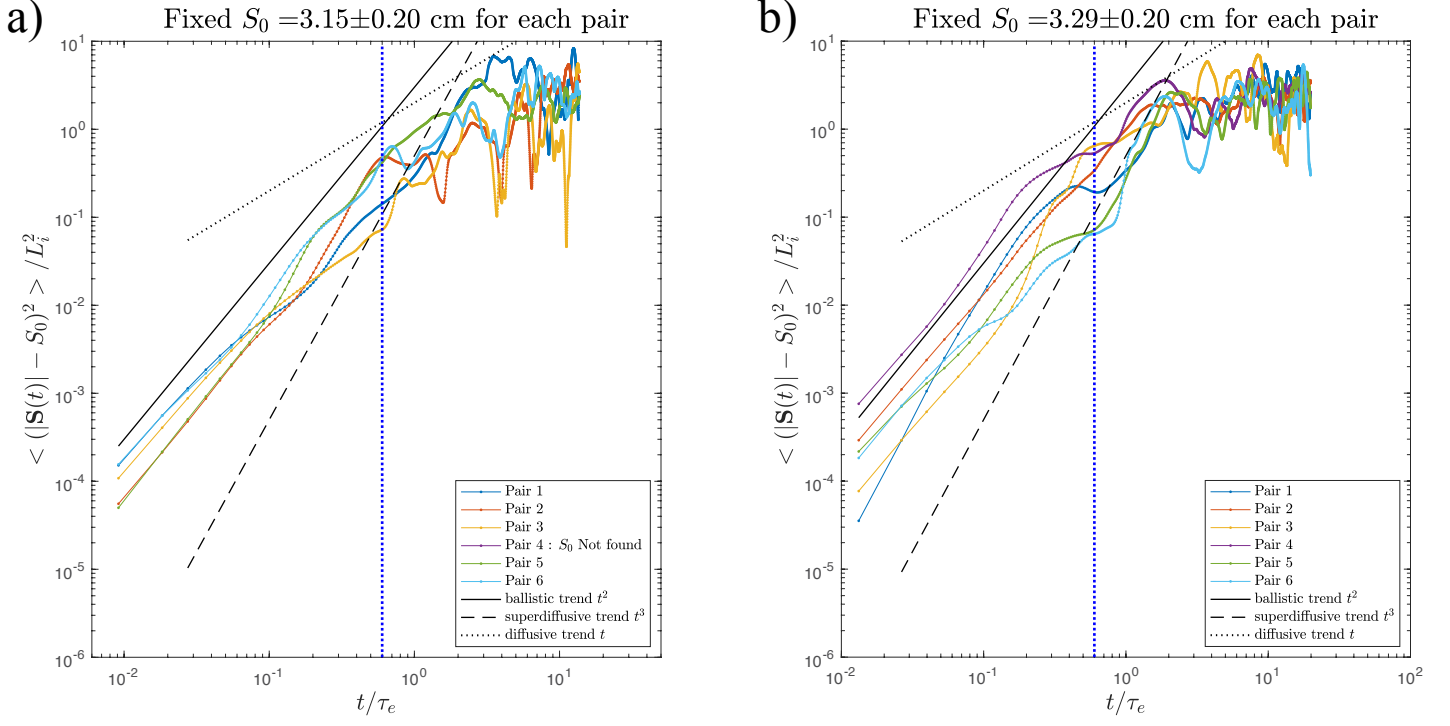


Figure 13: Mean square distance for each particle pair for $S_0 \sim 3$ cm as a function of time in experiments a) 5 and b) 11. The black solid, dashed and dotted lines represent slopes of 2, 3, and 1, respectively, characterizing the ballistic, super-diffusive and diffusive regimes. The time $T_L/\tau_e = 0.6$ is shown by a vertical blue dotted line. The separation of the particle pair 4 was never less than 5.5 cm in experiment 5, and so that pair is not shown in a).

positions for each characteristic pair 2 and 6, and 2 and 3, respectively, in experiments 5 and 11, shown in Figure 15, reveal that the particle pairs whose velocity differences were decorrelating slowly were actually close to the bottom wall or the free surface, i.e. they were located in two highly inhomogeneous regions.

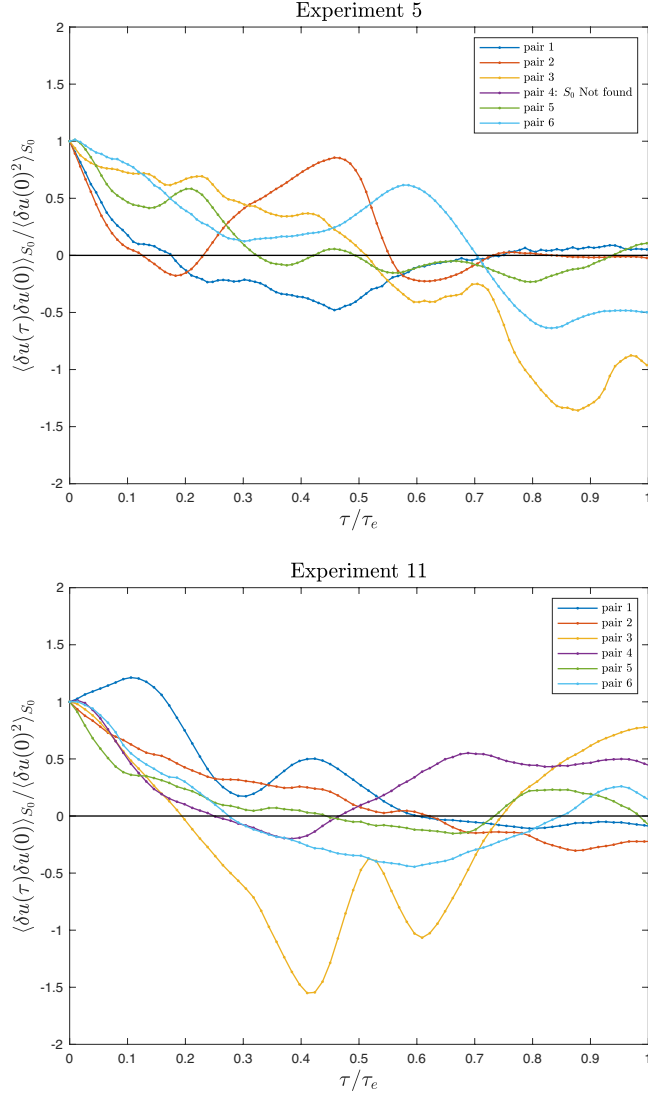
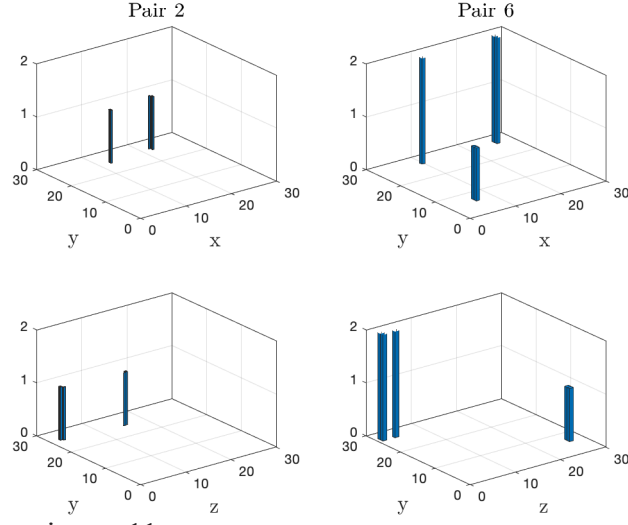


Figure 14: Lagrangian correlation of velocity differences between the particle pairs separated by S_0 , $\langle \delta u(\tau) \delta u(0) \rangle_{S_0} / \langle \delta u(0)^2 \rangle_{S_0}$, as a function of τ / τ_e for experiment 5 ($S_0 = 3.15$ cm) and experiment 11 ($S_0 = 3.29$ cm).

a) Experiment 5



b) Experiment 11

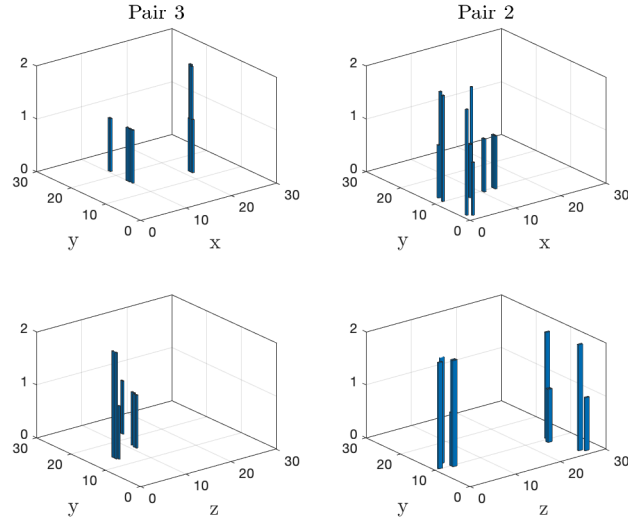


Figure 15: Histograms of the positions X, Y, Z for the particle pairs 2 and 6, and 2 and 3 in a) experiment 5 and b) experiment 11 when $S_0 \sim 3$ cm. Each bar represents the number of times the x , y or z particle positions of a pair were encountered when the particle-pair distance was about 3 cm (which corresponds to the initial times contributing to the Lagrangian correlation of velocity differences between the particle pairs in Figure 14).

601 7. Conclusions

602 In this experimental study, we departed from the theoretical framework
603 for homogeneous and isotropic turbulence and the dispersion of fluid (tracer)
604 particles to assess to what extent classic theories remain valid for the disper-
605 sion of large particles in inhomogeneous and anisotropic turbulence.

606 Our original experimental design consisted of stirring a fluid together with
607 a few almost neutrally-buoyant finite-size particles contained in a rectangular
608 tank including a mixed type of boundaries (no-slip and free surface). The
609 stirring of the two phases (fluid/particle) was achieved by a cylinder executing
610 a two-dimensional periodic Lissajoux figure enclosing a quarter of the volume
611 of the tank. Our approach consisted of recording the dynamics of the particles
612 in the entire volume of the tank, without using tracers. In doing so, we did
613 not directly probe the turbulence over the entire inertial range, but over
614 a limited scale range, which, in terms of length scales, extended from the
615 particle size to the tank's dimensions. However, the velocities of the finite-
616 size particles allowed us to determine the velocity-based properties of the
617 turbulence with tolerable accuracy.

618 Despite our initial expectations of particle collisions, only particle-cylinder
619 collisions had multiple occurrences, but their effect on the particle motion
620 remained limited. This can partly be explained by the fact that when parti-
621 cles are in the proximity of the cylinder, they frequently are engulfed in the
622 vortex surrounding the cylinder, which makes them flow around the cylinder
623 instead of colliding with it.

624 The dynamics of the particles was clearly indicative of anisotropy and
625 inhomogeneity of the turbulence at the particle scale. The walls and free-

626 surface contributed to the inhomogeneity, as shown by the velocity field. The
 627 absence of forcing motion in the direction along the axis of the cylinder re-
 628 sulted in a preferred velocity direction of the particles normal to the cylinder
 629 axis. Consequently, random preferential locations and trapping of particles
 630 along the x direction were recorded, especially at low stirring rates. Velocity
 631 fluctuations at the scale of the particles in this direction seemed insufficient to
 632 eject particles from their trapping regions. For a given period of the cylinder
 633 motion, the two opposite directions of stirring did not produce substantially
 634 different anisotropy, but produced a different inhomogeneity of the particle
 635 velocity fields.

636 Single-particle dispersion exhibited a ballistic regime at times shorter
 637 than the particle Lagrangian velocity auto-correlation time, and a short dif-
 638 fusive regime at longer times, in agreement with theoretical predictions for
 639 tracers in isotropic and homogeneous turbulence.

640 Particle-pair dispersion mostly agreed with the classic predictions for dis-
 641 persion in 3D turbulence when averaged over the six pairs, as ballistic and
 642 diffusive regimes were found. The super-diffusive regime was not observed
 643 because the time t_0 was not sufficiently smaller than the Lagrangian corre-
 644 lation time T_L . So, a temporal window for super-diffusion did not exist, and
 645 the ballistic regime transitioned directly to the diffusive regime. However,
 646 individually, some particle pairs briefly gave indications of a super-diffusive
 647 regime following the Richardson law. These particle pairs were found to
 648 be characterised by a more rapid decorrelation of their velocity differences
 649 compared to other pairs. We further made a link between the variability in
 650 the timescale of transition from the ballistic regime to Richardson's law and

the inhomogeneity of turbulence, by noting that larger decorrelation times tended to occur near the tank’s boundaries. Overall, both single-particle and particle-pair dispersion mostly agree with the ballistic and diffusive behaviours expected for 3D dispersion in homogeneous and isotropic turbulence despite the inhomogeneity and anisotropy of the turbulence in our experiments.

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790 absolute orientation of a sphere in a turbulent flow. Review of Scientific
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792 **Appendix A. Particle lifting and transient times**

793 By lifting time we mean the time it takes for particles to lift off the bottom
794 of the tank once the stirring motion has started. These lifting times inevitably
795 reflect the time it takes for momentum to reach the bottom wall after flow
796 initiation. Such a time is related to the time at which the cylinder reaches its
797 minimum distance from the bottom wall ($y_c = A$) during its first cycle, and
798 thus should be proportional to T . For initial downward cylinder motion, the
799 cylinder first reaches this height at a time $T/8$, which potentially translates
800 into an increase by a factor $3/1.75 \sim 1.7$ of the lifting times between the
801 cases with $T = 3$ s and $T = 1.75$ s. This appeared to be consistent with
802 what we observed, as the ratio of the mean of all the lifting times at $T = 3$
803 s to those at $T = 1.75$ s was 1.79. For initial upward cylinder motion, the
804 cylinder first reaches its lower height at a time $3T/8$. So, one would expect
805 that particle lifting in this latter case takes three times longer than for the
806 initial downward motion. We found an increase of that order, at $T = 3$ s,
807 as the ratio of the mean of all the lifting times for initial upward motion to
808 those for initial downward motion was 2.25. In any case, this lifting time

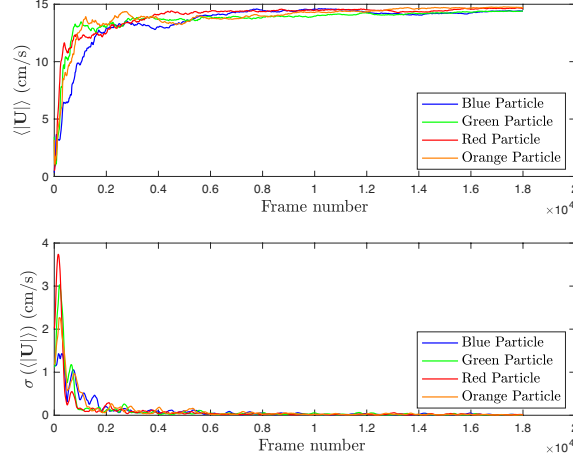


Figure A.16: (a) mean particle velocity $\langle |U| \rangle$ as a function of the number of frames. (b) Standard deviation $\sigma(\langle |U| \rangle)$ over neighbouring frames as a function of the number of frames. Data from experiment 11 have been used as an example.

809 can only be regarded as an indication of how long it takes for momentum
810 to reach the bottom wall after flow initiation, which takes from one to five
811 loops of the cylinder motion only.

812 To further assess how long it takes for the turbulent flow to be fully
813 established in the tank, we calculated the mean velocity of the particles as
814 a function of frame number. As shown in Figure A.16a, the mean velocity
815 reaches a plateau, whose start signals the onset of stationary turbulence in
816 the tank. The onset of the plateau is detected numerically by an algorithm
817 which estimates from which frame the standard deviation over a number of
818 neighbouring frames corresponding to one third of the cylinder period is first
819 less than 5% (Figure A.16b). Times t_s at which the transient is over in all
820 experiments are given in the last column of Table 1. They range from 20 to
821 34 cycles of the cylinder (equivalent to 3071 to 4711 frames over a total of

822 18,000 frames).

823 **Appendix B. Velocity time series, collisions and filtering**

824 The time series of the velocity are characterized by two types of peaks.
825 Some are short-lived (from a fifth to half a second), and result from a particle
826 colliding with the cylinder, and being kicked in either the y or z direction at
827 a speed that can exceed the maximal speed of the cylinder. Those kicks often
828 result in the particle subsequently rebounding against a wall. The transfer
829 of momentum that takes place is all the more important as the speed of
830 the cylinder is high. Table 1 of section 2.4 gives the total number of these
831 collisions for each experiment over 100 cycles. On average, a particle has a
832 collision with the cylinder every 4 to 7 cycles. Given the typical collision
833 duration, 4 to 10% of the time series recorded thus are affected by particle-
834 cylinder collisions. The second type of peaks last approximately one to two
835 seconds, and occur when particles happen to be in the wake of the cylin-
836 der. There are rare collisions with the cylinder rod, and we counted only
837 two occurrences of collision between particles across 11 experiments (only 8
838 experiments have been used).

839 To filter the collisions from the measured velocities, we assume that the
840 collisions only transfer momentum to the y and z particle velocity compo-
841 nents. The collisions are identified in the time series of U_y and U_z by auto-
842 matically finding peaks exceeding a velocity threshold that is adjusted using
843 the recorded videos. The velocities are then smoothed based on a local re-
844 gression using weighted linear least-squares and a second-degree polynomial
845 model, assigning lower weight to outliers in the regression. A zero weight

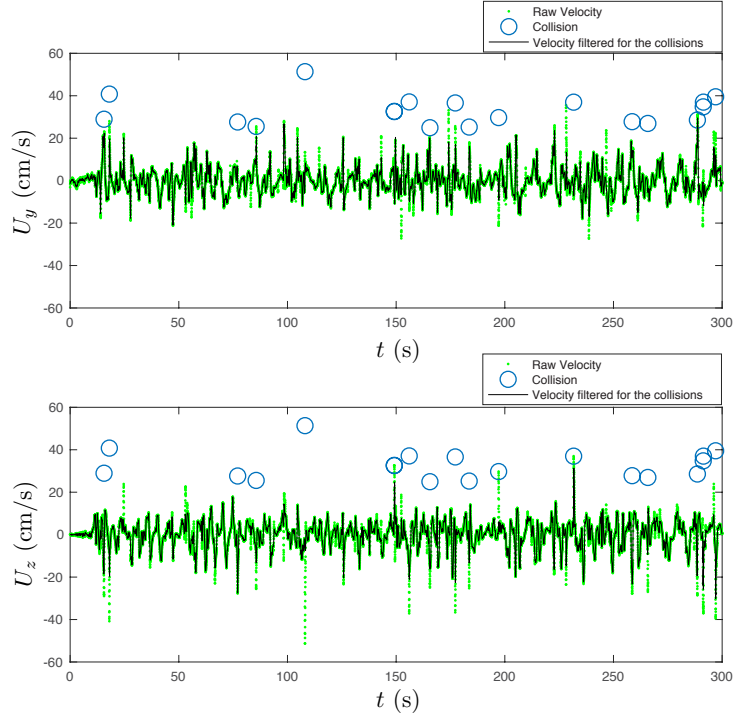


Figure B.17: Time series of the velocity components U_y and U_z for the green particle in experiment 6 before and after filtering the collisions.

846 is assigned to the data outside six mean absolute deviations. Figure B.17
847 shows an example of the procedure.

848 Figure B.18 shows the efficiency of the momentum transfer from the cylin-
849 der to the particles as well as the lack of impact from filtering. We note that
850 the transients were not removed from the time series of the velocity in this
851 instance. On average, the velocity of the particles is about 35% that of the
852 cylinder but the standard deviations are larger when the stirring is more vig-
853 orous (greater $U_c = \overline{u_c}$), which appears to result at least partly from particle
854 collisions, as shown by the fact that the standard deviation is substantially

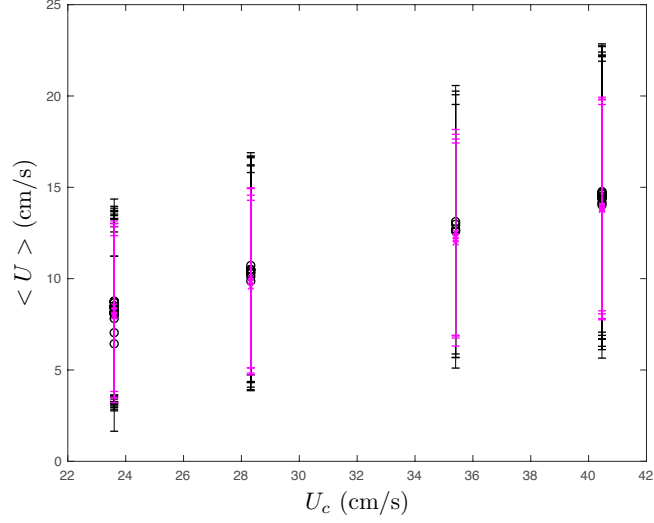


Figure B.18: Mean speed of the particles $\langle U \rangle = \langle |\mathbf{U}| \rangle$ including its standard deviation as a function of the mean cylinder velocity $U_c = \overline{u_c}$ for all experiments (in black). The mean and standard deviation from experiments 4, 6, 7 and 11 that were estimated after filtering the velocity for collisions with the cylinder are presented in magenta.

855 reduced when collisions are filtered (magenta lines).

856 We further assessed that the collisions have only minor impact on the
857 velocity and acceleration distributions (not shown).