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2 **Hydrodynamics and scale-up of bubble columns in the heterogeneous regime:**
3 **comparison of bubble size, gas holdup and liquid velocity measured**
4 **in 4 bubble columns from 0.15m to 3m in diameter**

5
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14
15 **Keywords**

16 Bubble Column, Sauter Diameter, Scale-up, Velocimetry, Heterogeneous Regime, Experimental,
17 Cross-correlation, Multiphase reactor, Clustering

18
19 **Highlights**

- 20 • Bubble Sauter diameter, gas holdup and axial liquid velocity measurements are performed
- 21 • 0.15m, 0.4m, 1m and 3m diameter bubble columns are investigated
- 22 • A wide experimental database is furnished to assist further model developments
- 23 • Void fraction and liquid velocities profiles happen to be self-similar in the heterogeneous
24 regime
- 25 • The entrained liquid flow rate, proportional to $D^{3/2}$, is only set by the column diameter
- 26 • The quantity $(gD)^{1/2}$ appears as a natural scale for mean and fluctuations velocities.

- 27 • Strong gas holdup gradients are present in the flow due to convective instabilities

28

29 **Abstract**

30 The development of CFD models coupled with Population Balance is a very promising topic
31 concerning multiphase reactors. In the case of bubbly flows and bubble columns, a serious lack of
32 local hydrodynamic characterizations still harms development and validation of relevant models. To
33 fill partially this gap, a new bubble size measurement technique, previously introduced by Maximiano
34 Raimundo et al. (2016), has been applied on a very wide range of bubble column diameters (from
35 0.15m to 3m) and superficial gas velocities (from 0.06m/s to 0.35m/s). Size measurements have been
36 coupled with others concerning gas holdup and axial liquid velocity, in order to provide an
37 experimental database allowing to clarify the scale-up rules and to assist future modelling works.
38 Average bubble sizes have been measured as globally similar at every scale. Measured holdup and
39 average liquid velocity confirm already reported behaviours at lower column diameters. Liquid
40 velocity fluctuations also follow self-similar radial profiles and are proportional to the average liquid
41 velocity at the centre of the column leading to a strong turbulence intensity. The fact that the
42 quantity $(gD)^{1/2}$ appears as a natural velocity scale and the presence of strong gas-holdup gradients
43 underline the similarity between bubble columns operated heterogeneous regime and free thermal
44 convection in pipes.

45 **1. Introduction**

46

47 Although being a major research topic for decades, the design and scale-up of bubble column
48 reactors is still a difficult task for chemical engineers, as associated hydrodynamic and transfer
49 phenomena are very sensitive to physical properties of fluids, operating conditions and geometrical
50 settings. A huge number of correlations have been developed in the past to assist scale-up by means
51 of 0D or 1D approaches (Deckwer, 1992; Kantarci et al., 2005, Besagni et al., 2018). Available
52 correlations are generally valid in rather narrow ranges of design parameters, physical properties and

53 operating conditions. Nowadays it is generally admitted that CFD can be used to secure industrial
54 designs when outside of validation ranges of empirical correlations. But preliminary parameter
55 adjustments are generally necessary to fit initial simulations on a given well investigated scale. The
56 fitting step keeps CFD far from being a full predictive tool. Parameters to be adjusted can concern
57 turbulence modelling, including Bubble Induced Turbulence (McClure et al., 2014 ; Joshi, 2001), but
58 above all, interfacial forces (Jakobsen et al., 2005). There is no real consensus concerning the forces
59 to consider to achieve realistic simulations, except for the drag law that all authors point out as
60 impacting mostly the calculated gas holdup. The use of lift, added mass, wall lubrication and
61 turbulent dispersion are sometimes suggested to improve the agreement between experimental
62 data and CFD simulations (Krishna et al., 2001; McClure et al., 2015). However, during a previous
63 work (Gemello et al., 2018b), it was found that time-averaged hydrodynamics (i.e. gas holdup and
64 liquid velocity profiles) of several bubble columns were satisfactorily predicted using an apparent
65 drag formulation as the only interfacial force. Nevertheless, the mean bubble diameter had to be
66 beforehand known to perform such simulations, as it conditions directly the drag force, and thus gas
67 holdup (Guedon et al., 2017). To overcome this limitation in presence of breakage and coalescence
68 phenomena, one powerful possibility consists in associating CFD with a population balance on Bubble
69 Size Distribution (BSD) (Buffo et al., 2013, Lehr et al., 2004, Sanya et al., 2005). If efficient, this would
70 make CFD a much more predictive tool.

71 Whether to develop interfacial force models for CFD or Population Balance kernels, experimental
72 data are essential, including information concerning bubble sizes in flow regimes of industrial
73 interest. Only a few experimental data are usable for this purpose. Major of them are based on
74 multitips optical probes (Chaumat et al., 2005, Xue, 2004, Xue et al., 2008, McClure et al., 2017).
75 Multitips technique ideally allows to measure both bubble chord and velocity distributions. The
76 technique is based on the processing of the phase-signal delay between probes located at different
77 heights. As often pointed out by authors, its accuracy is acceptable at the centre of the column,
78 where bubble have mostly vertical trajectories, but it rapidly decreases with the distance from it, as

79 bubble trajectories become more chaotic and include downward motion and the delay between
80 signals is not linkable with the axial bubble velocity anymore. In addition, the performances of
81 multitips probes in terms of minimum detectable bubble size are not clearly determined. An
82 alternative approach consists in the use of a measurement technique independent of the bubble
83 trajectory. This is the case of the cross correlation (CC) technique developed by Maximiano
84 Raimundo et al. (2016) and validated in the heterogeneous regime by comparison with endoscopic
85 measurements. The CC technique does not measure the global BSD, but only the mean Sauter
86 diameter, i.e. the ratio between 3rd and 2nd moments of the BSD. But the measurements have the
87 same accuracy regardless of the radial position. It has been applied recently to the study of the effect
88 of water quality and the gas sparger design, in a 400mm column diameter (Gemello et al., 2018a).

89 In the present work, the same cross correlation technique has been used to measure bubble sizes in
90 four bubbles columns from 0.15m to 3m in internal diameter, with the purpose of investigating the
91 effect of scale-up on bubble sizes, for an identical gas/liquid system. Distilled or demineralized water
92 are generally used in academic studies, but as tap water and air were the only fluids usable in the
93 biggest column, a similar system has been used at all scales. The tap water used in the present work
94 can be qualified as partially contaminated, as it exhibits a less coalescent behaviour than
95 demineralized water. One limitation of using tap water is that its quality depends on the location and
96 can lead to difficulties to repeat experiments elsewhere. To overcome this issue, It has been shown
97 in (Gemello et al., 2018a) that the tap water used in the present study behaves as a solution of
98 demineralized water containing 0.01% in weight of ethanol (Gemello et al., 2018a). In addition to the
99 bubble size, the global ($\langle\alpha_G\rangle$) and local (α_G) gas fractions have been measured via the liquid
100 elevation measurement and the optical probes used for the cross correlation technique, respectively.

101 Axial liquid velocity profiles (average and fluctuation RMS values) have been also measured with a
102 modified Pitot tube, so-called Pavlov tube technique (Forret et al., 2003). This technique is the only
103 one usable at high gas holdup in all column sizes. Measurements have been done at different V_{sg}
104 from 0.03m/s to 0.35m/s depending on bubble columns. In the following, the four experimental

105 setups are introduced, as well as the involved measurement techniques. Then results concerning gas
106 fraction, liquid velocity and bubble sizes are summarized and discussed. A discussion concerning the
107 presence of bubble clusters and of voids (regions with few bubbles) and the role of these meso-scale
108 structures on the hydrodynamics of the heterogeneous regime is finally proposed.

109 2. Experimental setups

110 In the present work, four different columns with 0.15 m, 0.4 m and 1 m and 3m inner diameters have
111 been tested. All the experiments were conducted with compressed air, which had been dried and
112 cooled, as the gas phase and with water, with no net liquid flow rate (batch mode), as the liquid
113 phase. In the different columns, a partially contaminated tap water is used. Table 1 reports water
114 analysis.

115 **Table 1: tap water analysis @ 20°C**

Surface tension (mN/m)	67
Conductivity @ 25°C ($\mu\text{S}/\text{cm}$)	559
Carbonate (mg/L)	≈ 0
Hydrogenocarbonate (mg/L)	251
pH	7.9

116
117 In the four columns, the gas was introduced into the column from the base. The volumetric flow rate
118 was determined by a row of several calibrated flow-meters, with an uncertainty given by suppliers
119 equal to 1.6%. The gas distributors are perforated plates, the number of holes and their diameters in
120 the different columns are given in the following table. The injectors have been designed to ensure
121 comparable porosities (at a value small enough to uncouple the gas injection and the bubble column
122 dynamics), and also to form gas jets with comparable ejection velocities (as the ejection velocity is
123 equal to the superficial gas velocity divided by the porosity of the injection plates) which are
124 expected to experience similar break-up process and thus to deliver comparable bubble size
125 distributions at injection in all columns. Columns of 0.15 and 0.4m inner diameter exhibit exactly the
126 same conditions of gas sparging with orifices 1mm in diameter and 10mm long. The injector for the
127 1m inner diameter column has the same porosity, i.e. the same gas velocity at the outlet of injections

128 holes (from 13 to 140m/s), but the diameter of holes is doubled. Indeed, the effect of the hole size
 129 can be considered as small as hole diameters stand much smaller than bubble sizes in the columns.
 130 The biggest column is equipped with a different sparger, with larger and shorter injection holes and
 131 with a lower porosity, but leading to comparable ejection gas velocities (from 37 to 100m/s).
 132 However, Gemello et al. (2018a) have studied the impact of the sparger on the bubble size in the
 133 0.4m inner diameter column used in the present study. It has been found that the sparger design has
 134 an important effect at the bottom of the column. This effect is due both to the spatial gas
 135 distribution, conditioned by the number of holes, and the bubble sizes generated by different
 136 spargers, which can be very disparate. But the effect of the sparger becomes negligible above a
 137 distance of 0.4m from the bottom of the column because of the dominant breakup phenomena
 138 whatever the water quality. This behaviour is in agreement with the usual observation that, in the
 139 heterogeneous regime, quantities such as local void fraction and velocities are weakly sensitive to
 140 injection conditions when $H_0/D > 2$ and when the data are gathered within the (almost) fully
 141 developed region (Forret et al., 2006). In addition, it will be shown in section 4 that the mean bubble
 142 sizes are similar for all columns and all flow conditions, probably thanks to an efficient break-up
 143 process of the gas jets formed at injection.

144 **Table 2: gas distributors characteristics in the different columns**

Column	Number of holes	Hole diameter (mm)	Length/diameter ratio of injectors	Porosity (%)	Pitch (mm)
Ø0.15m	55	1	10	0.24	15 (triangular)
Ø0.4m	391	1	10	0.24	15 (triangular)
Ø1m	613	2	10	0.25	37 (triangular)
Ø3m	164	9	0.5	0.15	200 (squared)

145
 146 The columns used in this study present of pairs of diametrically opposed holes at different heights
 147 (0.5m, 1m, 1.5m, 2.6m, 3.4m and 4.6m above the gas distributor in the case of the 1 m in diameter
 148 column), enabling the position of different technical means (Pavlov tube, optical probe...) or can be
 149 used as pressure taps. The scheme of the 1 m in diameter column is given in the Figure 1. The
 150 scheme of the other columns is very similar to this one.

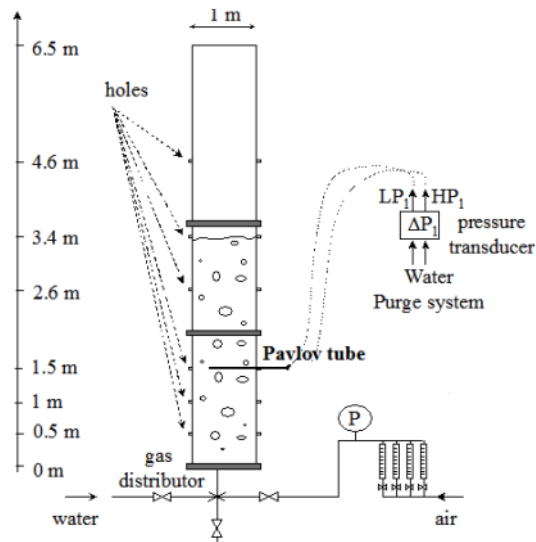


Figure 1: schematic view of the 1 m in diameter column and its perforated plate

151

152

153

154 In addition, the 0.15 m, 0.4 m and 1 m diameter columns were operated with a liquid static

155 height/column diameter "aspect ratio" (H_0/D) of 4. The 3 m diameter column has operated with 2.2

156 liquid static height/diameter ratio because the column is only 12 m high. Besagni et al. (2017a)

157 studied the effect of the aspect ratio on the gas holdup and suggested a critical aspect ratio of 5.

158 Nevertheless, exhaustive experimental results reported by these authors show that above 3-4, the

159 effect of the aspect ratio on gas holdup becomes very weak. This is also confirmed by the

160 measurements of Sasaki et al. (2016). Measurements have been performed at an elevation height of

161 2.5 times the column diameter ($H/D=2.5$), except in the 3m column, inside which measurements are

162 performed at $H/D=2$. According to Forret et al. (2006), such elevations are indeed within the fully

163 developed flow region. However, the recent work of Guan et al. (2016) may lead to moderate this

164 conclusion. They found that gas holdup and liquid velocity radial profiles were never totally

165 independent of the axial position in a column of 0.8m in diameter. Yet, using a uniform gas sparger,

166 the flow happens to be almost fully developed between H/D from 2 to 4. Guan et al. (2016) also

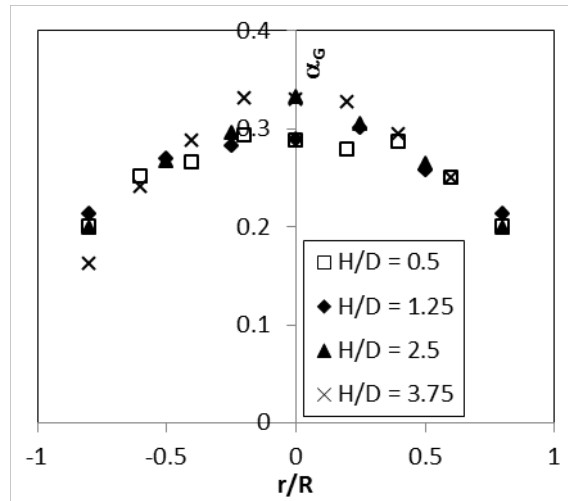
167 suggested that the height of flow development depends on the column diameter. Figure 2 reports

168 gas holdup profiles measured in the $\varnothing 0.4$ m column at $V_{sg}=0.16$ m/s and at different heights. Profiles

169 measured at $H/D = 2.5$ and 3.75 are similar, while some discrepancies are observed below, especially

170 in the middle of the column. In summary, the aspect ratio of the studied columns is considered to be
 171 sufficient to neglect its effect on hydrodynamics, and the height of measurement is comprised within
 172 the range of fully developed flow.

173



174

175 **Figure 2 : Comparison of gas holdup profiles measured in the Ø0.4m column at different H/D. V_{sg}=0.15m/s (H₀/D=4)**
 176

177

178 3. Measurement techniques

179 3.1. Gas holdup

180 The global gas holdup can be directly calculated by the visual observation of the expansion of liquid
 181 height by:

182

$$183 \langle \alpha_G \rangle = 1 - \frac{H_0}{H_D} \quad (1)$$

184

185 Where $\langle \alpha_G \rangle$ represents the global void fraction, H_0 represents the non-aerated liquid height and H_D
 186 the aerated liquid height. In the present study, the liquid height is measured by visual observation
 187 along a graduated rule, and fluctuations of liquid level induce a relatively high uncertainty of the

188 measurement, estimated at 10%. Sasaki et al. (2016) strongly decreased the uncertainty while using
189 image processing to detect liquid level. The local gas holdup was measured by a light reflective
190 optical probe. The beam is generated by a laser and is sent through the optical fiber to the point of
191 the probe. If the probe is in contact with the liquid phase, the light beam is refracted through the
192 liquid media. Otherwise, if the probe tip is in contact with the gas phase, the light beam is reflected
193 by the bubble into the probe tip and then detected by a photodiode where the light beam intensity is
194 converted into voltage. Adding the time that the probe detects the gas phase, the local gas hold up is
195 determined by the following equation.

196

$$197 \quad \alpha_G = \frac{\text{Cumulated gas time}}{\text{Experimental time}} \quad (2)$$

198

199 The local gas holdup detected with the optical probe was compared with global gas hold-up for
200 heterogeneous conditions in the 0.4m bubble column. The agreement was within $\pm 15\%$ (Maximiano
201 Raimundo et al., 2016), which is satisfactory owing to the uncertainty on the determination of $\langle \alpha_G \rangle$.

202

203 3.2. Cross-Correlation (Bubble size)

204 In order to characterize bubbles in dense, heterogeneous bubbly flows such as those encountered in
205 industrial bubble columns, a recently developed measuring technique based on the spatial
206 correlation of phase indicator functions is proposed (Maximiano Raimundo et al., 2016). The
207 normalized cross-correlation is a function that quantifies the similarity of two binary signals by
208 analysing both signals simultaneously:

$$209 \quad CC = \frac{\int_{t=0}^{t_{exp}} \text{Signal}_A(t) \times \text{Signal}_B(t) dt}{\int_{t=0}^{t_{exp}} \text{Signal}_A(t) dt} \quad (3)$$

210

211 Here $Signal_A$ and $Signal_B$ represent the raw signals coming from two different probes at the same
 212 elevation in the column at a radial distance (d) and t_{exp} represents the time of registration. The
 213 cross-correlation value is maximal if both probes are at the same point in space (distance between
 214 probes d of 0 mm), since the signals will be identical. In a single bubble-probe interaction, as the
 215 distance between the probes increases, the cross-correlation of the probe signals will decrease. The
 216 cross-correlation will be zero when the distance between probes becomes larger than the bubble
 217 horizontal diameter due to the fact that the same bubble cannot be detected by both probes at the
 218 same time. Nevertheless, in a bubble column, the cross-correlation is never zero even if the distance
 219 between probes reaches a value much larger than the largest bubble horizontal diameter. At large
 220 distances, the cross-correlation tends to the local gas hold-up α_G . It has been shown that some
 221 information related to the horizontal bubble size can be extracted from the initial linear slope of the
 222 cross-correlation curve as a function of probes distances (Maximiano Raimundo et al., 2016). The
 223 relationship between the Sauter mean value of horizontal bubble diameters, noted $d_{32,h}$, the
 224 correlation coefficient CC and the inter-probes distance d always expresses as follows:

$$226 \quad d_{32,h} = a \frac{d}{(CC(d)-1)} \quad (4)$$

227
 228 where the prefactor a is function of the bubble shape, as detailed in the following table (details in
 229 Maximiano Raimundo et al., 2016).

231 **Table 3: Prefactors a for various bubble shapes**

	Sphere	Oblate	Prolate with Ecc=0,7
Prefactor a	-1.472	-1.5978	-1.7

232
 233 To validate this method, it has been compared to endoscopic imaging. The Sauter mean horizontal
 234 diameters detected with the correlation technique happen to be in good agreement with those
 235 provided by endoscopic imaging. This latest method enables also the determination of the average

236 eccentricity (ecc) : the latter has been measured equal to 0.7 for all flow conditions and all column
237 dimensions except for the 3m I.D. column for which the eccentricity was not measured. Therefore,
238 the Sauter mean diameter d_{32} can be linked to the $d_{32,h}$ measured by the cross-correlation as follows.

239

$$240 \quad d_{32} = d_{32,h} * ecc^{1/3} \quad (5)$$

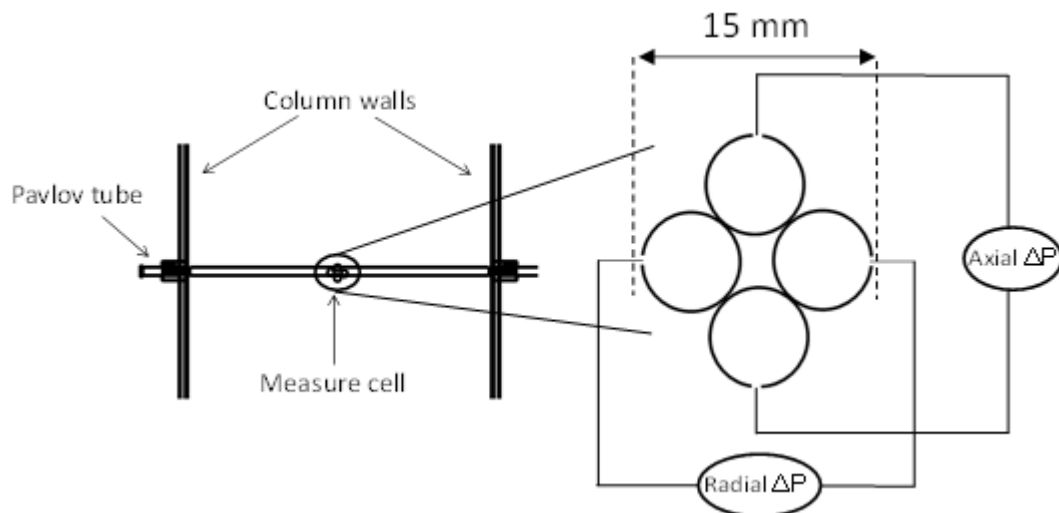
241

242 with $ecc = 0.7$.

243

244 3.3. Pavlov tube(axial liquid velocity)

245 The Pavlov tube used in this work is composed by four 5 mm diameter tubes, arranged as it is shown
246 in the right-hand side of the Figure 3. The four tubes are placed inside a 30 mm diameter tube, which
247 crosses horizontally the column, in order to obtain rigidity and to allow horizontal displacement. The
248 5 mm diameter tubes are completely closed, except for a 0.5 mm diameter lateral hole that assures
249 that the pressure inside the tube is equal to the one in the column.



250

251 **Figure 3: Schematic representation of the Pavlov tube (Axial measurements are performed along a vertical axis, radial**
252 **measurements are performed along a horizontal axis)**

253

254 The holes of the tubes on the vertical plan are placed in the same axis but in opposite
255 directions, as depicted in Figure 3. Additionally, each tube on the vertical direction is connected to

256 one of the chambers of a differential pressure sensor. These tubes serve to measure the axial
257 pressure difference, since the holes on the tubes are aligned with the column vertical axis. The
258 Pavlov tube allows also the measurement of the radial liquid velocity, quite low in this study, not
259 discussed in this paper. The measurements of the axial pressure differences (ΔP) can be used to
260 calculate the instantaneous axial liquid velocity recurring to the following equation.

$$261 \quad U_{(x,t)} = \begin{cases} \sqrt{\frac{2\Delta P(x,t)}{\rho_l}} & \text{if } \Delta P(x,t) \geq 0 \\ -\sqrt{\frac{-2\Delta P(x,t)}{\rho_l}} & \text{if } \Delta P(x,t) < 0 \end{cases} \quad (6)$$

262 The axial ΔP measurement was made with a differential pressure transmitter Rosemount 3051T with
263 a range of ± 60 mbar, a frequency of 8 Hz and resolution of 0.01 mbar. Let us mention that corrections
264 of eq.(6) accounting for local void fraction have been sometimes proposed. On one hand, the largest
265 of these corrections is the one proposed by Bosio and Malnes as discussed by Riemann et al. (1984).
266 According to that correction, the difference in velocity prediction remains smaller than 3% since
267 the void fraction are always below 35% in all our experiments. On another hand, these corrections
268 consider that the dynamic pressure collected from pitot or pavlov tubes in two-phase flows is
269 associated with the density and the velocity of the mixture. However, in bubbly flows, the orifices of
270 pitot or pavlov tubes are always wetted, meaning that the gas phase is never detected as such.
271 Therefore, the local stopping pressure detected with such sensors provides a measure of the local
272 liquid velocity even when the sensor is in the vicinity of a bubble, and no correction is required. All
273 Pavlov tube measurements presented hereafter are grounded on eq.(6).

274 The radial profiles are easily obtained by moving the measuring cell along the column radius. The
275 axial velocity fluctuations can be calculated through the instantaneous and the averaged velocity,
276 using Reynolds decomposition, as presented in the following equation.

277

$$278 \quad U(x,t) = U(x) + u'(x,t) \quad (7)$$

279

280 where $U(x)$ represents the mean liquid velocity, $u'(x,t)$ represents the liquid fluctuation velocity
281 at the instant t and $U(x,t)$ represents the liquid velocity at the instant t .

282

283 **4. Results**

284 Thereafter, the discussion focuses on the heterogeneous regime. On all columns, one can observe in
285 Figure 4, that the almost linear behaviour of $\langle\alpha_G\rangle$ with V_{sg} is observed only below 4-5cm/s,
286 approximately. This value corresponds to the transition to the heterogeneous regime.

287 *4.1. Gas holdup*

288 Global gas volume fractions have been measured for all columns and V_{sg} . Radial gas holdup profiles
289 have been measured with the optical probes at $H/D=2.5$ (Figure 4). Schweitzer et al. (2001) found
290 similar profiles of $\alpha_G(x)$ when normalized by the average gas holdup in the column $\langle\alpha_G\rangle$. Authors
291 suggest the correlation reported in eq.(8) to represent holdup profiles. The correlation has been
292 initially developed and validated in a 50mm inner diameter column for V_{sg} between 0.03 to 0.25m/s.
293 Its range of validity has been extended by Forret et al. (2006) to columns up to 1m of inner diameter.

294

$$295 \alpha_G(x) = \langle\alpha_G\rangle[-1.638(x^6 - 1) + 1.228(x^4 - 1) - 0.939(x^2 - 1)] \quad (8)$$

296

297 On Figure 4 (left), unsurprisingly it is verified that gas holdup follows the same profile at any scale,
298 with an average error below 5%. Concerning the average gas holdup, keeping in mind that the liquid
299 elevation measurement method eq.(1) is relatively imprecise (+/- 10%), different correlations
300 validated in air/water systems may be found in acceptable agreement with present experimental
301 data. As the gas holdups have been measured with the same technique and using the same water,
302 experimental data have been used to fit a new correlation in the aim to extract the specific effect of
303 scale up in the heterogeneous regime. Remember that existing models have been generally

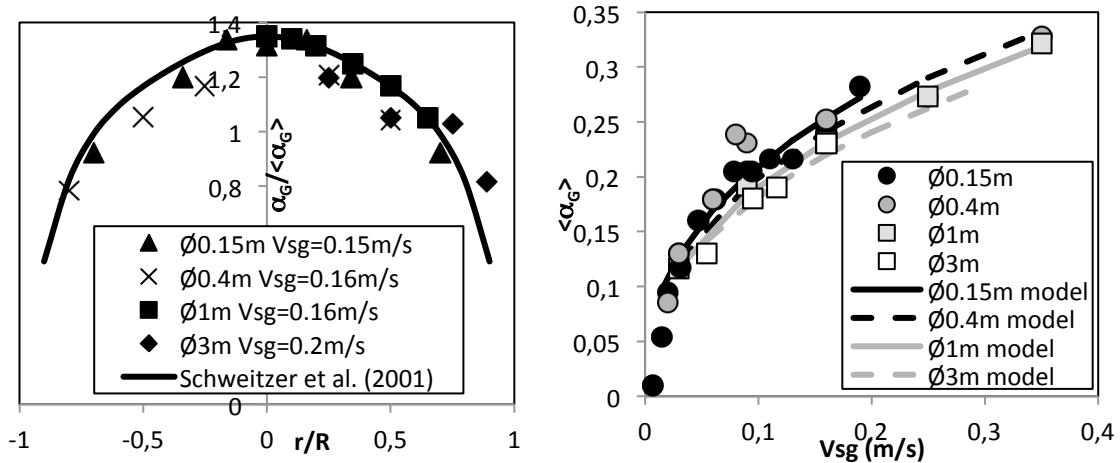
304 developed based on a single column diameter, generally lower than 0.4m. The following correlation
 305 has be found to predict $\langle \alpha_G \rangle$ with an error below 6% when $V_{sg} > 0.1\text{m/s}$:

306

$$307 \quad \langle \alpha_G \rangle = 0.49 \cdot V_{sg}^{0.41} \cdot D^{-0.047} \quad (9)$$

308

309 Measured and predicted average gas holdups are compared in Figure 4 (right). Only experiments
 310 corresponding to $V_{sg} > 0.05 \text{ m/s}$ are considered to fit eq.(9), given that gas holdup is proportional to
 311 V_{sg} in the homogeneous regime. It is observed a small, but not negligible effect of the column
 312 diameter on the gas holdup, especially in intermediate range of V_{sg} , between 0.1 and 0.2 m/s.
 313 Although most of existing correlations do not include the effect of D (Kantarci et al., 2005), the
 314 question of the impact of the column diameter on the gas holdup is still not completely settled as
 315 notably discussed by Rollbush et al. (2015). Additional experiments may although be necessary at
 316 higher D to confirm this trend, as at high V_{sg} measurements in 0.4 and 1m diameters are relatively
 317 close. Alternatively the eq. (9) can be replaced by an equation involving only V_{sg} with an exponent of
 318 0.41 and a prefactor of 0.5 instead of 0.49, with a mean error of 8%.



319

320 **Figure 4: Gas holdup profiles at H/D=2.5, normalized by the average gas holdup (left), and average gas holdup**
 321 **measurements (right). Comparison with correlations (eq. 8&9).**

322

323 4.2. Liquid velocity

324 In a similar manner, Forret et al. (2006) suggested to normalize axial liquid average velocity profiles
325 by the liquid velocity at the center U_0 , and found a good agreement with the following polynomial
326 model:

327

$$328 \quad U(x) = \frac{U_0}{a-c} [a \cdot \exp(-b \cdot x^2) - c] \quad (10)$$

329

330 With $a=2.976$, $b=0.943$, $c=1.848$. The eq. (10) has been compared to the present experimental
331 measurements of $U(x)$, and a very good agreement has been found, except in the largest column
332 where the magnitude of the liquid velocity close to the walls is slightly underestimated by the model.
333 The comparison between $U(x)/U_0$ and eq.(10) are reported in Figure 5. The liquid velocity at the
334 center can also be correlated with V_{sg} and D . The equation (11) presents the result of the regression,
335 which predicts U_0 with an average error below 8%:

336

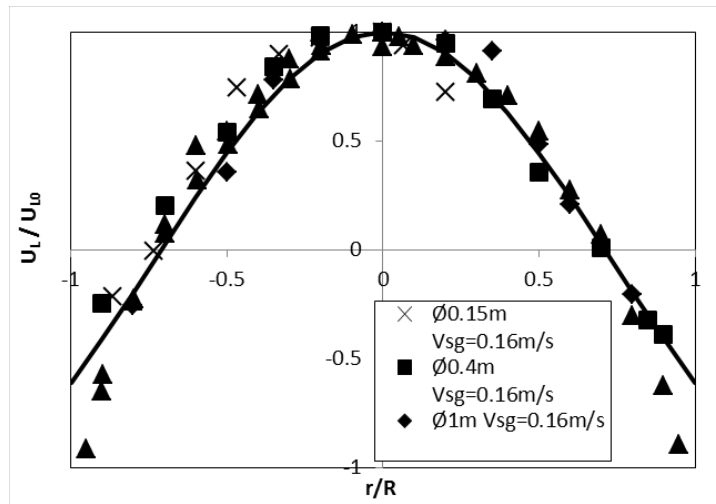
$$337 \quad U_0 = 1.35 \cdot V_{sg}^{0.16} \cdot D^{0.40} \quad (11)$$

338

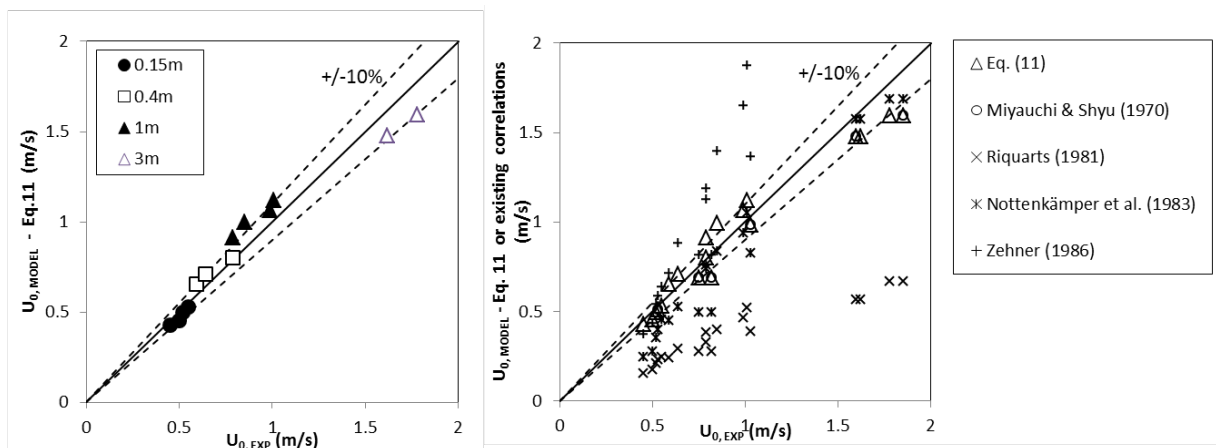
339 Figure 6 (left) presents the parity diagram between eq.(11) and experimental measurements in
340 different columns. In Figure 6 (right), experimental results are also compared to a short selection of
341 existing correlations of Miyauchi & Shyu (1970), Riquarts (1981), Nottenkämper (1983) and Zehner
342 (1986). In this figure, each experimental centreline velocity, reported in abscissa, is compared to the
343 velocity given by the different listed correlations, including eq.11, at the same operating V_{sg} and D ,
344 which are reported in ordinate. This comparison shows that our experiments are in good agreement
345 with previous works of Miyauchi & Shyu (1970) and Nottenkämper (1983), but not with others,
346 pointing out the disparity of experimental results obtained in different laboratories. This disparity can
347 have different origins, as the different measurement methods, different quality of water, different

348 gas sparging conditions or different column sizes. The present study allows to eliminate the 3 first
 349 possibilities.

350 While the exponent on D in eq. (11) is quite classical – in the range of 0.3-0.7, the exponent on Vsg
 351 can be considered lower than generally observed (0.3-0.5). This later result may be a consequence of
 352 the quality of water, which partially prevents coalescence when compared with non-contaminated
 353 water, resulting in a less pronounced heterogeneity of bubble sizes and possibly a lower liquid
 354 recirculation as suggested by Rollbusch et al. (2015). Thus, from the above results, it happens that
 355 the flows in bubble column operated in the heterogeneous regime self-organise, leading to self-
 356 similar radial profiles for the void fraction and for the mean liquid velocities.



357
 358 **Figure 5: Normalized liquid velocity profiles (left) and comparison between the axial average liquid velocity at the center**
 359 **U_0 and eq.(11).**



360
 361
 362 **Figure 6: Centre-line velocity parity diagram of eq.11 (left) ; parity diagram of different correlations (right)**

363

364

365 Besides, a well-known behaviour concerning the liquid velocity profile is observed: the section of the
366 columns can be divided into two surfaces of equal area: the core region when $r < \frac{\sqrt{2}}{4}D$ or $x < 0.71$, where
367 U is positive, and the external region where U is negative. In the core region, the liquid upward flux
368 can be calculated as:

369

$$370 \quad Q_{L,up} = \int_0^{\frac{\sqrt{2}}{4}D} 2\pi \cdot r \cdot (1 - \alpha_G) \cdot U(r) \cdot dr \quad (12)$$

371

372 The calculated liquid upward flux for three V_{sg} and for all the investigated columns are reported in
373 Figure . The liquid upward flux surprisingly happens to be independent of the superficial gas velocity
374 in the heterogeneous regime. Results correlates very well with $\propto D^{2.5}$, implying that the average
375 liquid velocity in the core region follows a trend $\propto \sqrt{D}$. One possible explanation of that could be the
376 predominant role of the natural velocity scale \sqrt{gD} . If we define a Froude number based on the
377 mean liquid velocity in the core region of section $S_{core} (= \pi D^2/8)$, a constant value of the Froude
378 number is observed:

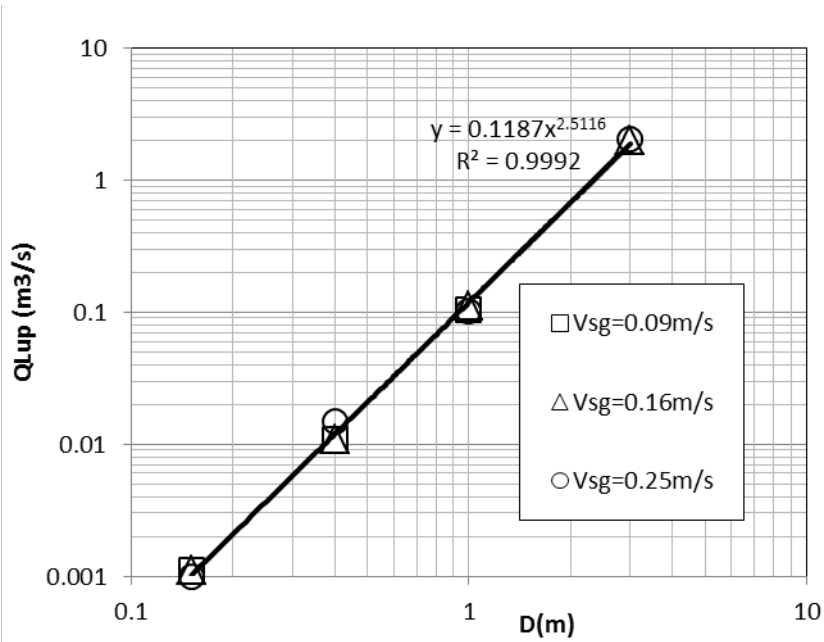
379

$$380 \quad Fr = \frac{Q_{L,up}}{S_{core}\sqrt{gD}} \approx 0.024 \quad (13)$$

381

382 This result also illustrates the close link between liquid volume fraction and velocity profiles, as both
383 change with the gas flow rate but not their mutual product. The above velocity scaling and the
384 observed flow organisation are reminiscent of free thermal convection in pipes for which the natural
385 scale for velocity has been shown to be the free fall velocity under buoyancy acceleration evaluated
386 for a length scale equal to the pipe diameter (Tisserand et al., 2010, Rusaouen et al., 2014).

387



388

389 **Figure 7: Liquid upward flux in the core region as a function of the bubble column diameter for the heterogeneous**
 390 **regime.**

391

392 *Relative velocity*

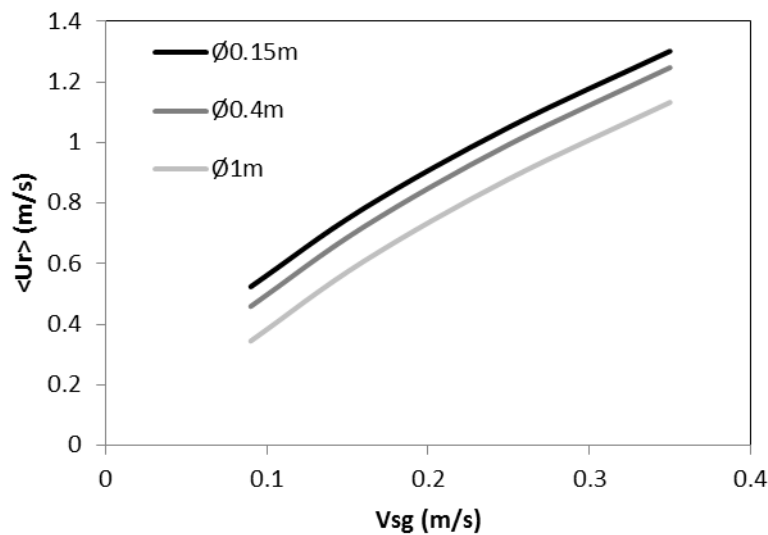
393 In the core region, the mean flow is directed upward and it is thus similar to a co-current upward
 394 two-phase flow. According to classical kinematic approaches (e.g. Zuber and Findlay 1965), the
 395 difference between the gas flow rate fraction defined as the volumetric gas flow divided by the sum
 396 of liquid and gas volumetric flow rates and the void fraction is controlled by the relative velocity
 397 between phases. In the same spirit, let us introduce the apparent (in the sense that it is a global
 398 quantity at the scale of the core region of the column and not a local one) relative velocity between
 399 gas and liquid $\langle Ur \rangle$ in the core region as the difference of mean velocities between gas and liquid in
 400 the core section. Each mean velocity is calculated as the phasic volumetric flow rate divided by the
 401 associated cross section, i.e. the core section multiplied by the volume fraction of the concerned
 402 phase in this section:

403

$$404 \langle Ur \rangle = \frac{Q_{G,core}}{\langle \alpha_G \rangle_{core} \cdot S_{core}} - \frac{Q_{L,up}}{(1 - \langle \alpha_G \rangle_{core}) \cdot S_{core}} \quad (14)$$

405

406 Where $Q_{G,core}$ is the gas flow rate in the core region, $Q_{L,core}$ the liquid flow rate in the core region is
 407 given by (eq.(13) and $\langle\alpha_G\rangle_{core}$ is computed from eq. (8) and is found $\approx 1.33 \langle\alpha_G\rangle$. $Q_{G,core}$ can be
 408 roughly estimated by the total gas flow rate Q_G . Note that this approximation tends to minimize the
 409 estimation of the relative velocity as the recirculation of a part of the bubbles is not taken into
 410 account. The estimation of relative velocities are reported in Figure 8 for $V_{sg} > 0.09\text{m/s}$ and for three
 411 columns. The relative velocity increases with V_{sg} and achieves values that are much higher than the
 412 terminal bubble velocity ($\approx 0.22\text{m/s}$). Such an increase in the bubble relative velocity in the
 413 heterogeneous regime has been evoked by Ruzicka (2013). Possible origin of this phenomenon is
 414 discussed below.



415
 416 **Figure 8: Estimation of the relative velocity in the core region, considering that the gas flow rate in the core region equals**
 417 **the totality (U_r max) or the half (U_r min) of the total gas flow rate.**

418
 419 *Fluctuating velocity*

420 RMS of axial liquid velocity fluctuations (u') have been measured in various operating conditions.
 421 Results presented in Figure 9 (left) point out that u' radial profiles also follow self-similar profiles. The
 422 following polynomial function has been regressed in eq. (15) and is also reported in Figure 9 (left) to
 423 illustrate the self-similarity of profiles.

424

425 $u'(x) = u'(0.71) \cdot [a + bx^2 + cx^4 + dx^6]$ (15)

426

427 With $a=0.48$, $b=2.25$, $c=-2.6$, $d=0.40$. The normalization of the u' profiles can be done whatever the
 428 position of reference (x) but it has been found that using the maximal u' value minimizes the error of
 429 residues of the regression. The maximal value of u' is always located at the boundary between
 430 positive and negative liquid velocities ($x = \sqrt{2}/2 \approx 0.71$). When comparing u'_{max} at different scales, it
 431 is found proportional to U_0 . As a consequence, the maximal u' value can be written as follows:

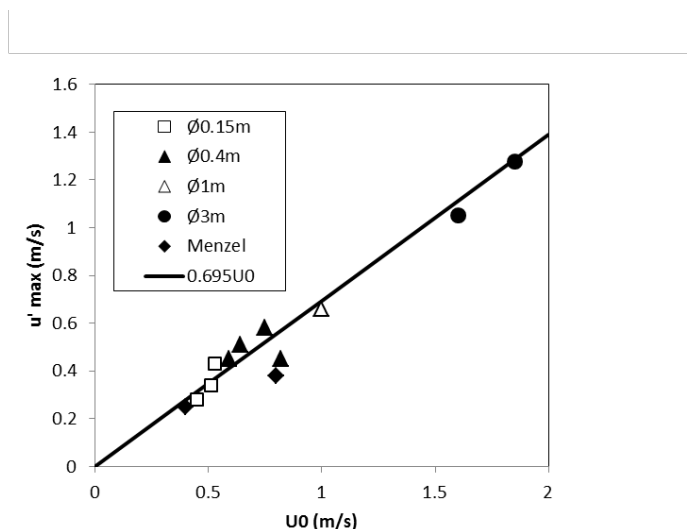
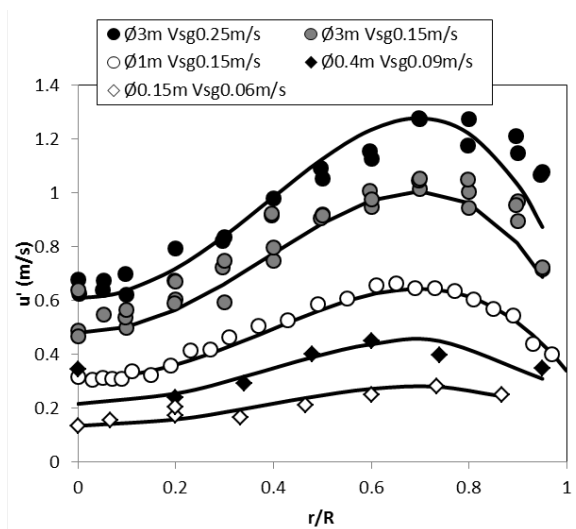
432

433 $u'(x = \sqrt{2}/2) = u'_{max} = 0.695 U_0$ (16)

434

435 u'_{max} is reported in Figure 9 (right) for the different columns. The experimental data of Menzel et al.
 436 (1990) using a hot wire probe is also reported on the same figure. The consistency between the
 437 values measured with different techniques confirms the reliability of Pavlov tube measurements
 438 concerning axial liquid velocity fluctuations. Note that in the core region, the liquid fluctuations are
 439 about half their maximum, corresponding to a turbulent intensity u'/U_0 about 25-30%. Hence, large
 440 velocity fluctuations are present over the entire cross-section at all flow conditions: that feature is a
 441 clear characteristic of the heterogeneous regime.

442



443

444 **Figure 9: u' radial profiles (left) and correlation between $u'(x=0,71)$ and U_0 (right).**

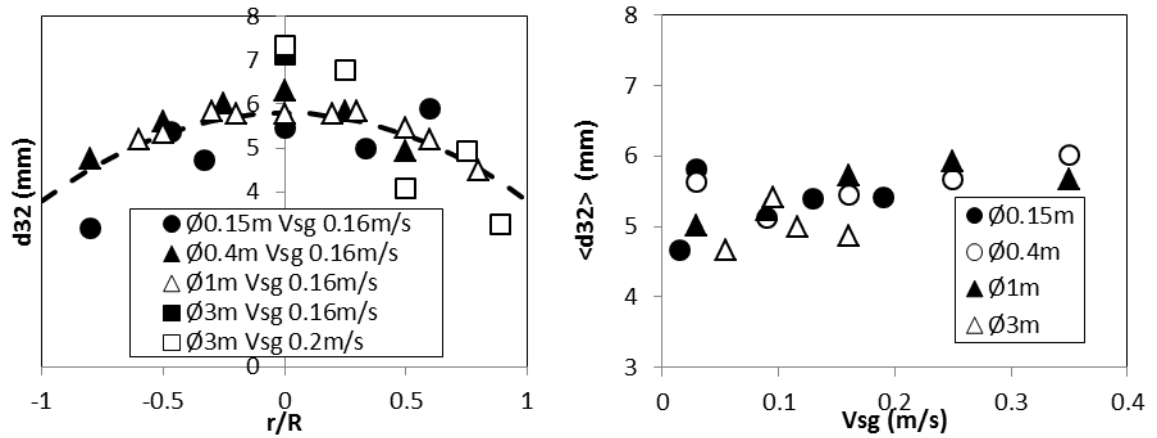
445

446 4.3. Bubble diameters

447 Sauter mean diameters measured with the cross-correlation technique are reported in Figure 10. On
448 the left, bubble Sauter diameter (d_{32}) profiles are reported at a given V_{sg} . On the right, volume
449 average Sauter diameters $\langle d_{32} \rangle$ for the 4 columns are given as a function of V_{sg} . $\langle d_{32} \rangle$ is computed
450 as the gas-volume averaged Sauter diameter over column sections. Let us recall that, in the up-flow
451 region, the typical relative uncertainty on the Sauter diameter is less than 10% for the large
452 superficial velocities considered here (Maximiano Raimundo et al., 2016). Important results can be
453 summarized as follows:

- 454 • The mean bubble diameter increases slowly from $\approx 4.5 - 5$ mm at low V_{sg} to 5.5-6mm at high
455 V_{sg}
- 456 • The mean bubble diameter is almost insensitive to the column diameter,
- 457 • Bubble size profiles follow roughly a parabolic shape, as suggested by the parabolic function
458 $(y=5.8-2x^2)$ reported in Figure 10.
- 459 • Bigger bubbles are measured at the center of the column, and the bubble size decreases with
460 x typically by 1 to 2mm indicating that a small amount of spatial segregation between upflow
461 and downflow regions occurs in these flows.

462 In all our experiments, the terminal velocity of the average bubbles varies over a limited range (0.21-
463 0.23m/s in clean water), and the bubbles always pertain to the same regime i.e. ellipsoids at high
464 particulate Reynolds number (the latter ranges between 950 and 1300) experiencing wobbling (Clift
465 et al., 1978). Hence, the present database has been built-up for nearly the same mean bubble size
466 irrespective of the dimension of the bubble column and of the amount of gas injected.



467

468 **Figure 10: Sauter diameter profiles in the 4 columns (right) and volume average Sauter diameter for the 4 columns versus**
 469 **V_{sg} . ($H/D=2.5$ except in the 3m column where $H/D=2$).**

470

471 As the mean bubble size does not exhibit strong variations for V_{sg} in the range of 0.03 to 0.35 m/s,
 472 this quantity does not explain by itself the transition between the homogeneous and the
 473 heterogeneous regime. Further investigation based on bubble size distributions may be required to
 474 identify a presumed link between coalescence phenomena and flow regime transition as suggested
 475 and reviewed by Besagni and Inzoli (2017b).

476

477 4.4. Discussion

478 Bubble size measurements suggest information concerning turbulence. Classical population balance
 479 kernels (Buffo et al., 2013) use the dissipation rate (ϵ) as a major parameter to predict breakage and
 480 coalescence. ϵ can classically be written as $\sim u'^3/\Lambda$, Λ being a turbulence macroscale. Besides, the
 481 global dissipation rate inside bubble columns can be estimated via different ways, but it is generally
 482 considered as not depending on the column diameter (Deckwer, 1992; Roels and Heijnen, 1980).
 483 Indeed in bubble columns ϵ is usually calculated as $g \cdot (\rho_L - \rho_G)/\rho_L \cdot V_{sg}$. The observed almost
 484 constant bubble size with scale-up is consistent with this theory. In the present study u' has been
 485 found to follow $\sim D^{0.4}$, this implies that Λ increases during scale up as $\sim D^{1.2}$, let say $\sim D$, which makes
 486 sense from a pure geometrical consideration. Further investigations concerning the size of
 487 turbulence macroscales in bubble columns may be useful to understand the link between flow

488 structures and turbulence production, as well as with breakage and coalescence phenomena.

489 Concerning velocity scales, the above discussion on the apparent relative velocity indicates that the

490 terminal velocity of bubbles is not the most relevant scale in these buoyancy driven bubbly flows.

491 Indeed, a natural velocity scale $(gD)^{0.5}$ arises from the analysis of the entrained liquid flow rate in the

492 central portion of the column. Such a scale is reminiscent of turbulent flows driven by convection. In

493 buoyancy driven flows such as in thermal convection, the equilibrium between flow convection and

494 Archimedian forcing leads to a velocity scaling as $V^2 \approx gL\Delta\rho/\rho$, where the density gradient $\Delta\rho$ is

495 evaluated at the length scale L . L is tentatively taken here as the column diameter. That choice is

496 supported here by the fact that we are considering the fully developed region, and, as the flow

497 happens to be self-similar in that zone, the height of the column is no longer a relevant parameter.

498 In addition to these large-scale gradients, localised variations in the density of the gas-liquid mixture,

499 equivalent then to variations in the local gas concentration, can also arise. Indeed, dispersed two-

500 phase flows can be prone to the formation of clusters, and Voronoï tessellations that are an efficient

501 way to detect and characterise such clusters (Monchaux et al., 2012) have been exploited here.

502 In the present experiments, we exploited the signals delivered by optical probes that provide the

503 arrival time of bubbles. From such a time history, it is straightforward to define successive Voronoï

504 time intervals ΔT_{bubble} containing a single bubble centred into it. Figure 11 provides the statistics

505 gathered on the axis of the 1m I.D. column at $V_{\text{sg}} = 25\text{cm/s}$: the data correspond to a record of 600

506 seconds during which 85000 bubbles have been detected. The abscissa in Figure 11 is the time

507 interval ΔT_{bubble} normalised by the mean value $\langle \Delta T_{\text{bubble}} \rangle$. The quantity $\Delta T_{\text{bubble}} / \langle \Delta T_{\text{bubble}} \rangle$ represents

508 the inverse of the ratio of the local and instantaneous gas concentration to the average gas hold-up.

509 Clearly, the measured probability density function differs from that of a random Poisson process

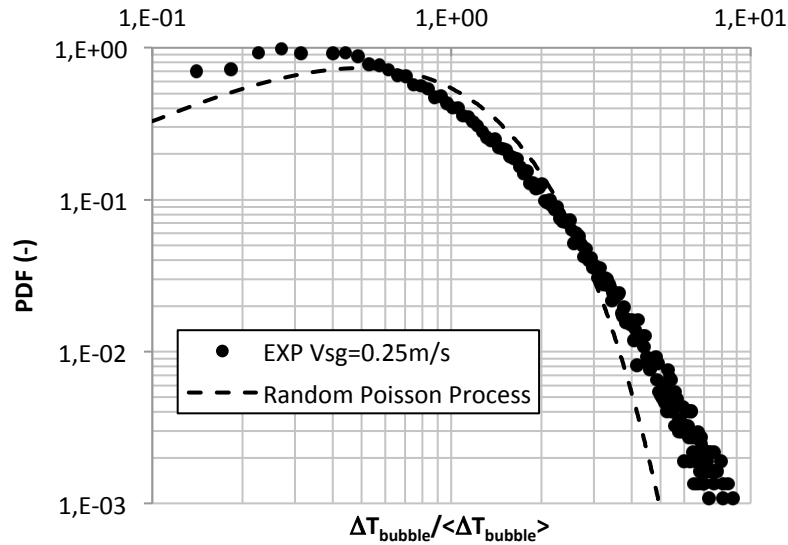
510 referred to as RPP in the sequel (Ferenc and Néda, 2007). In particular, the probability to found small

511 time intervals ΔT_{bubble} , below $0.6 \langle \Delta T_{\text{bubble}} \rangle$, is higher than in RPP indicating that clusters (i.e. regions

512 where bubbles accumulate) are more probable than in a random process. Similarly, the probability to

513 found large time intervals ΔT_{bubble} , above $4 \langle \Delta T_{\text{bubble}} \rangle$, is higher than in RPP indicating that voids (i.e.

514 regions with few bubbles) are also more probable than in RPP. This demonstrates that clusters and
515 voids are indeed present in the heterogeneous regime (this behaviour holds for others columns size
516 and superficial velocities). Moreover, that plot indicates that local, instantaneous concentrations
517 evolve between 0.1 and 10 times the average gas holdup. Local, instantaneous concentrations
518 varying over a so wide range imply quite strong fluctuations in density and thus in buoyancy.
519 Therefore, one expects strong local velocity differences between dense and dilute regions. The latter
520 can contribute to enhance the apparent relative velocity between phases as the bubbles are mostly
521 located in clusters while the liquid is mostly present in voids. This may explain the large apparent
522 relative velocity detected since the bubble terminal velocity in a still fluid is no longer the relevant
523 scale. Such structures are also prone to contribute to turbulence production as they produce strong
524 local and instantaneous shear rates. The velocity fluctuations shown in Figure 9 combined with the
525 correlation (13) support that statement. Finally, if one sticks to the mean values of bubble sizes, the
526 transition between homogeneous and heterogeneous regimes observed in the present experiments
527 is not related with coalescence; instead, convective instabilities that lead to the formation of dense
528 and dilute regions are believed to be responsible for that transition. To pursue along these lines, it
529 would be worthwhile to characterise these clusters and voids in terms of gas holdup and size
530 distributions and to examine how they evolve with flow conditions and column diameter. Another
531 key issue is how to quantitatively relate these clusters and voids with the apparent relative motion
532 between phases and with the turbulence production in the liquid phase. In particular it would be
533 relevant to examine the connections between clusters and void dimensions and the correlation
534 length scale Λ discussed above.



535

536 **Figure 11: Voronoï diagram deduced from an optical probe signal collected in the centre of the 1m diameter column**
 537 **operated at Vsg =25cm/s (dots). The dashed line represents the statistics of a Random Poisson Process.**

538

539 5. Conclusions

540 The objective of this work was to study specifically the scale-up of bubble columns in the
 541 heterogeneous regime using similar fluids and the same measurement techniques at very different
 542 scales, which has never been reported before in literature. The wide range of column diameter, from
 543 0.15 to 3m and of superficial gas velocities, from 0.05m/s to 0.35m/s, involved in this study makes
 544 the reported data an important material for validation of Population Balance and CFD models and for
 545 up-scaling issues. Correlations concerning gas holdup, mean liquid velocity and turbulence have been
 546 validated over a wide range of geometry and operating conditions. In particular, the self-similarity of
 547 the flow structure in terms of void fraction, liquid mean as well as fluctuating velocities have been
 548 demonstrated in the heterogeneous regime. This is believed to hold while the aspect ratio of the
 549 bubble column remains large enough so that end effects no longer affect the flow organisation in the
 550 central portion of the column. That self-similarity leads to an entrained liquid flow rate proportional
 551 to $D^2(gD)^{1/2}$, meaning that the entrainment capability of a bubble column is only set by its size and
 552 does not depend on the injected gas superficial velocity. This result also demonstrates that the
 553 velocity scale $(gD)^{1/2}$ is a key descriptor for bubble column scale-up. Also, both the average and the

554 fluctuations of the liquid velocity are very sensitive to the scale-up. Small dependencies to the
555 column diameter have been found on local and global gas holdup. The above results hold for all
556 columns and flow conditions pertaining to the heterogeneous regime, and for almost the same
557 average Sauter bubble diameters. The presence of strong concentration gradients has also been
558 demonstrated, and the impact of these clusters and voids on the flow characteristics and on
559 turbulence production deserves to be analysed further. As the turbulent dissipation rate plays an
560 important role on the BSD, the turbulence induced in the heterogeneous regime needs to be better
561 characterised in particular by determining its integral length scale.

562

563 **Abbreviations**

564	d	distance between probes (m or mm)
565	d_{32}	Local mean Sauter diameter (m or mm)
566	$\langle d_{32} \rangle$	volume averaged mean Sauter diameter (m or mm)
567	$d_{32,h}$	Local mean Sauter horizontal diameter (m or mm)
568	CC	cross-correlation
569	D	Column diameter (m)
570	ecc	bubble eccentricity
571	H	height from gas distributor (m)
572	H_0	static (non aerated) liquid height (m)
573	H_D	aerated liquid height (m)
574	Q_G	gas flow rate (m^3/s)
575	$Q_{G,core}$	gas flow rate in the core region(m^3/s)
576	$Q_{L,up}$	Upward liquid flow rate in the core region (m^3/s)
577	r	Radial position (m)
578	R	radius of the column (m)
579	S_c	Section of the core region (m^2)

580 $U(x,t)$ Liquid axial instantaneous velocity (m/s)
581 $U(x,t)$ Liquid axial average velocity (m/s)
582 U_0 Liquid axial velocity at the center of the column (m/s)
583 U_r Relative velocity between gas and liquid (m/s)
584 u' RMS of liquid axial velocity fluctuation (m/s)
585 V_{sg} Superficial gas velocity (m/s)
586 x normalized radial position ($=2r/D$)

587 *Symbols*

588 α_G Local gas holdup
589 $\langle \alpha_G \rangle$ Average gas holdup
590 ε Turbulence dissipation rate (w/kg)
591 Λ Turbulence macroscale (m)
592 ρ Liquid density (kg/m^3)

593

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