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Corresponding Author:	Simona Scollo, PhD Istituto Nazionale di Geofisica e Vulcanologia, Osservatorio Etneo 95123, ITALY	
Corresponding Author Secondary Information:		
Order of Authors:	Simona Scollo, PhD Costanza Bonadonna Irene Manzella	
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Abstract:	<p>Downward propagating instabilities are often observed at the bottom of volcanic plumes and clouds. These instabilities generate fingers that enhance the sedimentation of fine ash. Despite their potential influence on tephra dispersal and deposition, their dynamics is not entirely understood, undermining the accuracy of volcanic ash transport and dispersal models. Here, we present new laboratory experiments that investigate the effects of particle size, composition and concentration on finger generation and dynamics. The experimental set-up consists of a Plexiglas tank equipped with a removable plastic sheet that separates two different layers. The lower layer is a solution of water and sugar, initially denser than the upper layer, which consists of water and particles. Particles in the experiments include glass beads as well as andesitic, rhyolitic, and basaltic volcanic ash. During the experiments, we removed the horizontal plastic sheet separating the two fluids. Particles were illuminated with a laser and filmed with a HD camera; the Particle Image Velocimetry (PIV) is used to analyse finger dynamics. Results show that both the number and the downward advance speed of fingers increase with particle concentration in the upper layer, while finger speed increases with particle size but is independent of particle composition. An increase in particle concentration and turbulence is estimated to take place inside the fingers, which could promote aggregation in subaerial fallout events. Finally, finger number, finger speed and particle concentration were observed to decrease with time after the formation of fingers. A similar pattern could occur in volcanic clouds when the mass supply from the eruptive vent is reduced. Observed evolution of the experiments through time also indicates that there must be a threshold of fine ash concentration and mass eruption rate below which fingers do not form; this is also confirmed by field observations.</p>	
Response to Reviewers:	<p>Dear Executive Editor,</p> <p>We have accepted all your suggestions indicated on your annotated paper and made the final changes indicated on your annotated paper.</p> <p>Thanking for your co-operation,</p> <p>Best Regards, Simona Scollo, On the behalf of all authors</p>	

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1 **Settling-driven gravitational instabilities associated with volcanic clouds:**
2 **new insights from experimental investigations**

3
4 Simona Scollo¹, Costanza Bonadonna², Irene Manzella²

5 Istituto Nazionale di Geofisica e Vulcanologia, Osservatorio Etneo, Sezione di Catania, Italy

6 Département des sciences de la Terre, Université de Genève, Suisse

7
8 **Abstract**

9
10 Downward propagating instabilities are often observed at the bottom of volcanic plumes and clouds.
11 These instabilities generate fingers that enhance the sedimentation of fine ash. Despite their
12 potential influence on tephra dispersal and deposition, their dynamics is not entirely understood,
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26 concentration were observed to decrease with time after the formation of fingers. A similar pattern

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28 evolution of the experiments through time also indicates that there must be a threshold of fine ash
29 concentration and mass eruption rate below which fingers do not form; this is also confirmed by
30 field observations.

31 *Key words:* Tephra; Volcanic Plumes; Volcanic Ash; Laboratory Experiments; PIV Analysis;
32 Particle Aggregation.

33

34 1. INTRODUCTION

35 During explosive volcanic eruptions, a large number of volcanic particles is injected into the
36 atmosphere with the potential of generating significant hazards to nearby communities and various
37 economic sectors. While fine ash in distal area may produce long-term health risks and is extremely
38 dangerous for aircraft jet engines due to the accumulation of melted glass particles and erosion of
39 turbine blades, proximal fallout can cause collapse of buildings and damage to agriculture,
40 vegetation, lifelines, road networks and critical infrastructures (e.g. Blong, 2000; Miller and
41 Casadevall, 2000). The volcanic crisis of Eyafjallajökull volcano (2010, Iceland) and Cordón Caulle
42 volcano (2011, Chile), represent the most recent examples of widespread economic disruption
43 caused by volcanic ash (e.g. Alexander, 2013; Oxford, 2010; Elissondo et al., 2016; Sammons et al.,
44 2010; Wilson et al., 2013). Volcanic risk can be mitigated thanks to accurate forecasting of tephra
45 dispersal that builds on a good understanding and description of volcanic plumes and cloud
46 dynamics and sedimentation. There is a variety of volcanic ash dispersal models based on different
47 assumptions and modelling strategies (see Folch et al. (2012) for a review). Sensitivity analyses
48 have demonstrated that, when eruption source parameters are well constrained, eruptive phenomena
49 such as particle dispersal and sedimentation can be reproduced with good accuracy (e.g. Costa et
50 al., 2006; Scollo et al., 2010; Bonadonna et al., 2012). However, even after an accurate model
51 calibration, differences between field data and model results can reach up to 150% (e.g. Scollo et
52 al., 2008). Causes of these discrepancies include the fact that not all the physical processes of
53 volcanic plumes and clouds are fully described. Among these processes, the generation and
54 dynamics of fingers associated with settling-driven gravitational instabilities, also called convective
55 instabilities, could play an important role (e.g. Carazzo and Jellinek, 2013; Durant, 2015; Manzella
56 et al., 2015; Figure 1).

57 In the presence of particle-laden fluids, such as volcanic plumes and clouds, gravitational
58 instabilities are induced by particle settling across the density interface (Hoyal et al., 1999).
59 Initially the configuration is gravitationally stable: the lighter particle laden fluid (e.g. volcanic

60 current) is emplaced above the denser one (e.g. atmospheric layer). Small variations in density at
61 different points of the interface occur due to particle settling, generating instabilities. As a
62 consequence, vertical gravity currents, called fingers, start to develop in the lower layer and lead to
63 convective motion (Turner, 1979), which drives the vertical transport of particles in the lower layer
64 (e.g. Carazzo and Jellinek, 2012; Hoyal et al., 1999; Manzella et al., 2015). The main condition for
65 the formation of settling-driven gravitational instabilities is that the particle suspension behaves as a
66 continuum and this happens if the finger downward velocity has to be greater than particle settling
67 velocity (Hoyal et al., 1999). This condition (e.g. the particles to be coupled with the fluid and
68 efficiently mixed) is satisfied, according to Carazzo and Jellinek (2012), when both the Stokes
69 number ($St = \frac{1}{f} \frac{\rho_p}{18} \frac{d_p^2 V}{\mu L}$) and the Sedimentation number ($\Sigma = \frac{1}{f} \frac{\rho_p}{18} \frac{d_p^2 g}{\mu V}$) are <1 , where f is the drag
70 factor, ρ_p the particle density (kg m^{-3}), d_p the particle diameter (m), μ the dynamic viscosity (Pa s),
71 V (m s^{-1}) and L (m) the characteristic velocity and length for the flow, and g the acceleration due to
72 gravity (m s^{-2}).

73 Gravitational instabilities have often been observed in many volcanic plumes and clouds, such as
74 those associated with the eruption of Mount St. Helens 1980 (USA), Montserrat 1997 (West Indies),
75 Eyafjallajökull 2010 (Iceland), Ruapehu 1996 (New Zealand) and Etna 2013 (Italy) (Bonadonna et
76 al., 2002; Bonadonna et al., 2005a; Bonadonna et al. 2005b; Bonadonna et al., 2011; Durant et al.,
77 2009; Manzella et al., 2015; Schultz et al., 2006) (e.g. Figure 1). In recent decades, several authors
78 have used laboratory experiments to study the effects of gravitational instabilities on tephra
79 sedimentation. For example, Carey (1997) examined the settling behaviour of volcanic ash (20-180
80 μm diameter) onto a water surface based on an experimental apparatus. The experimental set-up
81 consisted of a 1.5-m-high settling column positioned over a 30 cm x 30 cm x 70 cm glass tank filled
82 with water. Particles fell at a constant rate at the top of the column, accumulated on the water
83 surface and then descended into the tank where they were photographed. Carey (1997) observed
84 that the formation of fingers is directly linked to the reduction of particle settling velocity at the air-

85 water interface, which increases the concentration at the boundary layer. He also proved that the
86 particle settling in the water column was accelerated by the formation of diffuse vertical gravity
87 currents driven by gravitational instabilities that reduced the residence time of fine ash. Carazzo and
88 Jellinek (2012) studied gravitational instabilities in volcanic plumes through both laboratory
89 experiments and theoretical considerations. They found that finger formation reduced the residence
90 time of fine ash into the gravitationally unstable particle boundary layer of volcanic clouds.
91 Gravitational instabilities could therefore explain the unusual patterns of some tephra deposits (e.g.
92 Bonadonna et al., 2002; Bonadonna et al., 2005) and/or the premature sedimentation of fine ash that
93 are often explained by particle aggregation (e.g. Carey and Sigurdsson, 1982).

94 Models of sedimentation associated with gravitation instabilities were developed by Hoyal et al.
95 (1999), recently modified by Manzella et al. (2015). Their formulation builds on the mass balance
96 equation between the incoming and outgoing flux at the density interface for two different
97 conditions: an upper quiescent layer (i.e. no external forcing of the fluid motion) and an upper
98 turbulent layer (i.e. external forcing of the fluid motion). Furthermore, Cardoso and Zarrebini
99 (2001) analysed buoyant particle-laden flows both experimentally and theoretically. They found
100 that the development of particle-rich fingers was related to unstable particle stratification and that
101 both the concentration of particles at the source of the plume, as well as the size of the particles, had
102 notable influence on the sedimentation pattern in the environment below the surface current.
103 Recently, Manzella et al. (2015) analysed gravitational instabilities during the 2010 eruption of
104 Eyjafjallajökull volcano (Iceland) that transported fine ash to the ground at a speed of ~1 m/s,
105 various orders of magnitude faster than the predicted terminal fall velocities of the smallest
106 observed particles. These results were confirmed by specific laboratory experiments using glass
107 beads in a density-stratified aqueous solution. They also showed how particle aggregation was
108 strongly linked with sedimentation driven by fingers. The relationship between particle aggregation
109 and gravitational instabilities was also suggested by Carazzo and Jellinek (2012).

110 Particle Imaging Velocimetry (PIV) is a widely used technique in fluid dynamics. It enables
111 measuring the velocity of a fluid through the tracking of several particles able to reflect the light of
112 a laser sheet (e.g. Adrian, 1991; Adrian, 1995; Adrian, 2005; Grant, 1997; Raffel et al., 2007). In
113 volcanology, the PIV technique has already been applied to characterize plume dynamics, to
114 measure the flow velocity for various ranges of particle size, overpressure ratios and densities and
115 to analyse the effect of collision kinetic energy and atmospheric water vapour in subsaturated
116 condition on ash aggregation (e.g. Saffaraval et al., 2012, Telling and Dufek, 2012, Chojnicki et al.,
117 2014, Chojnicki et al., 2015a, Chojnicki et al., 2015b).

118 In our experiments, particles generating fingers are embedded in the fluid and, thanks to their
119 potential for reflecting laser light, are used as PIV trackers. Experiments are carried out and
120 analysed by the PIV technique (Section 2) in order to investigate the influence of particle size,
121 composition and concentration on the formation and dynamics of the fingers (Section 3).
122 Experimental results are then discussed and compared with observations of fingers occurring during
123 explosive volcanic eruptions (Section 4).

124

125 **2. METHODS**

126 *2.1 Experimental setup*

127 Our experimental set-up is described in details by Manzella et al. (2015). The set-up comprises a
128 Plexiglas tank of 30.3 cm x 50 cm x 7.5 cm (with x corresponding to the length (L), y to the height
129 (H) and z to the width (W)) equipped with a removable sheet for the partition of two separate layers
130 (Figure 2). The upper partition ($H_1 = 13.5$ cm), which is filled with water and particles, is
131 characterized by an initial lower density than the lower partition ($H_2 = 25.1$ cm) that consists of a
132 solution of water and sugar. The lower layer density was fixed with a value of 1008.4 kg/m^3 , while
133 variations in the density in the upper layer depend on the concentration and on the different
134 densities of particles and, in our experiments, range between 999.8 and 1001.5 kg/m^3 (see Manzella
135 et al. (2015) as GSA data repository for the formulation). The experiments are carried out under

136 isothermal conditions and the configuration is suitable to represent the state in which the plume and
137 fingers are advected at wind speed and the dynamic conditions are similar to those in volcanic
138 clouds (Manzella et al., 2015). The experiments entail removing the horizontal barrier that separates
139 the two fluids, and then observing the instabilities formed at the boundary of the two layers
140 propagating downward (Manzella et al., 2015). Similarly, we consider two different set-ups defined
141 as unmixed and mixed conditions. During unmixed experiments, particles are fully suspended
142 before the beginning of the experiment but they do not undergo additional external stirring once the
143 experiment starts, while, during mixed experiments, particles are continuously mixed with a rotary
144 stirrer that is stopped 1-2 seconds before the removal of the separation between the two layers (see
145 also Figure DR4 of the repository material of Manzella et al. (2015)). The stirrer is set at a speed of
146 30 rpm with a paddle of 6.9 cm (length) by 3.4 cm (diameter).

147 Experiments were carried out to examine the fluid dynamics associated with finger formation
148 using a PIV measuring system and image analysis. To this end, we recorded the experiments with a
149 high speed/high definition camera while a 2 Watt Neodymium-doped YAG (Yttrium Aluminium
150 Garnet) laser (RayPower 2000 by Dantec Dynamics), located at about 1 m from the frontal tank
151 wall, generates a green light to illuminate the particles used as tracer for the PIV analysis (Figure 2).
152 We were then able to measure the number and speed of fingers by image analysis and, therefore, to
153 assess the effect of concentration, size and particle composition on their generation and dynamics.

154 *2.2 Experimental conditions*

155 Particles used in our experiments include 4 grain-size classes of Glass Beads (GB), i.e. with
156 diameter $< 32 \mu\text{m}$, between $32\text{-}45 \mu\text{m}$, between $45\text{-}63 \mu\text{m}$, and between $63\text{-}90 \mu\text{m}$ as well as 6
157 grain-size classes of Andesitic, Rhyolitic, and Basaltic Volcanic Ash (Andes-VA; Rhyol-VA;
158 Basalt-VA, respectively), i.e. with diameter $< 32 \mu\text{m}$, $32\text{-}45 \mu\text{m}$, $45\text{-}63 \mu\text{m}$, $63\text{-}90 \mu\text{m}$, $90\text{-}125 \mu\text{m}$,
159 and $125\text{-}180 \mu\text{m}$ (Table 1). An additional class was also considered, where we mixed all the
160 particles with diameter $<125 \mu\text{m}$ (called $<125 \mu\text{m}$ in Table 1) to study the effect of using a widely
161 polydisperse mixture. Populations of different particle size were obtained through mechanical

162 sieving at the University of Geneva and the above-mentioned grain-size classes have been named
163 after the sieves used which follow the international standard classification ISO3310. Laser
164 diffraction analyses carried out with the CILAS 1180 instrument on selected samples indicate a
165 good sorting inside the considered ranges (Folk and Ward, 1957).

166 For the andesitic composition, we used samples of volcanic ash erupted during the 2010 eruption
167 of Eyjafjallajökull volcano (Iceland). This eruption produced a continuous volcanic plume up to 10
168 km above sea level between 14 April and 21 May 2010 (Gudmundsson et al., 2012). Glass
169 composition ranges from benmoreite to trachyte with a silica content between 56 and 68 wt% and a
170 total alkali from 7.3 and 9.1wt% (Cioni et al., 2014). The ash considered in our experiments was
171 sampled between 4 and 8 May 2010 (Bonadonna et al., 2011). For the rhyolitic composition, we
172 used samples of volcanic ash erupted during the May 2008 eruption of Chaitén volcano (Chile)
173 (Alfano et al., 2011; Alfano et al., 2012; Alfano et al., 2016). The bulk magma composition of the
174 main phase of this eruption varies between 73.0 and 75.5 wt% of SiO₂ (Alfano et al., 2011). Finally,
175 for the basaltic composition, we used samples of volcanic ash from the 1992 eruption of Cerro
176 Negro (Nicaragua) that lasted for about 21 days and was associated with a volcanic plume up to
177 about 7 km a.s.l. (e.g. Connor and Connor, 2006; Connor et al, 1993). The silica content varies
178 between 48.64 and 52.15 wt% (Roggensack et al., 1997). The density of individual size classes is
179 complex to determine; however, based on the detailed analysis of Eychenne and Le Penneç (2012),
180 we can assume that the density of fine ash is close to their Dense Rock Equivalent (DRE) value.
181 The mean DRE value (measured with a helium pycnometer) of Eyjafjallajökull, Chaitén and Cerro
182 Negro ash is 2738 kg m⁻³ (Bonadonna et al. 2011), 2240 kg m⁻³ (Alfano et al. 2012) and 2988 kg
183 m⁻³ (measured for this work), respectively.

184 Three concentrations were considered to generate fingers: $C1 = 3$ g/l, $C2 = 4$ g/l, $C3 = 5$ g/l. The
185 concentrations were chosen based on experimental constraints as reported in Manzella et al. (2015).
186 In fact, both lower and higher concentrations are not detectable experimentally based on the grey
187 scale measuring strategy. The particle volumetric concentration in the experiments is then in the

188 order of 10^{-9} , which, together with their capacity to reflect the laser light, confirms that the particles
189 used can be exploited as tracers in PIV analysis. This is also supported by the fact that they are
190 coupled with the fluid since both their Stokes and Sedimentation numbers are < 1 (i.e. they are in
191 the range of 10^{-4} - 10^{-1} and 0.5-1, respectively) for the grain-size and particle composition analysed in
192 our experiments (Carazzo and Jellinek, 2012). A detailed list of experiments is reported in Table 1.
193 In unmixed conditions, different tests were carried using GB with different concentrations
194 (experiment 1-10), GB with different sizes (experiments 11-17), Andes-VA with different sizes
195 (experiments 18-24) and Andes-VA with different concentrations (experiments 25-26). Experiments
196 with mixed conditions were carried out only for GB and Andes-VA in the range between 45 and 63
197 μm (experiments 28-29) and for Andes-VA with diameter $< 125 \mu\text{m}$ (experiments 27 and 30).
198 However, the experiment 27 did not provide a good PIV analysis and was not considered for further
199 analysis. Experiments were finally carried out for Rhyol-VA and Basalt-VA with all the grain size
200 classes considered (experiments 31-34) in unmixed conditions. Experiments with GB were repeated
201 up to three times to verify the repeatability of the measurements (e.g. experiments 12-13, 14-15 and
202 16-17), while experiments with volcanic ash were carried out only once because of the limited
203 amount of material available. It is worth mentioning that GB particles are more visible than
204 volcanic ash particles, reflecting the laser light more efficiently. Among volcanic particles, the
205 Andes-VA ones reflect the laser light best and therefore, once the effect of the composition has
206 been studied, these were preferred for tests with volcanic ash.

207 *2.3 Data analysis*

208 Images of 1624 x 1600 pixel sizes were taken at of 0.03 s time steps and each particle captured by
209 the camera that scattered the laser light was used for the PIV analysis with the Dynamic Studio
210 Software (DANTEC, http://www.cefd-imech.ac.vn/lab/3D_PIV/DynamicStudio%20Manual.pdf).
211 PIV technique is based on the fact that the image intensity field at each instant corresponds to the
212 position of the particles reflecting the laser light and it assumes that between two instants t and $t +$
213 Δt , i.e. two consecutive exposures to the laser light, all particles inside a previously defined

214 interrogation window have moved together with the fluid with the same displacement vector, ΔX .
 215 This interrogation area (IA) should be small enough to respect this assumption but large enough to
 216 contain at least 10 particles reflecting the laser light to evaluate the intensity field. On the other
 217 hand, the particle volume fraction should be smaller than 10^{-4} , so that they are easily visible and do
 218 not influence the fluid flow. For this reason, in order to find the most suitable interrogation window
 219 and thus increase the accuracy of the analysis, the Dynamic Studio Software uses an iterative
 220 process that reduces the size of the interrogation area progressively from 128x128 to 16x16 pixels.
 221 In this framework, using a spatially statistical cross-correlation function which relates the difference
 222 in image intensity between two instants, we are able to evaluate the displacement of the fluid and
 223 the velocity vector for each interrogation area (Raffel et al., 2007). In addition, the DANTEC
 224 software is also used to evaluate the finger speed (m/s) measuring the position of the finger front at
 225 different times, the divergence (s^{-1}) and the vorticity (s^{-1}) fields.

226 As also described in the Dynamic Studio software manual, the divergence of a 3D vector
 227 velocity field \bar{U} is defined as:

$$228 \quad \text{div}(\bar{U}) = \frac{\partial U}{\partial x} + \frac{\partial V}{\partial y} + \frac{\partial W}{\partial z}. \quad (1)$$

229 For planar data gradients, as the one analysed with PIV, it reduces to:

$$230 \quad \text{div}(\bar{UV}) = \frac{\partial U}{\partial x} + \frac{\partial V}{\partial y} \quad (2)$$

231 with x , y and z indicating the axis associated with the length, height, and width in Figure 2.

232 Non-zero divergence values could indicate local changes in density and, therefore, local changes of
 233 particles concentration or, when the fluid is incompressible, a non-negligible variation of the
 234 velocity in the z direction.

235 Vorticity is a vector quantity, which corresponds to the rotation of the fluids. For planar data
 236 gradient only the z -component of vorticity, ω_z , can be calculated:

$$237 \quad \omega_z = \frac{\partial V}{\partial x} - \frac{\partial U}{\partial y}. \quad (3)$$

238 According to Tritton (1988), turbulent flows are characterised by non-zero, fluctuating vorticity.
239 Even if the studied gravitational instabilities cannot be considered two-dimensional because they
240 have a component in the out of plane direction ($z = W$ in Figure 2), this component is significantly
241 smaller than the longitudinal ($x = L$ in Figure 2) and vertical ($y = H$ in Figure 2) dimensions of the
242 tank, i.e. a few millimetres versus tens of centimetres. In addition, a single finger is mostly axi-
243 symmetrical with respect to the flow direction, so we can assume that what we observe in a single
244 finger in the x - y plane would be similar to what we could observe in the y - z plane. As a result, we
245 consider that the PIV two-dimensional analysis can globally capture the main flow dynamics
246 involved in the gravitational instabilities.

247

248 **3. RESULTS**

249 Figure 3 shows selected images of the experiments using GB and Andes-VA particles between 32
250 and 45 μm and between 63 and 90 μm with C1 as concentration and in unmixed condition
251 (experiments 11, 15, 20 and 22 in Table 1). Fingers are clearly visible a few seconds ($> 3\text{s}$) after the
252 sheet is removed and are of approximately the same size in each experiment.

253 Our analysis shows that the descending fingers have an irregular shape during the formation
254 stage, and descend with large caps at their tips (e.g. Figure 3). The number of fingers increases with
255 particle concentration (Figure 4a), but does not depend on the particle size and composition (Figure
256 4b). The mean wavelength, given by L/n where L is the length of the box (30.3 cm) and n is the
257 number of fingers, ranges between 2.2 and 2.9 cm. Finger speed has a poor dependence on the
258 particle concentration in the range of concentration investigated here (Figure 4c) and increases with
259 particle size (Figure 4d). The experimental error bars in Figures 4a and 4b was evaluated by the
260 standard deviation obtained from the mean value of finger number in ten images taken 3 s after
261 removing the sheet. The mean and standard deviation in Figures 4c and 4d are instead evaluated by
262 the analysis of five fingers over the course of one whole experiment.

263 It is worth mentioning that particles greater than 125 μm did not generate fingers (experiment 18
264 in Table 1). The number of fingers also depends on the experimental conditions of the upper layer.
265 Figures 5a and 5b illustrate experiments with a “mixed” upper layer using GB and Andes-VA
266 between 45 and 63 μm (experiments 28 and 29 in Table 1) showing a higher number of fingers (2-3
267 fingers more for both cases) with respect to “unmixed” experiments. Finger speed, instead, is
268 independent of initial mixing.

269 We also analysed the evolution of finger dynamics with time. In general, the downward
270 movement of fingers was not steady and we observed similar oscillations to those reported in
271 Carazzo and Jellinek (2012). For the same class size, we found a general decrease of finger number
272 with time mainly at the interface due to the decrease of the particle concentration in the upper layer.
273 As an example, the experiment 11 in Table 1 (i.e. GB with size between 32-45 μm) shows 11 and 8
274 fingers, respectively, 5 and 30 s after the sheet was removed (Figures 6a and b). The speed of
275 fingers is about 4.5 ± 1.1 mm/s and 2.4 ± 0.2 mm/s after about 7 and 20 s, respectively. This
276 behaviour was similar for different class sizes (e.g. Figures 6c and 6d). Figure 7 shows the variation
277 of the finger number with respect to time for GB between 45 and 63 μm (experiment 3). At the
278 beginning of the experiment, the number of fingers is 11 ± 1 and after about 30 s the number of
279 fingers remains almost constant (between 7 ± 1 and 6 ± 1). A power law fits the evolution with time
280 well ($R^2=0.88$; Figure 7). We found that the finger speed was 4.6 ± 1.1 , 3.0 ± 1.0 and 2.4 ± 1.3
281 mm/s at 10 s, at 20 s and 30 s, respectively. The study of finger evolution with time was also carried
282 out for Andes-VA particles (experiments 19, 29, and 30) and for volcanic ash with a wide range of
283 size ($< 125 \mu\text{m}$) showing similar trends. As an example, Figure 8 shows the experiment 24 in Table
284 1 carried out with Andes-VA. At the beginning of the experiment (< 10 s), fingers contain particles
285 with the widest size range. In fact, even though particle size cannot be quantitatively assessed with
286 PIV, a qualitative assessment can be made based on backscattering because larger particles show a
287 smaller backscattering of the laser light and are less visible in the images retrieved by the camera

288 than smaller particles. In agreement with our previous observations, this experiment also shows
289 how both the number and speed of fingers decrease with time. In particular, we observe 12 and 11
290 fingers after 5 and 10 s and 9 and 8 fingers after 15 sec and 20 s, respectively. The associated speed
291 is 7.9 ± 1.2 , 6.1 ± 1.0 , 4.6 ± 1.7 and 4.3 ± 0.8 mm/s after 5, 10, 15 and 20 s, respectively.

292 Finally, we investigated the divergence and vorticity fields for GB and Andes-VA with different
293 particle size and, in general, we found that divergence was zero everywhere except at the interface
294 and inside the fingers. Figure 9 shows how the highest variation of the relative values of divergence
295 and vorticity are concentrated in the fingers, where we also notice an increase of the brightness
296 coming from the laser reflection of the particles (Figure 9a). An increase in brightness is hence
297 associated with an increasing number of reflecting particles. As aforementioned, a non-zero
298 divergence for PIV planar data could be associated with a significant velocity component along the
299 out of plane direction (width in Figure 2), which cannot be excluded considering the 3D nature of
300 fingers. However, the occurrence of the highest variations in divergence, even if small, combined
301 with an increase of brightness within the fingers (Figures 9a and 9b), could suggest a temporary and
302 localized variation in particle concentration. By contrast, a fluctuation of vorticity values in the x - y
303 plane can be associated with a turbulence motion regardless of the 3D nature of the fingers. We can
304 then infer that gravitational instabilities could be likely associated with an increase of concentration
305 and turbulence with respect to initial conditions.

306

307 **4. DISCUSSION**

308 Our experiments confirm previous findings that gravitational instabilities have a marked effect on
309 the sedimentation of volcanic ash (e.g. Carazzo and Jellinek, 2012; Manzella et al., 2015) and the
310 analysis of these instabilities provide new insights into the effect of particle concentration, size and
311 composition and into the evolution of fingers with time. Based on the analysis of divergence and
312 vorticity, the formation of gravitational instabilities also has implications on particle aggregation.

313 Results show how the number of fingers depends largely on particle concentration in the upper
314 layer, as already concluded by Hoyal et al. (1999), while finger speed mostly depends on particle
315 size. Even though basaltic particles were more difficult to analyze than silicic particles due to their
316 dark colour that does not reflect the laser light well, particle composition seems to play a negligible
317 role on finger dynamics (e.g. Figure 4).

318 We have also shown how the number of fingers decreases with a drop in particle concentration
319 in the upper layer of the tank. Hence, there must be a critical value of particle concentration below
320 which fingers cannot form. In this sense, our experimental observations suggest that only volcanic
321 clouds characterized by a relatively high mass load of particles, and, therefore, volcanic eruptions
322 associated with a large Mass Eruption Rate (MER), are likely to form gravitational instabilities. As
323 an example, gravitational instabilities were clearly observed during the 23rd November 2013
324 explosive event of Etna volcano (Italy) that was characterized by a MER of about 10^5 kg/s
325 (Andronico et al., 2015) (see Figure 1a). The eruptive plume of the 4th May 2010 Eyjafjallajökull
326 eruption (Iceland) (Manzella et al., 2015) and of the 17th June 1996 eruption of Ruapehu volcano
327 (New Zealand) (Bonadonna et al., 2005), which generated well-developed gravitational instabilities,
328 were also characterized by a MER of about 10^5 kg/s (e.g. Degruyter and Bonadonna, 2013; Ripepe
329 et al., 2011; Bonadonna et al., 2005) (see Figures 1b and 1c). Gravitational instabilities were also
330 observed at the bottom of volcanic clouds associated with the thermal plumes of the August-
331 October 1997 Vulcanian explosions of Montserrat volcano, which injected an average of about 10^8 -
332 10^9 kg of tephra into the atmosphere in just a few seconds (Bonadonna et al., 2002) (Figure 1d). By
333 contrast, gravitational instabilities were not observed during the 2011 and 2012 explosive events of
334 Etna volcano that were characterized by a lower MER ($\sim 10^4$ kg/s) (Andronico et al., 2014).

335 Another fundamental condition for the formation of gravitational instabilities in volcanic clouds
336 is the presence of fine ash. Indeed, gravitational instabilities only formed in our experiments in
337 association with the sedimentation of particles < 125 μm . Our results match those reported by Carey
338 (2007), although he used a different set-up and different concentration, but similar particle

339 composition (i.e. 1991 Pinatubo dacitic tephra) and fluid (i.e. water). In addition, both sets of
340 experiments were performed in water, and, therefore, the critical cut-off size of 125 μm could be
341 even smaller in air (Carazzo and Jellinek, 2012). All previously mentioned eruptions (i.e. Etna 23
342 November 2013, Eyjafjallajökull 2010, Montserrat 1997 and Ruapehu 1996) were characterized by
343 the presence of particles $< 125 \mu\text{m}$, i.e. $\sim 6\text{wt}\%$ for Ruapehu 1996, $\sim 40\text{wt}\%$ for Eyjafjallajökull
344 2010 and $\sim 50\text{-}80\text{wt}\%$ for the August-October 1997 Vulcanian explosions of Montserrat
345 (Bonadonna et al., 2002; Bonadonna and Houghton 2005; Bonadonna et al., 2011). In addition,
346 volcanic lightning that is typically associated with particle-laden jet and abundance of fine particles
347 (Cimarelli et al., 2013), was observed during the 23 November 2013 Etna eruption.

348 Theory reported in Manzella et al. (2015) shows how an increase in particle concentration in the
349 upper layer would increase g' , and, therefore, the finger speed. In fact, finger velocity is equal to:
350 $v_f = g'^{\frac{2}{5}}(v_p \frac{1}{4} \pi \delta^2)^{1/5}$, where v_p is the particle settling velocity, δ is the Particle Boundary Layer
351 (PBL) thickness, g' is the reduced gravity of the PBL given by $g' = g \frac{\rho_{PBL} - \rho_a}{\rho_a}$, where g is the
352 gravity and ρ_{PBL} and ρ_a are the density of PBL and of the atmosphere, respectively. As a result, the
353 higher the particle concentration in volcanic plumes, the higher the finger speed. Although we
354 investigated a small range, a slight increase of the finger speed with concentration is also confirmed
355 by our experiments and by theory (e.g. Hoyal et al., 1999). This aspect should however be explored
356 further by enlarging the concentration range. Moreover, the theory of Hoyal et al. (1999) can
357 explain the observed increase of particle speed with particle size and the negligible effect of particle
358 composition on finger dynamics. First, the larger the particle size, the larger the particle velocity v_p ,
359 and, therefore, the finger velocity v_f . Second, the main difference among the particle composition is
360 the variation in density that, however, is negligible for particles $< 125 \mu\text{m}$ (e.g. Bonadonna and
361 Phillips, 2003; Eychenne and Le Pennec, 2012). In fact, the difference in DRE values is only
362 between 2240 kg/m^3 for Chaitén rhyolitic ash and 2988 kg/m^3 for Cerro Negro basaltic ash.
363 Furthermore, volcanic clouds represent a polydisperse mixture containing a wide range of particle

364 sizes even at medial-distal locations. Based on our results and theory, we expect that i) the number
365 of fingers decreases with the distance from the vent and ii) the finger speed decreases with the
366 distance from the vent for a combined effect of the reduction of both volcanic ash concentration and
367 particle size in the upper layer.

368 Aggregation of ash particles into clusters with a higher terminal fall velocity leads to a reduction
369 of the atmospheric lifetime (e.g. Brown et al., 2012; Costa et al., 2006; Durant, 2015). It has already
370 been suggested that aggregation could be enhanced inside the fingers due to the high particle
371 concentration (e.g. Carazzo and Jellinek, 2012). Our analysis of divergence and vorticity provides
372 additional evidence that indicates the potential role of gravitational instabilities in forming particle
373 aggregates. Fluctuations of divergence and vorticity inside the fingers could represent an increase of
374 particle concentration and turbulence with respect to the surrounding regions, which increases the
375 probability of collisions, and, therefore, promotes aggregation. Nonetheless, the formation of ash
376 clusters within volcanic plumes and clouds and the sedimentation of single ash clusters
377 independently of gravitational instabilities cannot be excluded, in particular when the cluster
378 settling velocity is higher than the finger settling velocity (e.g. Manzella et al., 2015).

379 Finally, our results clearly show that the highest values of finger number and finger speed are
380 associated with their initial formation and both decrease with time. This could be related to the
381 decrease of particle concentration in the upper layer, which in our experiment progressively
382 decreases with time. However, during an eruption, volcanic plumes are continuously fed at the
383 eruptive vent and the decrease in finger number and speed could only occur at the end of the
384 explosive activity. This also supports the idea that particle aggregation may be more efficient at the
385 beginning of finger formation when the concentration is the highest.

386

387 **5. CONCLUSIONS**

388 A comprehensive physical characterization of the sedimentation processes occurring in volcanic
389 plumes and clouds relies on a better understanding of the gravitational instabilities. We carried out

390 experiments using a Plexiglas tank of 50 cm x 30.3 cm x 7.5 cm equipped with a horizontal
391 removable plastic pet sheet to separate the two layers. The upper layer was made up of water and
392 particles, while the lower layer was a solution of water and sugar that was initially denser than the
393 upper layer. After removing the horizontal plastic pet sheet, particles were illuminated with a laser
394 and filmed with a HD camera and analysed by PIV. Our experimental investigations provide new
395 insights into the mechanisms characterizing finger formation and finger dynamics. In particular:

- 396 1) Number of fingers and finger speed increase with particle concentration in the upper layer;
397 this is in agreement with previous experimental observations (e.g., Carazzo and Jellinek,
398 2012; Manzella et al., 2015) and supports field observations, where fingers have been
399 observed only in volcanic plumes with relatively high MER (i.e. $MER \geq 10^5$ kg/s);
- 400 2) Gravitational instabilities were observed only with particles $< 125 \mu\text{m}$; this also concurs with
401 previous experimental observations (i.e. Carey 2007; Carazzo and Jellinek, 2012) and
402 confirms the idea that a relative abundance of fine ash is necessary to generate fingers.
403 However, the size cut-off in air could be smaller due to different buoyancy;
- 404 3) Number of fingers and finger speed are independent of particle composition, suggesting that
405 finger formation can occur independently of magma composition;
- 406 4) The relation between gravitational instabilities and particle aggregation was explored based
407 on the analysis of divergence and vorticity inside the fingers. These values suggest
408 heterogeneity in particle concentration and an increase in turbulent motion that need further
409 exploration with experiments including the analysis of the 3D component. Given that a high
410 concentration of particles $< 125 \mu\text{m}$ and turbulence are both factors promoting aggregation,
411 we can conclude that particle aggregation could easily occur both at the base of the cloud
412 where fingers form and inside fingers.

413

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423

424 **Figures and Table Captions**

425 **Figure 1.** Gravitational instabilities associated with:

426 a) 23 November 2013 lava fountain of Etna volcano, Italy (photo by
427 http://www.tboeckel.de/EFSF/efsf_etna/Etna2013/Etna_11_13/volcano_etna_11_2013_e.htm); b)
428 Eyjafjallajökull plume on 4 May 2010, Iceland (photo by C. Bonadonna); c) 17 June 1996 eruption
429 of Ruapehu volcano, New Zealand (photo from
430 <http://www.natgeocreative.com/photography/1302290>); d) Vulcanian explosion in September 1997,
431 Montserrat (photo adjusted from Bonadonna et al. (2002)).

432 **Figure 2.** Experimental set-up comprising a Plexiglas tank of 30.3 cm x 50 cm x 7.5 cm (x
433 corresponding to the length (L) direction, y to the height (H), z to the width (W)), a removable pet
434 sheet, a laser and a HD camera. Particles, HD Camera, and laser instrument are not to scale. The
435 stirrer used in our mixed experiments is shown in Figure DR4 of the repository material of
436 Manzella et al. (2015).

437 **Figure 3.** Images of experiments with: a) GB with diameter between 32-45 μm (experiment 11 in
438 Table 1); b) GB with diameter between 63-90 μm (experiment 15 in Table 1); c) Andes-VA with
439 diameter between 32-45 μm (experiment 22 in Table 1); and d) Andes-VA with diameter between
440 63-90 μm (experiment 20 in Table 1). Images (26 cm x 16.5 cm) are taken about 10 seconds after
441 removing the horizontal pet sheet separating the two fluids in unmixed conditions.

442 **Figure 4.** Plots showing the number of fingers with respect to: a) particle concentration in the upper
443 layer (g/l) for GB and Andes-VA and b) particle size (μm) for GB, Andes-VA, Rhyol-VA and
444 Basalt-VA; and the speed of fingers with respect to: c) particle concentration in the upper layer (g/l)
445 for GB, Andes-VA, and Theory (Hoyal et al., 1999) and d) particle size for GB and Andes-VA
446 (μm) (experiments 19 to 26 and 31 to 43 in Table 1).

447 **Figure 5.** Images (26 cm x 16.5 cm) showing the number of fingers for the experiments with a
448 mixed upper layer using a) GB between 45 and 63 μm (experiment 28 in Table 1) and b) Andes-VA
449 between 45 and 63 μm (experiment 29 in Table 1).

450 **Figure 6.** Images (26 cm x 16.5 cm) of experiments with GB of diameter between 32-45 μm
451 (experiment 11 in Table 1) taken a) 5 s and b) 30 s and of diameter between 45-63 μm (experiment
452 3 in Table 1) taken c) 5 s and d) 30 s after removing the horizontal pet sheet. Arrows indicate
453 observed fingers.

454 **Figure 7.** Plot showing the evolution of the finger number with time for GB between 45-63 μm
455 (experiment 3 in Table 1).

456 **Figure 8.** Images (26 cm x 16.5 cm) of experiment 24 in Table 1 that includes Andes-VA with a
457 wide range of particle sizes ($< 125 \mu\text{m}$) imaged about a) 5 s, b) 10 s, c) 15 s, and d) 20 s after
458 removing the horizontal pet sheet.

459 **Figure 9.** Image of the experiment 7 (Table 1) for GB between 45-63 μm a) after 15 s the finger
460 formation; b) divergence (s^{-1}) and c) vorticity (s^{-1}) fields and d) divergence and vorticity values
461 measured along the orange line across the finger in c).

462

463 **Table 1.** Summary of experiments: experiment number; particle composition including glass beads
464 (GB), andesitic volcanic ash (Andes-VA), rhyolitic volcanic ash (Rhyol-VA), and basaltic volcanic
465 ash (Basalt-VA); particle size (S) in μm ; Concentration in g/l; the upper layer was both quiescent
466 (i.e. unmixed experiments, or continually mixed using a rotary stirrer (i.e. mixed experiments))
467 described in Manzella et al. (2015).

468

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1 **Settling-driven gravitational instabilities associated with volcanic clouds:**
2 **new insights from experimental investigations**

3
4 Simona Scollo¹, Costanza Bonadonna², Irene Manzella²

5 Istituto Nazionale di Geofisica e Vulcanologia, Osservatorio Etneo, Sezione di Catania, Italy

6 Département des sciences de la Terre, Université de Genève, Suisse

7
8 **Abstract**

9
10 **Downward propagating instabilities** are often observed at the bottom of volcanic plumes and clouds.
11 These instabilities generate fingers that enhance the sedimentation of fine ash. **Despite** their
12 potential influence on tephra dispersal and deposition, their dynamics is not entirely understood,
13 undermining the accuracy of volcanic ash transport and dispersal models. Here, we present new
14 laboratory experiments that investigate the effects of particle size, composition and concentration
15 on finger generation and dynamics. The experimental set-up consists of a Plexiglas tank equipped
16 with a removable **plastic** sheet **that separates two different** layers. The lower **layer** is a solution of
17 water and sugar, initially denser than the upper **layer, which consists of** water and particles.
18 **Particles in the experiments** include glass beads as well as andesitic, rhyolitic, and basaltic volcanic
19 ash. During the experiments, we removed the horizontal **plastic sheet** separating the two fluids.
20 Particles were illuminated with a laser and filmed with a HD camera; the Particle Image
21 Velocimetry (PIV) is used to analyse finger dynamics. Results show that both the number and the
22 **downward advance speed** of fingers increase with particle concentration in the upper layer, while
23 finger speed increases with particle size but is independent of particle composition. An increase in
24 particle concentration and turbulence is estimated to take place inside the fingers, which could
25 promote **aggregation in subaerial fallout events**. Finally, finger number, finger speed and particle
26 concentration were observed to decrease with time after the formation of fingers. A similar pattern

27 could occur in volcanic clouds **when** the mass supply from the eruptive vent is reduced. **Observed**
28 **evolution** of the experiments **through** time also indicates that there must be a threshold of fine ash
29 concentration and mass eruption rate below which fingers do not form; **this is** also confirmed by
30 field observations.

31 *Key words:* Tephra; Volcanic Plumes; Volcanic Ash; Laboratory Experiments; PIV Analysis;
32 Particle Aggregation.

33

34 1. INTRODUCTION

35 During explosive volcanic eruptions, a large **number** of volcanic particles is injected into the
36 atmosphere with the potential of generating significant hazards to nearby communities and various
37 economic sectors. While fine ash **in distal area** may produce long-term health risks and is extremely
38 dangerous for aircraft jet engines due to the accumulation of melted glass particles and erosion of
39 turbine blades, **proximal fallout** can cause collapse of buildings and damage to agriculture,
40 vegetation, lifelines, road networks and critical infrastructures (e.g. Blong, 2000; Miller and
41 Casadevall, 2000). The volcanic crisis of Eyafjallajökull volcano (2010, Iceland) and Cordón Caulle
42 volcano (2011, Chile), represent the most recent examples of widespread economic disruption
43 caused by volcanic ash (e.g. Alexander, 2013; Oxford, 2010; Elissondo et al., 2016; Sammons et al.,
44 2010; Wilson et al., 2013). Volcanic risk can be mitigated thanks to accurate forecasting of tephra
45 dispersal that builds on a good understanding and description of volcanic plumes and cloud
46 dynamics and sedimentation. There is a variety of volcanic ash dispersal models based on different
47 assumptions and modelling strategies (see Folch et al. (2012) for a review). Sensitivity analyses
48 have demonstrated that, when eruption source parameters are well constrained, eruptive phenomena
49 such as particle dispersal and sedimentation can be reproduced with good accuracy (e.g. Costa et
50 al., 2006; Scollo et al., 2010; Bonadonna et al., 2012). However, even after an accurate model
51 calibration, differences between field data and model results can reach up to 150% (e.g. Scollo et
52 al., 2008). Causes of these discrepancies include the fact that not all the physical processes of
53 volcanic plumes and clouds are fully described. Among these processes, the generation and
54 dynamics of fingers associated with settling-driven gravitational instabilities, also called convective
55 instabilities, could play an important role (e.g. Carazzo and Jellinek, 2013; Durant, 2015; Manzella
56 et al., 2015; Figure 1).

57 In the presence of particle-laden fluids, such as volcanic plumes and clouds, gravitational
58 instabilities are induced by particle settling across the density interface (Hoyal et al., 1999).
59 Initially the configuration is gravitationally stable: the lighter particle laden fluid (e.g. volcanic

60 current) is emplaced above the denser one (e.g. atmospheric layer). Small variations in density at
 61 different points of the interface occur due to particle settling, generating instabilities. As a
 62 consequence, vertical gravity currents, called fingers, start to develop in the lower layer and lead to
 63 convective motion (Turner, 1979), which drives the vertical transport of particles in the lower layer
 64 (e.g. Carazzo and Jellinek, 2012; Hoyal et al., 1999; Manzella et al., 2015). The main condition for
 65 the formation of settling-driven gravitational instabilities is that the particle suspension behaves as a
 66 continuum and this happens if the finger downward velocity has to be greater than particle settling
 67 velocity (Hoyal et al., 1999). This condition (e.g. the particles to be coupled with the fluid and
 68 efficiently mixed) is satisfied, according to Carazzo and Jellinek (2012), when both the Stokes
 69 number ($St = \frac{1}{f} \frac{\rho_p}{18 \mu} \frac{d_p^2 V}{L}$) and the Sedimentation number ($\Sigma = \frac{1}{f} \frac{\rho_p}{18 \mu} \frac{d_p^2 g}{V}$) are <1 , where f is the drag
 70 factor, ρ_p the particle density (kg m^{-3}), d_p the particle diameter (m), μ the dynamic viscosity (Pa s),
 71 V (m s^{-1}) and L (m) the characteristic velocity and length for the flow, and g the acceleration due to
 72 gravity (m s^{-2}).
 73 Gravitational instabilities have often been observed in many volcanic plumes and clouds, such as
 74 those associated with the eruption of Mount St. Helens 1980 (USA), Montserrat 1997 (West Indies),
 75 Eyafjallajökull 2010 (Iceland), Ruapehu 1996 (New Zealand) and Etna 2013 (Italy) (Bonadonna et
 76 al., 2002; Bonadonna et al., 2005a; Bonadonna et al. 2005b; Bonadonna et al., 2011; Durant et al.,
 77 2009; Manzella et al., 2015; Schultz et al., 2006) (e.g. Figure 1). In recent decades, several authors
 78 have used laboratory experiments to study the effects of gravitational instabilities on tephra
 79 sedimentation. For example, Carey (1997) examined the settling behaviour of volcanic ash (20-180
 80 μm diameter) onto a water surface based on an experimental apparatus. The experimental set-up
 81 consisted of a 1.5-m-high settling column positioned over a 30 cm x 30 cm x 70 cm glass tank filled
 82 with water. Particles fell at a constant rate at the top of the column, accumulated on the water
 83 surface and then descended into the tank where they were photographed. Carey (1997) observed
 84 that the formation of fingers is directly linked to the reduction of particle settling velocity at the air-

85 water interface, which increases the concentration at the boundary layer. He also proved that the
86 particle settling in the water column was accelerated by the formation of diffuse vertical gravity
87 currents driven by gravitational instabilities that reduced the residence time of fine ash. Carazzo and
88 Jellinek (2012) studied gravitational instabilities in volcanic plumes through both laboratory
89 experiments and theoretical considerations. They found that finger formation reduced the residence
90 time of fine ash into the gravitationally unstable particle boundary layer of volcanic clouds.
91 Gravitational instabilities could therefore explain the unusual patterns of some tephra deposits (e.g.
92 Bonadonna et al., 2002; Bonadonna et al., 2005) and/or the premature sedimentation of fine ash that
93 are often explained by particle aggregation (e.g. Carey and Sigurdsson, 1982).

94 Models of sedimentation associated with gravitation instabilities were developed by Hoyal et al.
95 (1999), recently modified by Manzella et al. (2015). Their formulation builds on the mass balance
96 equation between the incoming and outgoing flux at the density interface for two different
97 conditions: an upper quiescent layer (i.e. no external forcing of the fluid motion) and an upper
98 turbulent layer (i.e. external forcing of the fluid motion). Furthermore, Cardoso and Zarrebini
99 (2001) analysed buoyant particle-laden flows both experimentally and theoretically. They found
100 that the development of particle-rich fingers was related to unstable particle stratification and that
101 both the concentration of particles at the source of the plume, as well as the size of the particles, had
102 notable influence on the sedimentation pattern in the environment below the surface current.
103 Recently, Manzella et al. (2015) analysed gravitational instabilities during the 2010 eruption of
104 Eyjafjallajökull volcano (Iceland) that transported fine ash to the ground at a speed of ~1 m/s,
105 various orders of magnitude faster than the predicted terminal fall velocities of the smallest
106 observed particles. These results were confirmed by specific laboratory experiments using glass
107 beads in a density-stratified aqueous solution. They also showed how particle aggregation was
108 strongly linked with sedimentation driven by fingers. The relationship between particle aggregation
109 and gravitational instabilities was also suggested by Carazzo and Jellinek (2012).

110 Particle Imaging Velocimetry (PIV) is a widely used technique in fluid dynamics. It enables
111 measuring the velocity of a fluid through the tracking of several particles able to reflect the light of
112 a laser sheet (e.g. Adrian, 1991; Adrian, 1995; Adrian, 2005; Grant, 1997; Raffel et al., 2007). In
113 volcanology, the PIV technique has already been applied to characterize plume dynamics, to
114 measure the flow velocity for various ranges of particle size, overpressure ratios and densities and
115 to analyse the effect of collision kinetic energy and atmospheric water vapour in subsaturated
116 condition on ash aggregation (e.g. Saffaraval et al., 2012, Telling and Dufek, 2012, Chojnicki et al.,
117 2014, Chojnicki et al., 2015a, Chojnicki et al., 2015b).

118 In our experiments, particles generating fingers are embedded in the fluid and, thanks to their
119 potential for reflecting laser light, are used as PIV trackers. Experiments are carried out and
120 analysed by the PIV technique (Section 2) in order to investigate the influence of particle size,
121 composition and concentration on the formation and dynamics of the fingers (Section 3).
122 Experimental results are then discussed and compared with observations of fingers occurring during
123 explosive volcanic eruptions (Section 4).

124

125 **2. METHODS**

126 *2.1 Experimental setup*

127 Our experimental set-up is described in details by Manzella et al. (2015). The set-up comprises a
128 Plexiglas tank of 30.3 cm x 50 cm x 7.5 cm (with x corresponding to the length (L), y to the height
129 (H) and z to the width (W)) equipped with a removable sheet for the partition of two separate layers
130 (Figure 2). The upper partition ($H_1 = 13.5$ cm), which is filled with water and particles, is
131 characterized by an initial lower density than the lower partition ($H_2 = 25.1$ cm) that consists of a
132 solution of water and sugar. The lower layer density was fixed with a value of 1008.4 kg/m³, while
133 variations in the density in the upper layer depend on the concentration and on the different
134 densities of particles and, in our experiments, range between 999.8 and 1001.5 kg/m³ (see Manzella
135 et al. (2015) as GSA data repository for the formulation). The experiments are carried out under

136 isothermal conditions and the configuration is suitable to represent the state in which the plume and
137 fingers are advected at wind speed and the dynamic conditions are similar to those in volcanic
138 clouds (Manzella et al., 2015). The experiments entail removing the horizontal barrier that separates
139 the two fluids, and then observing the instabilities formed at the boundary of the two layers
140 propagating downward (Manzella et al., 2015). Similarly, we consider two different set-ups defined
141 as unmixed and mixed conditions. During unmixed experiments, particles are fully suspended
142 before the beginning of the experiment but they do not undergo additional external stirring once the
143 experiment starts, while, during mixed experiments, particles are continuously mixed with a rotary
144 stirrer that is stopped 1-2 seconds before the removal of the separation between the two layers (see
145 also Figure DR4 of the repository material of Manzella et al. (2015)). The stirrer is set at a speed of
146 30 rpm with a paddle of 6.9 cm (length) by 3.4 cm (diameter).

147 Experiments were carried out to examine the fluid dynamics associated with finger formation
148 using a PIV measuring system and image analysis. To this end, we recorded the experiments with a
149 high speed/high definition camera while a 2 Watt Neodymium-doped YAG (Yttrium Aluminium
150 Garnet) laser (RayPower 2000 by Dantec Dynamics), located at about 1 m from the frontal tank
151 wall, generates a green light to illuminate the particles used as tracer for the PIV analysis (Figure 2).
152 We were then able to measure the number and speed of fingers by image analysis and, therefore, to
153 assess the effect of concentration, size and particle composition on their generation and dynamics.

154 *2.2 Experimental conditions*

155 Particles used in our experiments include 4 grain-size classes of Glass Beads (GB), i.e. with
156 diameter $< 32 \mu\text{m}$, between $32\text{-}45 \mu\text{m}$, between $45\text{-}63 \mu\text{m}$, and between $63\text{-}90 \mu\text{m}$ as well as 6
157 grain-size classes of Andesitic, Rhyolitic, and Basaltic Volcanic Ash (Andes-VA; Rhyol-VA;
158 Basalt-VA, respectively), i.e. with diameter $< 32 \mu\text{m}$, $32\text{-}45 \mu\text{m}$, $45\text{-}63 \mu\text{m}$, $63\text{-}90 \mu\text{m}$, $90\text{-}125 \mu\text{m}$,
159 and $125\text{-}180 \mu\text{m}$ (Table 1). An additional class was also considered, where we mixed all the
160 particles with diameter $<125 \mu\text{m}$ (called $<125 \mu\text{m}$ in Table 1) to study the effect of using a widely
161 polydisperse mixture. Populations of different particle size were obtained through mechanical

162 sieving at the University of Geneva and the above-mentioned grain-size classes have been named
163 after the sieves used which follow the international standard classification ISO3310. Laser
164 diffraction analyses carried out with the CILAS 1180 instrument on selected samples indicate a
165 good sorting inside the considered ranges (Folk and Ward, 1957).

166 For the andesitic composition, we used samples of volcanic ash erupted during the 2010 eruption
167 of Eyjafjallajökull volcano (Iceland). This eruption produced a continuous volcanic plume up to 10
168 km above sea level between 14 April and 21 May 2010 (Gudmundsson et al., 2012). Glass
169 composition ranges from benmoreite to trachyte with a silica content between 56 and 68 wt% and a
170 total alkali from 7.3 and 9.1wt% (Cioni et al., 2014). The ash considered in our experiments was
171 sampled between 4 and 8 May 2010 (Bonadonna et al., 2011). For the rhyolitic composition, we
172 used samples of volcanic ash erupted during the May 2008 eruption of Chaitén volcano (Chile)
173 (Alfano et al., 2011; Alfano et al., 2012; Alfano et al., 2016). The bulk magma composition of the
174 main phase of this eruption varies between 73.0 and 75.5 wt% of SiO₂ (Alfano et al., 2011). Finally,
175 for the basaltic composition, we used samples of volcanic ash from the 1992 eruption of Cerro
176 Negro (Nicaragua) that lasted for about 21 days and was associated with a volcanic plume up to
177 about 7 km a.s.l. (e.g. Connor and Connor, 2006; Connor et al, 1993). The silica content varies
178 between 48.64 and 52.15 wt% (Roggensack et al., 1997). The density of individual size classes is
179 complex to determine; however, based on the detailed analysis of Eychenne and Le Penneç (2012),
180 we can assume that the density of fine ash is close to their Dense Rock Equivalent (DRE) value.
181 The mean DRE value (measured with a helium pycnometer) of Eyjafjallajökull, Chaitén and Cerro
182 Negro ash is 2738 kg m⁻³ (Bonadonna et al. 2011), 2240 kg m⁻³ (Alfano et al. 2012) and 2988 kg
183 m⁻³ (measured for this work), respectively.

184 Three concentrations were considered to generate fingers: $C1 = 3$ g/l, $C2 = 4$ g/l, $C3 = 5$ g/l. The
185 concentrations were chosen based on experimental constraints as reported in Manzella et al. (2015).
186 In fact, both lower and higher concentrations are not detectable experimentally based on the grey
187 scale measuring strategy. The particle volumetric concentration in the experiments is then in the

188 order of 10^{-9} , which, together with their capacity to reflect the laser light, confirms that the particles
189 used can be exploited as tracers in PIV analysis. This is also supported by the fact that they are
190 coupled with the fluid since both their Stokes and Sedimentation numbers are < 1 (i.e. they are in
191 the range of 10^{-4} - 10^{-1} and 0.5-1, respectively) for the grain-size and particle composition analysed in
192 our experiments (Carazzo and Jellinek, 2012). A detailed list of experiments is reported in Table 1.
193 In unmixed conditions, different tests were carried using GB with different concentrations
194 (experiment 1-10), GB with different sizes (experiments 11-17), Andes-VA with different sizes
195 (experiments 18-24) and Andes-VA with different concentrations (experiments 25-26). Experiments
196 with mixed conditions were carried out only for GB and Andes-VA in the range between 45 and 63
197 μm (experiments 28-29) and for Andes-VA with diameter $< 125 \mu\text{m}$ (experiments 27 and 30).
198 However, the experiment 27 did not provide a good PIV analysis and was not considered for further
199 analysis. Experiments were finally carried out for Rhyol-VA and Basalt-VA with all the grain size
200 classes considered (experiments 31-34) in unmixed conditions. Experiments with GB were repeated
201 up to three times to verify the repeatability of the measurements (e.g. experiments 12-13, 14-15 and
202 16-17), while experiments with volcanic ash were carried out only once because of the limited
203 amount of material available. It is worth mentioning that GB particles are more visible than
204 volcanic ash particles, reflecting the laser light more efficiently. Among volcanic particles, the
205 Andes-VA ones reflect the laser light best and therefore, once the effect of the composition has
206 been studied, these were preferred for tests with volcanic ash.

207 *2.3 Data analysis*

208 Images of 1624 x 1600 pixel sizes were taken at of 0.03 s time steps and each particle captured by
209 the camera that scattered the laser light was used for the PIV analysis with the Dynamic Studio
210 Software (DANTEC, http://www.cefd-imech.ac.vn/lab/3D_PIV/DynamicStudio%20Manual.pdf).
211 PIV technique is based on the fact that the image intensity field at each instant corresponds to the
212 position of the particles reflecting the laser light and it assumes that between two instants t and $t +$
213 Δt , i.e. two consecutive exposures to the laser light, all particles inside a previously defined

214 interrogation window have moved together with the fluid with the same displacement vector, ΔX .
 215 This interrogation area (IA) should be small enough to respect this assumption but large enough to
 216 contain at least 10 particles reflecting the laser light to evaluate the intensity field. On the other
 217 hand, the particle volume fraction should be smaller than 10^{-4} , so that they are easily visible and do
 218 not influence the fluid flow. For this reason, in order to find the most suitable interrogation window
 219 and thus increase the accuracy of the analysis, the Dynamic Studio Software uses an iterative
 220 process that reduces the size of the interrogation area progressively from 128x128 to 16x16 pixels.
 221 In this framework, using a spatially statistical cross-correlation function which relates the difference
 222 in image intensity between two instants, we are able to evaluate the displacement of the fluid and
 223 the velocity vector for each interrogation area (Raffel et al., 2007). In addition, the DANTEC
 224 software is also used to evaluate the finger speed (m/s) measuring the position of the finger front at
 225 different times, the divergence (s^{-1}) and the vorticity (s^{-1}) fields.

226 As also described in the Dynamic Studio software manual, the divergence of a 3D vector
 227 velocity field \bar{U} is defined as:

$$228 \quad \text{div}(\bar{U}) = \frac{\partial U}{\partial x} + \frac{\partial V}{\partial y} + \frac{\partial W}{\partial z}. \quad (1)$$

229 For planar data gradients, as the one analysed with PIV, it reduces to:

$$230 \quad \text{div}(\overline{UV}) = \frac{\partial U}{\partial x} + \frac{\partial V}{\partial y} \quad (2)$$

231 with x , y and z indicating the axis associated with the length, height, and width in Figure 2.

232 Non-zero divergence values could indicate local changes in density and, therefore, local changes of
 233 particles concentration or, when the fluid is incompressible, a non-negligible variation of the
 234 velocity in the z direction.

235 Vorticity is a vector quantity, which corresponds to the rotation of the fluids. For planar data
 236 gradient only the z -component of vorticity, ω_z , can be calculated:

$$237 \quad \omega_z = \frac{\partial V}{\partial x} - \frac{\partial U}{\partial y}. \quad (3)$$

238 According to Tritton (1988), turbulent flows are characterised by non-zero, fluctuating vorticity.
239 Even if the studied gravitational instabilities cannot be considered two-dimensional because they
240 have a component in the out of plane direction ($z = W$ in Figure 2), this component is significantly
241 smaller than the longitudinal ($x = L$ in Figure 2) and vertical ($y = H$ in Figure 2) dimensions of the
242 tank, i.e. a few millimetres versus tens of centimetres. In addition, a single finger is mostly axi-
243 symmetrical with respect to the flow direction, so we can assume that what we observe in a single
244 finger in the x - y plane would be similar to what we could observe in the y - z plane. As a result, we
245 consider that the PIV two-dimensional analysis can globally capture the main flow dynamics
246 involved in the gravitational instabilities.

247

248 **3. RESULTS**

249 Figure 3 shows selected images of the experiments using GB and Andes-VA particles between 32
250 and 45 μm and between 63 and 90 μm with C1 as concentration and in unmixed condition
251 (experiments 11, 15, 20 and 22 in Table 1). Fingers are clearly visible a few seconds ($> 3\text{s}$) after the
252 sheet is removed and are of approximately the same size in each experiment.

253 Our analysis shows that the descending fingers have an irregular shape during the formation
254 stage, and descend with large caps at their tips (e.g. Figure 3). The number of fingers increases with
255 particle concentration (Figure 4a), but does not depend on the particle size and composition (Figure
256 4b). The mean wavelength, given by L/n where L is the length of the box (30.3 cm) and n is the
257 number of fingers, ranges between 2.2 and 2.9 cm. Finger speed has a poor dependence on the
258 particle concentration in the range of concentration investigated here (Figure 4c) and increases with
259 particle size (Figure 4d). The experimental error bars in Figures 4a and 4b was evaluated by the
260 standard deviation obtained from the mean value of finger number in ten images taken 3 s after
261 removing the sheet. The mean and standard deviation in Figures 4c and 4d are instead evaluated by
262 the analysis of five fingers over the course of one whole experiment.

263 It is worth mentioning that particles greater than 125 μm did not generate fingers (experiment 18
264 in Table 1). The number of fingers also depends on the experimental conditions of the upper layer.
265 Figures 5a and 5b illustrate experiments with a “mixed” upper layer using GB and Andes-VA
266 between 45 and 63 μm (experiments 28 and 29 in Table 1) showing a higher number of fingers (2-3
267 fingers more for both cases) with respect to “unmixed” experiments. Finger speed, instead, is
268 independent of initial mixing.

269 We also analysed the evolution of finger dynamics with time. In general, the downward
270 movement of fingers was not steady and we observed similar oscillations to those reported in
271 Carazzo and Jellinek (2012). For the same class size, we found a general decrease of finger number
272 with time mainly at the interface due to the decrease of the particle concentration in the upper layer.
273 As an example, the experiment 11 in Table 1 (i.e. GB with size between 32-45 μm) shows 11 and 8
274 fingers, respectively, 5 and 30 s after the sheet was removed (Figures 6a and b). The speed of
275 fingers is about 4.5 ± 1.1 mm/s and 2.4 ± 0.2 mm/s after about 7 and 20 s, respectively. This
276 behaviour was similar for different class sizes (e.g. Figures 6c and 6d). Figure 7 shows the variation
277 of the finger number with respect to time for GB between 45 and 63 μm (experiment 3). At the
278 beginning of the experiment, the number of fingers is 11 ± 1 and after about 30 s the number of
279 fingers remains almost constant (between 7 ± 1 and 6 ± 1). A power law fits the evolution with time
280 well ($R^2=0.88$; Figure 7). We found that the finger speed was 4.6 ± 1.1 , 3.0 ± 1.0 and 2.4 ± 1.3
281 mm/s at 10 s, at 20 s and 30 s, respectively. The study of finger evolution with time was also carried
282 out for Andes-VA particles (experiments 19, 29, and 30) and for volcanic ash with a wide range of
283 size ($< 125 \mu\text{m}$) showing similar trends. As an example, Figure 8 shows the experiment 24 in Table
284 1 carried out with Andes-VA. At the beginning of the experiment (< 10 s), fingers contain particles
285 with the widest size range. In fact, even though particle size cannot be quantitatively assessed with
286 PIV, a qualitative assessment can be made based on backscattering because larger particles show a
287 smaller backscattering of the laser light and are less visible in the images retrieved by the camera

288 than smaller particles. In agreement with our previous observations, this experiment also shows
289 how both the number and speed of fingers decrease with time. In particular, we observe 12 and 11
290 fingers after 5 and 10 s and 9 and 8 fingers after 15 sec and 20 s, respectively. The associated speed
291 is 7.9 ± 1.2 , 6.1 ± 1.0 , 4.6 ± 1.7 and 4.3 ± 0.8 mm/s after 5, 10, 15 and 20 s, respectively.

292 Finally, we investigated the divergence and vorticity fields for GB and Andes-VA with different
293 particle size and, in general, we found that divergence was zero everywhere except at the interface
294 and inside the fingers. Figure 9 shows how the highest variation of the relative values of divergence
295 and vorticity are concentrated in the fingers, where we also notice an increase of the brightness
296 coming from the laser reflection of the particles (Figure 9a). An increase in brightness is hence
297 associated with an increasing number of reflecting particles. As aforementioned, a non-zero
298 divergence for PIV planar data could be associated with a significant velocity component along the
299 out of plane direction (width in Figure 2), which cannot be excluded considering the 3D nature of
300 fingers. However, the occurrence of the highest variations in divergence, even if small, combined
301 with an increase of brightness within the fingers (Figures 9a and 9b), could suggest a temporary and
302 localized variation in particle concentration. By contrast, a fluctuation of vorticity values in the x - y
303 plane can be associated with a turbulence motion regardless of the 3D nature of the fingers. We can
304 then infer that gravitational instabilities could be likely associated with an increase of concentration
305 and turbulence with respect to initial conditions.

306

307 **4. DISCUSSION**

308 Our experiments confirm previous findings that gravitational instabilities have a marked effect on
309 the sedimentation of volcanic ash (e.g. Carazzo and Jellinek, 2012; Manzella et al., 2015) and the
310 analysis of these instabilities provide new insights into the effect of particle concentration, size and
311 composition and into the evolution of fingers with time. Based on the analysis of divergence and
312 vorticity, the formation of gravitational instabilities also has implications on particle aggregation.

313 Results show how the number of fingers depends largely on particle concentration in the upper
314 layer, as already concluded by Hoyal et al. (1999), while finger speed mostly depends on particle
315 size. Even though basaltic particles were more difficult to analyze than silicic particles due to their
316 dark colour that does not reflect the laser light well, particle composition seems to play a negligible
317 role on finger dynamics (e.g. Figure 4).

318 We have also shown how the number of fingers decreases with a drop in particle concentration
319 in the upper layer of the tank. Hence, there must be a critical value of particle concentration below
320 which fingers cannot form. In this sense, our experimental observations suggest that only volcanic
321 clouds characterized by a relatively high mass load of particles, and, therefore, volcanic eruptions
322 associated with a large Mass Eruption Rate (MER), are likely to form gravitational instabilities. As
323 an example, gravitational instabilities were clearly observed during the 23rd November 2013
324 explosive event of Etna volcano (Italy) that was characterized by a MER of about 10^5 kg/s
325 (Andronico et al., 2015) (see Figure 1a). The eruptive plume of the 4th May 2010 Eyjafjallajökull
326 eruption (Iceland) (Manzella et al., 2015) and of the 17th June 1996 eruption of Ruapehu volcano
327 (New Zealand) (Bonadonna et al., 2005), which generated well-developed gravitational instabilities,
328 were also characterized by a MER of about 10^5 kg/s (e.g. Degruyter and Bonadonna, 2013; Ripepe
329 et al., 2011; Bonadonna et al., 2005) (see Figures 1b and 1c). Gravitational instabilities were also
330 observed at the bottom of volcanic clouds associated with the thermal plumes of the August-
331 October 1997 Vulcanian explosions of Montserrat volcano, which injected an average of about 10^8 -
332 10^9 kg of tephra into the atmosphere in just a few seconds (Bonadonna et al., 2002) (Figure 1d). By
333 contrast, gravitational instabilities were not observed during the 2011 and 2012 explosive events of
334 Etna volcano that were characterized by a lower MER ($\sim 10^4$ kg/s) (Andronico et al., 2014).

335 Another fundamental condition for the formation of gravitational instabilities in volcanic clouds
336 is the presence of fine ash. Indeed, gravitational instabilities only formed in our experiments in
337 association with the sedimentation of particles <125 μm . Our results match those reported by Carey
338 (2007), although he used a different set-up and different concentration, but similar particle

339 composition (i.e. 1991 Pinatubo dacitic tephra) and fluid (i.e. water). In addition, both sets of
340 experiments were performed in water, and, therefore, the critical cut-off size of 125 μm could be
341 even smaller in air (Carazzo and Jellinek, 2012). All previously mentioned eruptions (i.e. Etna 23
342 November 2013, Eyjafjallajökull 2010, Montserrat 1997 and Ruapehu 1996) were characterized by
343 the presence of particles $< 125 \mu\text{m}$, i.e. $\sim 6\text{wt}\%$ for Ruapehu 1996, $\sim 40\text{wt}\%$ for Eyjafjallajökull
344 2010 and $\sim 50\text{-}80\text{wt}\%$ for the August-October 1997 Vulcanian explosions of Montserrat
345 (Bonadonna et al., 2002; Bonadonna and Houghton 2005; Bonadonna et al., 2011). In addition,
346 volcanic lightning that is typically associated with particle-laden jet and abundance of fine particles
347 (Cimarelli et al., 2013), was observed during the 23 November 2013 Etna eruption.

348 Theory reported in Manzella et al. (2015) shows how an increase in particle concentration in the
349 upper layer would increase g' , and, therefore, the finger speed. In fact, finger velocity is equal to:
350 $v_f = g'^{\frac{2}{5}}(v_p \frac{1}{4} \pi \delta^2)^{1/5}$, where v_p is the particle settling velocity, δ is the Particle Boundary Layer
351 (PBL) thickness, g' is the reduced gravity of the PBL given by $g' = g \frac{\rho_{PBL} - \rho_a}{\rho_a}$, where g is the
352 gravity and ρ_{PBL} and ρ_a are the density of PBL and of the atmosphere, respectively. As a result, the
353 higher the particle concentration in volcanic plumes, the higher the finger speed. Although we
354 investigated a small range, a slight increase of the finger speed with concentration is also confirmed
355 by our experiments and by theory (e.g. Hoyal et al., 1999). This aspect should however be explored
356 further by enlarging the concentration range. Moreover, the theory of Hoyal et al. (1999) can
357 explain the observed increase of particle speed with particle size and the negligible effect of particle
358 composition on finger dynamics. First, the larger the particle size, the larger the particle velocity v_p ,
359 and, therefore, the finger velocity v_f . Second, the main difference among the particle composition is
360 the variation in density that, however, is negligible for particles $< 125 \mu\text{m}$ (e.g. Bonadonna and
361 Phillips, 2003; Eychenne and Le Pennec, 2012). In fact, the difference in DRE values is only
362 between 2240 kg/m^3 for Chaitén rhyolitic ash and 2988 kg/m^3 for Cerro Negro basaltic ash.
363 Furthermore, volcanic clouds represent a polydisperse mixture containing a wide range of particle

364 sizes even at medial-distal locations. Based on our results and theory, we expect that i) the number
365 of fingers decreases with the distance from the vent and ii) the finger speed decreases with the
366 distance from the vent for a combined effect of the reduction of both volcanic ash concentration and
367 particle size in the upper layer.

368 Aggregation of ash particles into clusters with a higher terminal fall velocity leads to a reduction
369 of the atmospheric lifetime (e.g. Brown et al., 2012; Costa et al., 2006; Durant, 2015). It has already
370 been suggested that aggregation could be enhanced inside the fingers due to the high particle
371 concentration (e.g. Carazzo and Jellinek, 2012). Our analysis of divergence and vorticity provides
372 additional evidence that indicates the potential role of gravitational instabilities in forming particle
373 aggregates. **Fluctuations of divergence** and vorticity inside the fingers **could** represent an increase of
374 particle concentration and turbulence with respect to the surrounding regions, **which** increases the
375 probability of collisions, **and, therefore, promotes** aggregation. Nonetheless, the formation of ash
376 clusters within volcanic plumes and clouds and the sedimentation of single ash clusters
377 independently of gravitational instabilities cannot be excluded, in particular when the cluster
378 **settling** velocity is higher than the finger **settling** velocity (e.g. Manzella et al., 2015).

379 Finally, our results clearly show that the highest values of finger number and finger speed are
380 associated with their initial formation and both decrease with time. This could be related to the
381 decrease of particle concentration in the upper layer, **which in our experiment progressively**
382 decreases with time. However, during an eruption, volcanic plumes are continuously fed at the
383 eruptive vent and the decrease in finger number and speed could only occur at the end of the
384 explosive activity. This also supports the idea that particle aggregation may be more efficient at the
385 beginning of finger formation when the concentration is the highest.

386

387 **5. CONCLUSIONS**

388 A comprehensive physical characterization of the sedimentation processes occurring in volcanic
389 plumes and clouds relies on a better understanding of the gravitational instabilities. We carried out

390 experiments using a Plexiglas tank of 50 cm x 30.3 cm x 7.5 cm equipped with a horizontal
391 removable **plastic** pet sheet **to separate the two** layers. The upper layer was made up of water and
392 particles, while the lower layer was a solution of water and sugar that was initially denser than the
393 upper layer. After removing the horizontal **plastic** pet sheet, particles were illuminated with a laser
394 and filmed with a HD camera and analysed by PIV. Our experimental investigations provide new
395 insights into the mechanisms characterizing finger formation and **finger** dynamics. In particular:

- 396 1) **Number of fingers** and **finger** speed increase with particle concentration in the upper layer;
397 this is in agreement with previous experimental observations (e.g., Carazzo and Jellinek,
398 2012; Manzella et al., 2015) and supports field observations, where fingers have been
399 observed only in volcanic plumes with relatively high MER (i.e. $MER \geq 10^5$ kg/s);
- 400 2) Gravitational instabilities were observed only with particles $< 125 \mu\text{m}$; this also concurs with
401 previous experimental observations (i.e. Carey 2007; Carazzo and Jellinek, 2012) and
402 confirms the idea that a relative abundance of fine ash is necessary to generate fingers.
403 However, the size cut-off in air could be smaller due to different buoyancy;
- 404 3) **Number of fingers and finger speed** are independent of particle composition, suggesting that
405 finger formation can occur independently of **magma composition**;
- 406 4) The relation between gravitational instabilities and **particle aggregation** was explored based
407 on the analysis of divergence and vorticity inside the fingers. These values suggest
408 heterogeneity in particle concentration and an increase in turbulent motion that need further
409 exploration with experiments including the analysis of the 3D component. Given that a high
410 concentration of particles $< 125 \mu\text{m}$ and turbulence are both factors promoting aggregation,
411 we can conclude that particle aggregation could easily occur both at the base of the cloud
412 where fingers form and inside fingers.

413

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423

424 **Figures and Table Captions**

425 **Figure 1.** Gravitational instabilities associated with:

426 a) 23 November 2013 lava fountain of Etna volcano, Italy (photo by
427 http://www.tboeckel.de/EFSF/efsf_etna/Etna2013/Etna_11_13/volcano_etna_11_2013_e.htm); b)
428 Eyjafjallajökull plume on 4 May 2010, Iceland (photo by C. Bonadonna); c) 17 June 1996 eruption
429 of Ruapehu volcano, New Zealand (photo from
430 <http://www.natgeocreative.com/photography/1302290>); d) Vulcanian explosion in September 1997,
431 Montserrat (photo adjusted from Bonadonna et al. (2002)).

432 **Figure 2.** Experimental set-up comprising a Plexiglas tank of 30.3 cm x 50 cm x 7.5 cm (*x*
433 corresponding to the length (*L*) direction, *y* to the height (*H*), *z* to the width (*W*)), a removable pet
434 sheet, a laser and a HD camera. Particles, HD Camera, and laser instrument are not to scale. The
435 stirrer used in our mixed experiments is shown in Figure DR4 of the repository material of
436 Manzella et al. (2015).

437 **Figure 3.** Images of experiments with: a) GB with diameter between 32-45 μm (experiment 11 in
438 Table 1); b) GB with diameter between 63-90 μm (experiment 15 in Table 1); c) Andes-VA with
439 diameter between 32-45 μm (experiment 22 in Table 1); and d) Andes-VA with diameter between
440 63-90 μm (experiment 20 in Table 1). Images (26 cm x 16.5 cm) are taken about 10 seconds after
441 removing the horizontal pet sheet separating the two fluids in unmixed conditions.

442 **Figure 4.** Plots showing the number of fingers with respect to: a) particle concentration in the upper
443 layer (g/l) for GB and Andes-VA and b) particle size (μm) for GB, Andes-VA, Rhyol-VA and
444 Basalt-VA; and the speed of fingers with respect to: c) particle concentration in the upper layer (g/l)
445 for GB, Andes-VA, and Theory (Hoyal et al., 1999) and d) particle size for GB and Andes-VA
446 (μm) (experiments 19 to 26 and 31 to 43 in Table 1).

447 **Figure 5.** Images (26 cm x 16.5 cm) showing the number of fingers for the experiments with a
448 mixed upper layer using a) GB between 45 and 63 μm (experiment 28 in Table 1) and b) Andes-VA
449 between 45 and 63 μm (experiment 29 in Table 1).

450 **Figure 6.** Images (26 cm x 16.5 cm) of experiments with GB of diameter between 32-45 μm
451 (experiment 11 in Table 1) taken a) 5 s and b) 30 s and of diameter between 45-63 μm (experiment
452 3 in Table 1) taken c) 5 s and d) 30 s after removing the horizontal pet sheet. Arrows indicate
453 observed fingers.

454 **Figure 7.** Plot showing the evolution of the finger number with time for GB between 45-63 μm
455 (experiment 3 in Table 1).

456 **Figure 8.** Images (26 cm x 16.5 cm) of experiment 24 in Table 1 that includes Andes-VA with a
457 wide range of particle sizes ($< 125 \mu\text{m}$) imaged about a) 5 s, b) 10 s, c) 15 s, and d) 20 s after
458 removing the horizontal pet sheet.

459 **Figure 9.** Image of the experiment 7 (Table 1) for GB between 45-63 μm a) after 15 s the finger
460 formation; b) divergence (s^{-1}) and c) vorticity (s^{-1}) fields and d) divergence and vorticity values
461 measured along the orange line across the finger in c).

462

463 **Table 1.** Summary of experiments: experiment number; particle composition including glass beads
464 (GB), andesitic volcanic ash (Andes-VA), rhyolitic volcanic ash (Rhyol-VA), and basaltic volcanic
465 ash (Basalt-VA); particle size (S) in μm ; Concentration in g/l; the upper layer was both quiescent
466 (i.e. unmixed experiments, or continually mixed using a rotary stirrer (i.e. mixed experiments))
467 described in Manzella et al. (2015).

468

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Figure 1



Figure 2

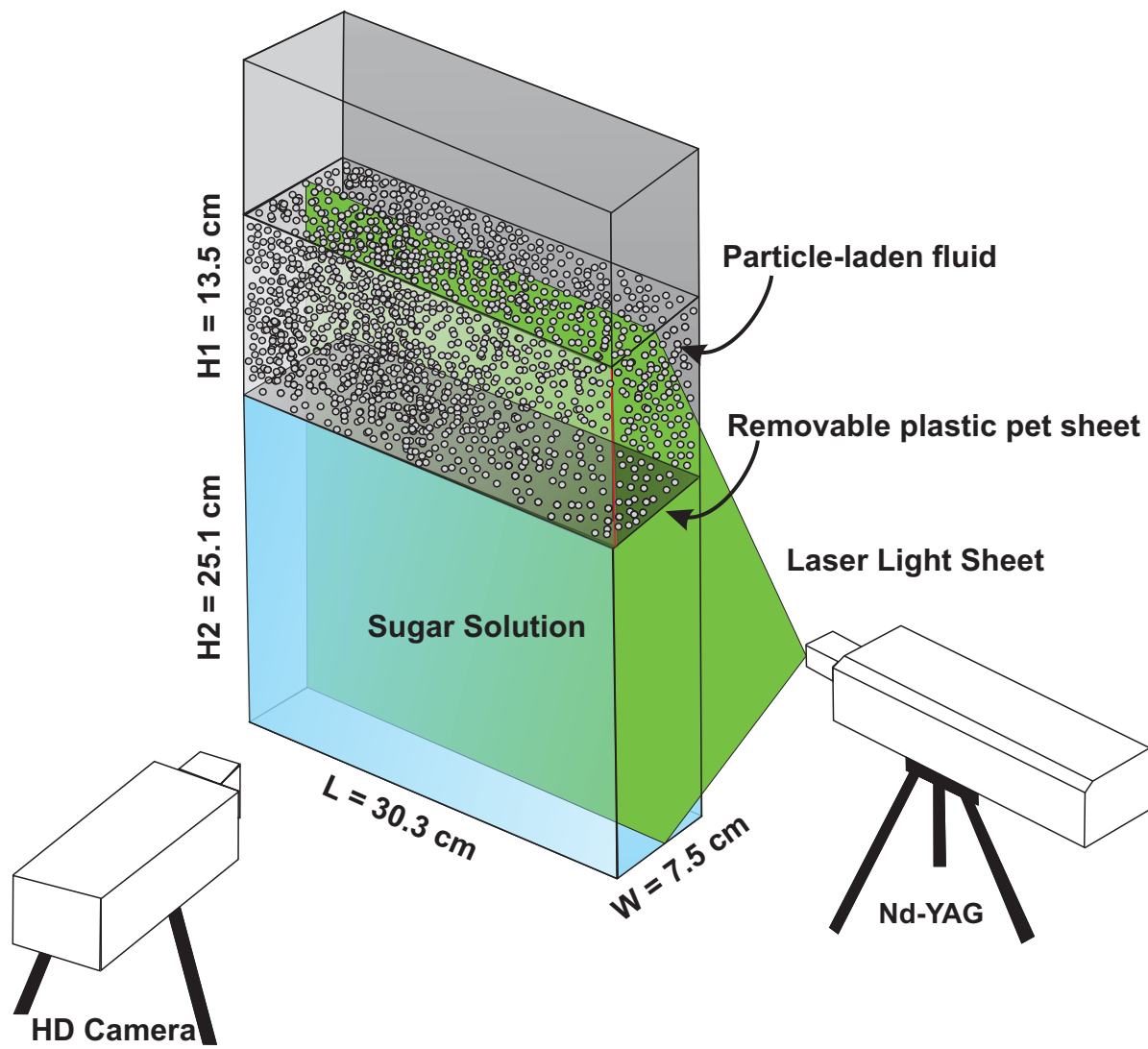


Figure 3

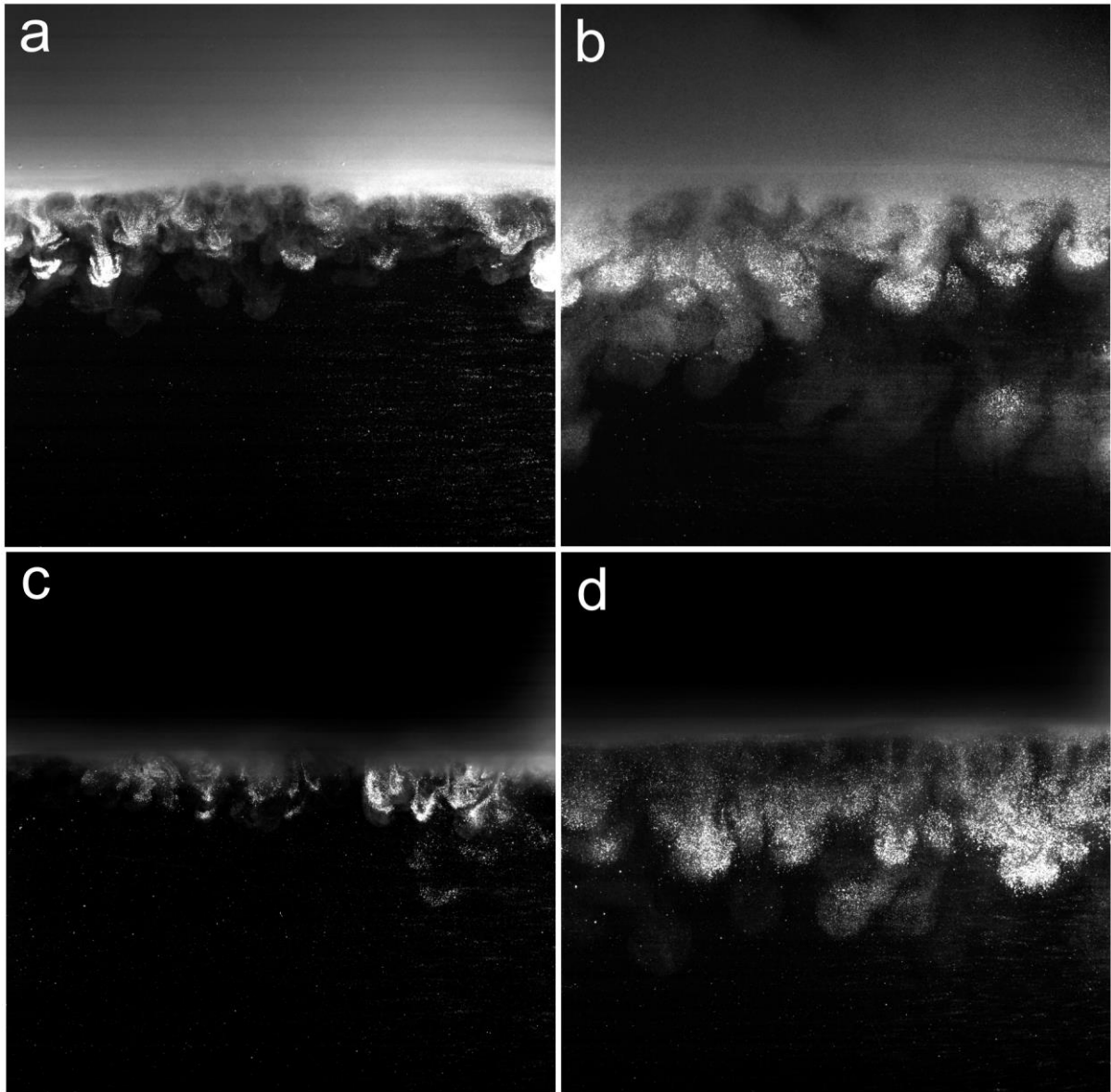


Figure 4

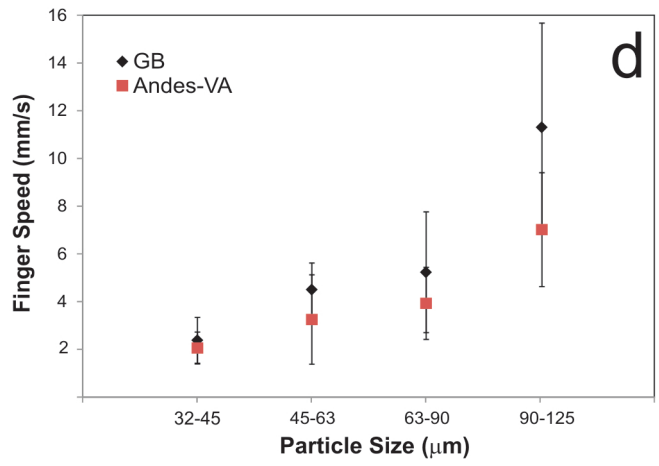
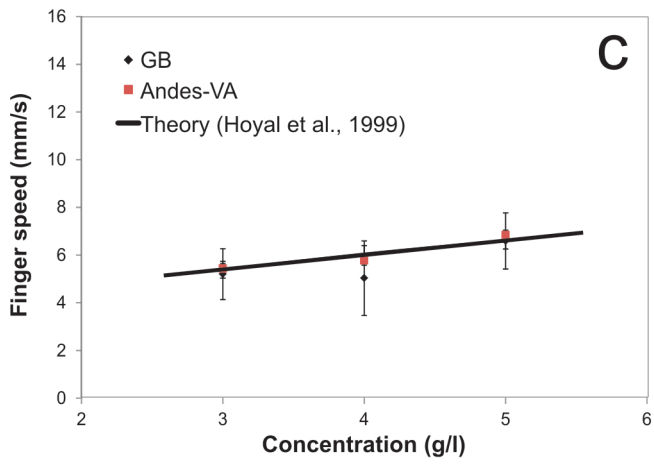
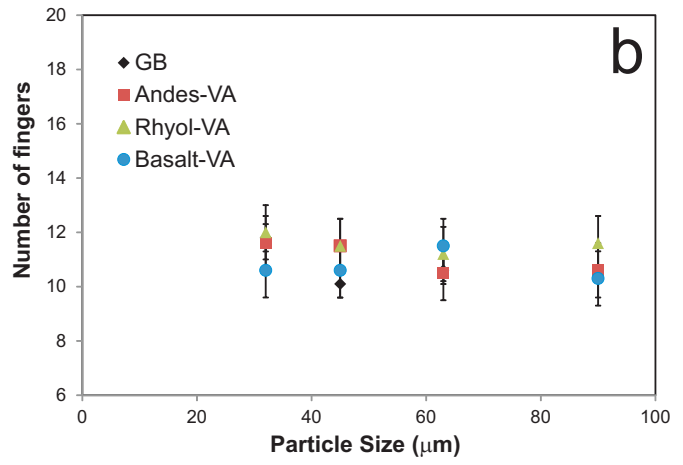
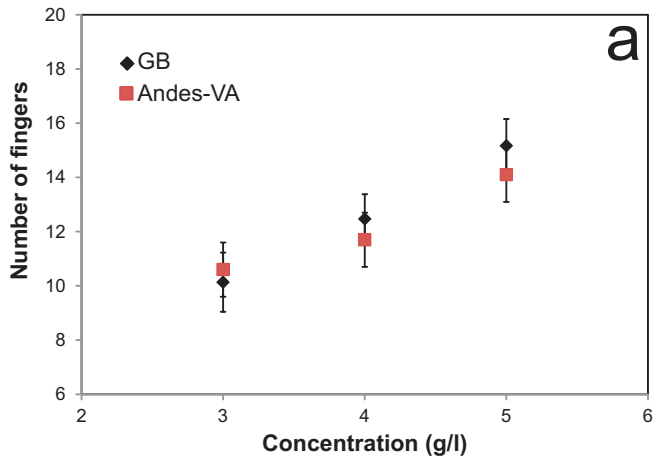


Figure 5

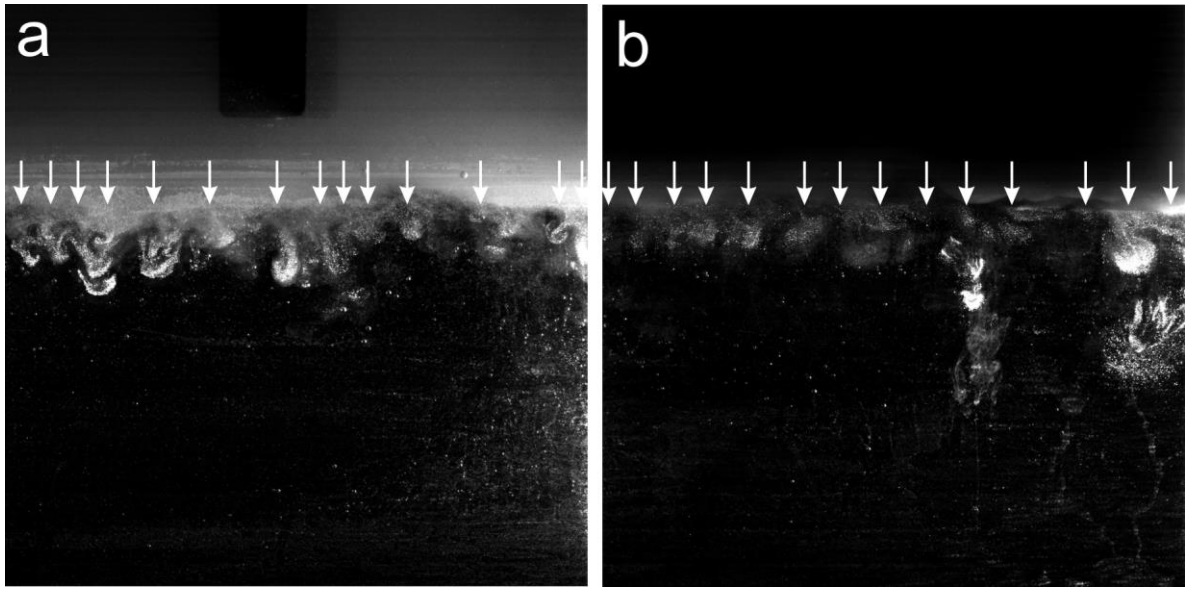


Figure 6

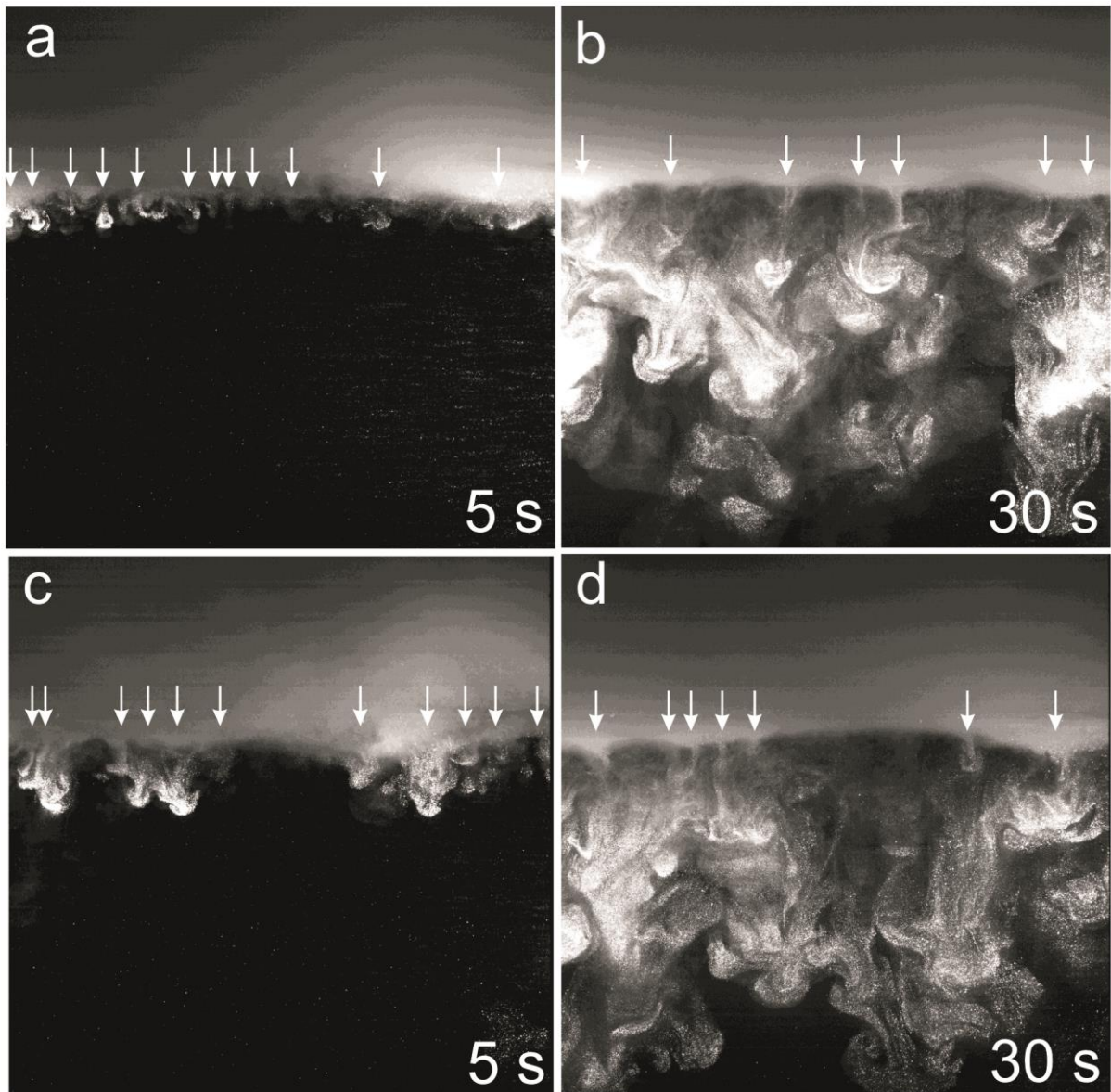


Figure 7

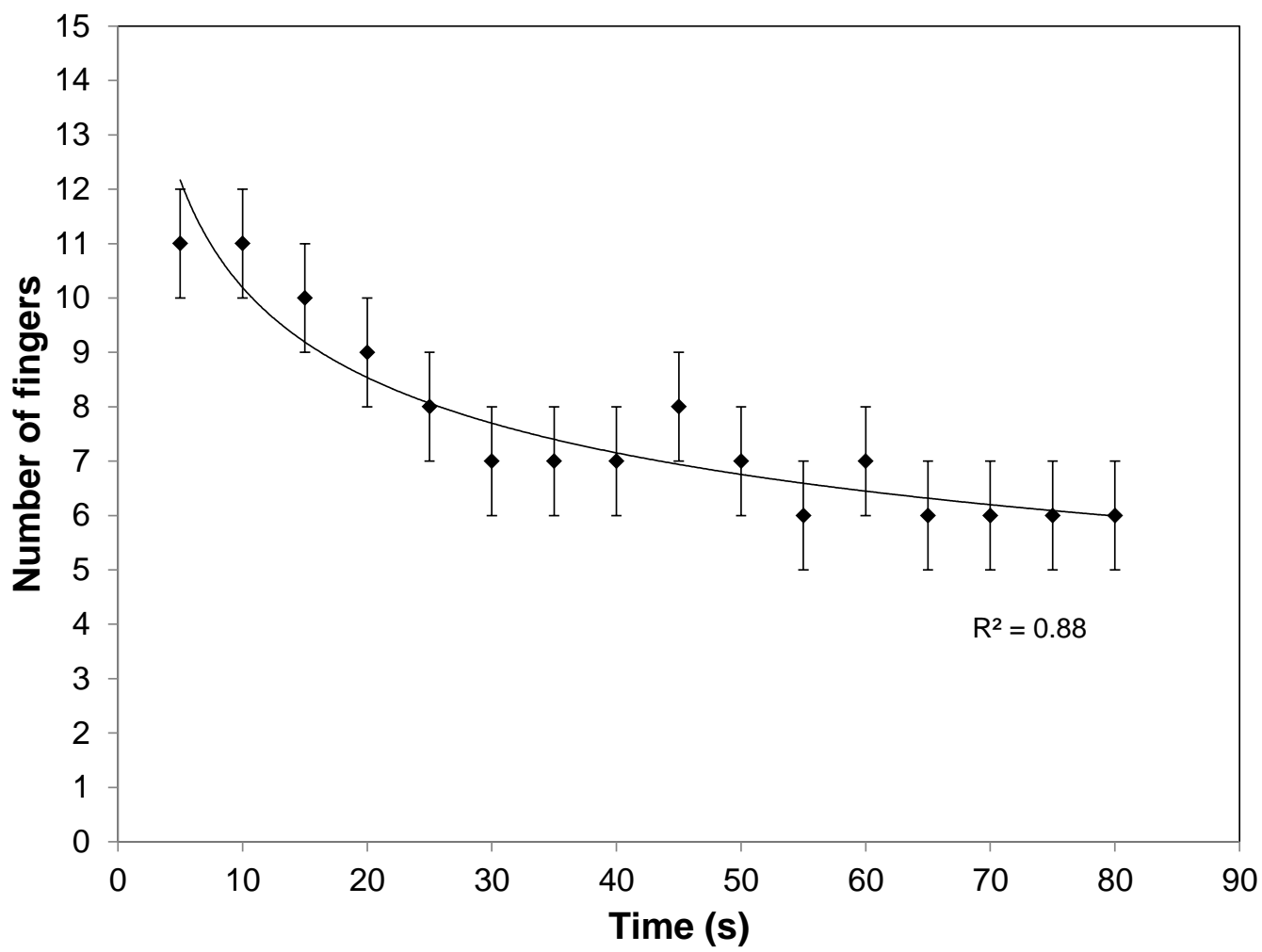


Figure 8

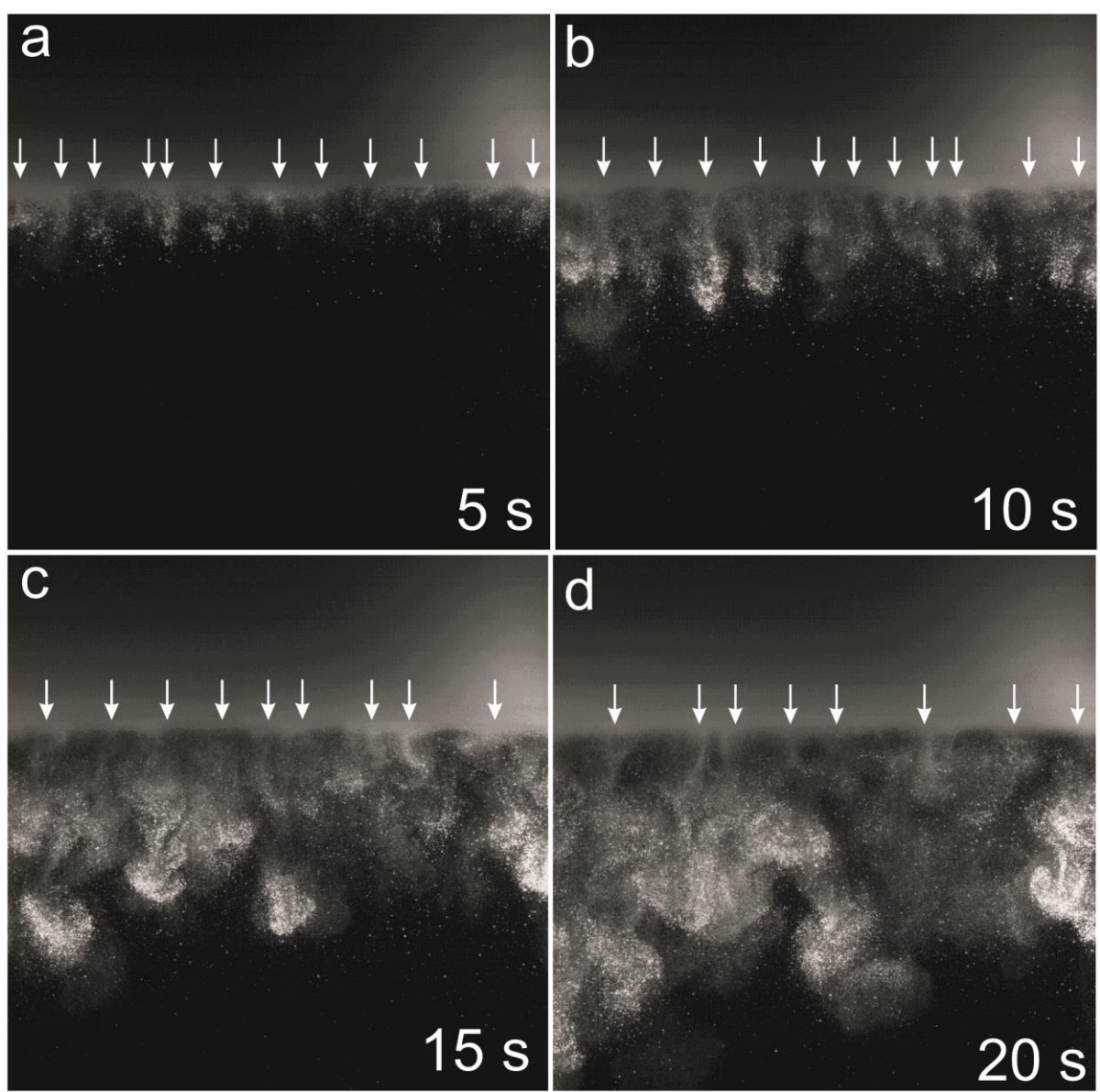


Figure 9

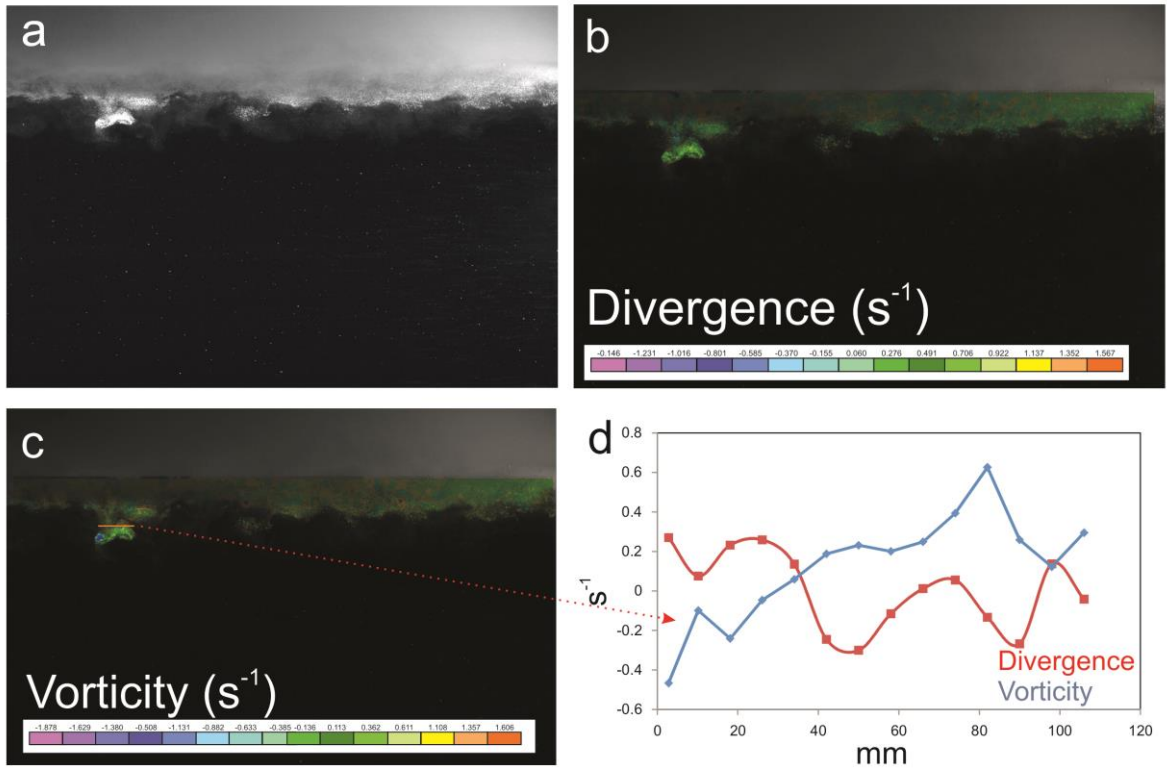


Table 1

Experiment number	Particle composition	Particle size (μm)	Concentration (g/l)	Experiment conditions
1	GB	45-63	3	Unmixed
2	GB	45-63	3	Unmixed
3	GB	45-63	3	Unmixed
4	GB	45-63	4	Unmixed
5	GB	45-63	4	Unmixed
6	GB	45-63	4	Unmixed
7	GB	45-63	4	Unmixed
8	GB	45-63	5	Unmixed
9	GB	45-63	5	Unmixed
10	GB	45-63	5	Unmixed
11	GB	32-45	3	Unmixed
12	GB	32-45	3	Unmixed
13	GB	32-45	3	Unmixed
14	GB	63-90	3	Unmixed
15	GB	63-90	3	Unmixed
16	GB	<32	3	Unmixed
17	GB	<32	3	Unmixed
18	Andes-VA	125-180	3	Unmixed
19	Andes-VA	90-125	3	Unmixed
20	Andes-VA	63-90	3	Unmixed
21	Andes-VA	45-63	3	Unmixed
22	Andes-VA	32-45	3	Unmixed
23	Andes-VA	<32	3	Unmixed
24	Andes-VA	<125	3	Unmixed
25	Andes-VA	63-90	4	Unmixed
26	Andes-VA	63-90	5	Unmixed
27	Andes-VA	<125	3	Mixed
28	GB	45-63	3	Mixed
29	Andes-VA	45-63	3	Mixed
30	Andes-VA	<125	3	Mixed
31	Rhyol-VA	<32	3	Unmixed
32	Rhyol-VA	32-45	3	Unmixed
33	Rhyol-VA	45-63	3	Unmixed
34	Rhyol-VA	63-90	3	Unmixed
35	Rhyol-VA	90-125	3	Unmixed
36	Rhyol-VA	<125	3	Unmixed
37	Basalt-VA	<32	3	Unmixed
38	Basalt-VA	32-45	3	Unmixed
39	Basalt-VA	45-63	3	Unmixed
40	Basalt-VA	63-90	3	Unmixed
41	Basalt-VA	90-125	3	Unmixed
42	Basalt-VA	<125	3	Unmixed
43	Basalt-VA	<32	3	Unmixed