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# 1 Success and breakdown of Tanner's law for drops of 2 dense granular suspensions

3 Alice Pelosse<sup>†</sup>, Élisabeth Guazzelli and Matthieu Roché

4 Université Paris Cité, CNRS, Matière et Systèmes Complexes UMR 7057, F-75013, Paris

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6 The spreading of viscous drops of density-matched suspensions on a solid surface is  
7 experimentally investigated at the global drop scale. The spreading dynamics still obeys  
8 Tanner's law provided one uses an effective viscosity which happens to be smaller than the  
9 bulk viscosity of the suspension. When the height of the drop is of the order of the particle  
10 size, Tanner's law breaks down as the particles start to freeze and the pure fluid drains out of  
11 the solid matrix.

12 **Key words:**

---

## 13 1. Introduction

14 The spreading of a liquid on a solid substrate is a common phenomenon, e.g. on plant leaves  
15 after the impact of raindrops or insecticide droplets, or during the coating of surfaces in  
16 industry. In these examples, the spreading fluid is not always a pure liquid as it may carry  
17 rigid bodies, such as fine dust particles in raindrops or solid particles added to the coating  
18 fluid. It is therefore of great importance to examine whether the spreading laws established  
19 for a pure fluid are still valid for such a complex fluid.

20 In this work, we focus on granular suspensions, a type of complex fluid extensively studied  
21 in bulk-flow situations, but quite unexplored when confined by a free interface such as for  
22 a spreading droplet. Granular suspensions are made of large particles (i.e. having diameter  
23  $d > 10 \mu\text{m}$ ) such that Brownian motion and colloidal forces are negligible. Despite their high  
24 degree of heterogeneity at the microscopic scale, these particulate systems can be seen as  
25 continuous effective fluids at the macroscopic scale, with a bulk viscosity that solely depends  
26 on the particle volume fraction,  $\phi$ , and is notably independent of particle diameter  $d$  for  
27 monodisperse rigid spheres (see e.g. Guazzelli & Pouliquen 2018).

28 Such a macroscopic description fails when the size of the system approaches that of the  
29 particle as seen in thin films (Palma & Lhuissier 2019; Gans *et al.* 2019) or near an advancing  
30 contact line (Zhao *et al.* 2020; Pelosse *et al.* 2023). In that latter case, the relation between  
31 the dynamic contact angle and the spreading velocity is similar to the classic Cox-Voinov  
32 law observed in the case of a pure liquid (Voinov 1976; Cox 1986). However, the wetting  
33 viscosity involved in this law differs from that of the bulk as it depends not only on  $\phi$  but

<sup>†</sup> Email address for correspondence: [alice.pelosse@gmail.com](mailto:alice.pelosse@gmail.com)

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34 also on  $d$ . This observation is linked to the ability of the particles to approach the contact  
 35 line close enough to affect dissipation (Zhao *et al.* 2020; Pelosse *et al.* 2023).

36 In the present work, the spreading of viscous drops of density-matched suspensions (having  
 37 a volume  $V_0$ , a surface tension with air  $\gamma$ , and a density  $\rho$ ) on a solid surface is experimentally  
 38 investigated at the global drop scale, i.e. by recording the time evolution of their radius,  $R(t)$ .  
 39 For pure fluids, the radius growth is expected to follow Tanner's law (Tanner 1979) such  
 40 that  $R(t) = A t^n$ , where both the factor  $A$  and the exponent  $n$  depend on  $V_0$ . For drop radii  
 41 smaller than the capillary length, i.e.  $R_0 \sim V_0^{1/3} < \ell_c = \sqrt{\gamma/\rho g}$  (with  $\ell_c = 1.8$  mm in  
 42 the present work), the spreading is driven by the balance of capillary and viscous forces,  
 43 leading to  $A \propto (V_0^3 \gamma/\eta)^{1/10}$  and  $n = 1/10$  (De Gennes 1985). Conversely, for large drops,  
 44 i.e.  $R_0 > \ell_c$ , gravity prevails over capillarity and the spreading behaviour is established  
 45 through the balance between gravity and viscous dissipation, resulting in  $A \propto (V_0^3 \rho g/\eta)^{1/8}$   
 46 and  $n = 1/8$  (Lopez *et al.* 1976; Hocking 1983). This paper discusses the validity of Tanner's  
 47 law for large drops of granular suspensions and compares the present dynamics at the global  
 48 drop scale to that previously obtained at a local scale in the vicinity of the contact angle.

## 49 2. Experimental methods

50 Two different types of granular suspensions have been used in the experiments. The first  
 51 combination of particles and fluid consists of spherical polystyrene beads (Dynoseeds  
 52 TS, Microbeads, Norway) suspended in a density-matched Newtonian PEG copolymer  
 53 [Poly(ethylene glycol-ran-propylene glycol) monobutyl ether] (Sigma) widely used in pre-  
 54 vious experimental work (see e.g. Guazzelli & Pouliquen 2018). The fluid density  $\rho =$   
 55  $1056 \text{ kg/m}^3$  is close to that of polystyrene and its dynamic viscosity is measured to be  
 56  $\eta_f = 2.4 \pm 0.1 \text{ Pa}\cdot\text{s}$  at  $22^\circ\text{C}$ . Different batches of particles are used with varying mean  
 57 diameters  $d = 20, 40, 80, 140, 250$  and  $550 \mu\text{m}$  (with dispersion in size of 10%). The  
 58 suspension mixture is made by weighting a mass of suspending fluid and adding the amount  
 59 of solid needed to reach the desired particle volume fraction  $\phi = 40\%$  in most of the situations  
 60 presented in §3. Larger loading in particles are also investigated as described in §3.4. Mixing is  
 61 achieved by (i) first slowly stirring with a spatula and (ii) then using a rolling device overnight.  
 62 The particles are found to be completely wet by the fluid and to experience no aggregation.  
 63 The surface tension of the suspensions is that of the PEG copolymer,  $\gamma = \gamma_f \approx 35 \text{ mN m}^{-1}$ ,  
 64 as confirmed by pendant drop experiments. The second type of suspension consists of  $60\text{-}\mu\text{m}$   
 65 PMMA spheres (Spheromers CA, Microbeads, Norway) immersed in a fluid chosen to match  
 66 the density and index of the particles. This fluid is a mixture of Triton X-100 (73 wt%), zinc  
 67 chloride (16 wt%), and water (11 wt%) having a viscosity  $\eta_f = 3.3 \pm 0.1 \text{ Pa}\cdot\text{s}$  at  $22^\circ\text{C}$ . A  
 68 fluorescent dye, Rhodamine 6G, is added to the fluid to aid the visualisation with a laser  
 69 sheet as described in §3.3. Only a single solid volume fraction of  $\phi = 40\%$  has been studied  
 70 for this suspension mixture.

71 Drops comprising the smallest particles (from  $20 \mu\text{m}$  to  $250 \mu\text{m}$ ) are made using a syringe  
 72 pump pushing the target volume of fluid at a flow rate  $1 \text{ mL}/\text{min}$  out of a  $3 \text{ mm}$  needle (inner  
 73 diameter). To avoid transient heterogeneity in the suspension microstructure, at least  $3 \text{ mL}$   
 74 of suspension is discarded before any experiments. For the largest  $550 \mu\text{m}$  particles, due to  
 75 concerns about confinement effects in the tubing, the drops are made manually with a spatula  
 76 ensuring a better control of  $\phi$ . In all cases, the drops are deposited onto (Neyco Fused Silica)  
 77 Quartz substrates carefully enough to produce axisymmetric spreading. To this extent, before  
 78 each experiment, the substrate is cleaned thoroughly with deionised water and ethanol and  
 79 treated with a plasma cleaner to avoid pinning of the advancing contact line. Under these  
 80 conditions, the continuous phase of the suspensions wets completely the surface of the wafer.

81 The spreading of the drop is recorded by a (Imaging Source) monochrome digital camera

82 (with a spatial resolution of 23 pixels/mm) located 50 cm above the solid substrate and  
 83 operated at a frame rate of  $1 \text{ s}^{-1}$  which provides a capture of both early and long-time  
 84 dynamics. Automatic measurement of the radius is performed by a Python script that (i) first  
 85 blurs locally the raw picture over roughly 5 pixels, (ii) then detects the edges of the drop  
 86 using the (scikit-image toolbox) Canny edge detector with a Gaussian width of 1, and (iii)  
 87 finally returns the best circle fitting the outer radius using the (scikit-image toolbox) Hough  
 88 transform function. Additional side-view recording is performed by a camera positioned at a  
 89 few centimetres from the drop (with spatial resolution of  $0.6 \mu\text{m}/\text{pixel}$ ) which is synchronised  
 90 with the top-view camera. Post-treatment of the recording uses the Sobel filter to detect drop  
 91 contour and extract the drop profile as a function of time (Pelosse *et al.* 2023).

92 In our experiments, the initial time is ill-defined, see figure 1(a). Indeed, as the drop  
 93 touches the solid surface and starts spreading, it is attached to the needle for a certain amount  
 94 of time which goes from a few seconds to one minute depending on the drop composition.  
 95 The growth of the radius according to Tanner's law is therefore only investigated once the  
 96 drop is fully detached from the needle, i.e. at a delayed time  $t_0$ . The radius data are therefore  
 97 fitted with the parameterised function  $R(t, t_0, A) = A(t + t_0)^{1/8}$ , where  $A = k(\rho g V_0^3 / \eta_f)^{1/8}$   
 98 with  $k$  a constant,  $V_0$  the drop volume inferred from its mass, and  $\eta_f$  the viscosity of the PEG  
 99 copolymer measured by a capillary viscometer for each experiment. The fitted parameters  $t_0$   
 100 and  $A$  are estimated numerically. An alternative method is to use an initial radius instead of  
 101 an initial time, i.e.  $R(t, R_0, A') = R_0 + A't^{1/8}$  (Saiseau *et al.* 2022). This is equally efficient  
 102 but we have chosen the time-offset correction in the present data analysis. For smaller drops,  
 103 a transition from a (short-time) capillary to a (long-time) gravity regime has been reported  
 104 (Cazabat & Cohen Stuart 1986; Levinson *et al.* 1988). Such changes in power law are not  
 105 observed with the drop sizes investigated in the present work. It should be stressed that the  
 106 time offset correction only affect the behaviour at short time and does not modify that at long  
 107 time which is undoubtedly driven by gravity with a power law exponent of  $1/8$  as evidenced  
 108 in figure 1(a) discussed in § 3.1.

## 109 3. Results

110

### 3.1. Tanner's law

111 Figure 1(a) shows the typical time-evolution of the radius of drops consisting of pure  
 112 fluid and granular suspensions. The colour coding indicates the different fluids while the  
 113 symbol coding the varying drop volume. The raw data are plotted versus time  $t$  in the inset  
 114 whereas the main graph displays the normalised radius  $R^* = R(\eta_f / V_0^3)^{1/8}$  versus  $t + t_0$ .  
 115 This radius normalisation removes volume and viscosity effects, as evidenced by the tight  
 116 collapse of the curves for a given fluid. The time offset corrects the initial bending seen  
 117 in the inset as already explained at the end of § 2. The spreading clearly obeys a gravity-  
 118 driven dynamic, i.e.  $R \sim t^{1/8}$ , as expected for the relatively large Bond numbers considered,  
 119  $2.6 \leq Bo = (3V_0/4\pi)^{2/3}/\ell_c^2 \leq 11.9$ . Drops of suspensions of any volume and concentration  
 120 seem to obey Tanner's law established for continuous Newtonian fluids. However, there are  
 121 two main differences. First, the curves of the main log-log plot for suspension drops exhibit  
 122 a small vertical shift compared to those for the pure fluid which indicates that the factor  $A$   
 123 of Tanner's law differs when adding particles to the fluid. Second, for the largest particles  
 124 ( $d = 550 \mu\text{m}$ ), the dynamic slows down for  $t \gtrsim 200 \text{ s}$ . Deviations from Tanner's law will be  
 125 successively discussed in the following § 3.2 and 3.4.

4

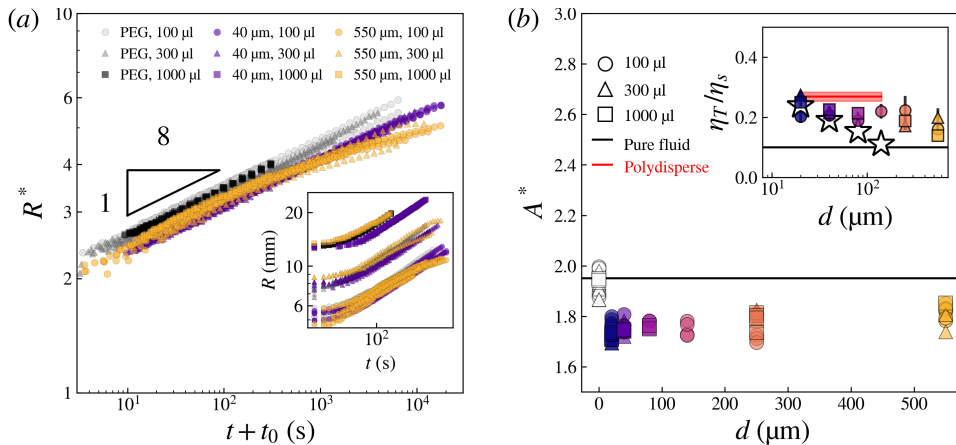


Figure 1: (a) Normalised radius  $R^* = R(\eta_f/V_0^3)^{1/8}$  versus time (with the  $t_0$  offset) for different drop volume  $V_0 = 100, 300,$  and  $1000 \mu\text{L}$  and different fluids: suspensions with  $d = 40 \mu\text{m}$  (purple symbols) and  $550 \mu\text{m}$  (gold symbols) at  $\phi = 40\%$  and pure PEG (grey and black symbols). Inset: raw data of the radius versus time. (b) Normalised factor  $A^* = A(\eta_f/V_0^3)^{1/8}$  versus particle diameter  $d$  (from  $20 \mu\text{m}$  to  $550 \mu\text{m}$ ). The open symbols correspond to experiments with pure PEG. Inset: ratio of the relative effective viscosity obtained from Tanner's law,  $\eta_T$ , to the relative bulk viscosity,  $\eta_s$ , versus particle diameter  $d$ . The red horizontal line corresponds to this ratio for a polydisperse suspension made of particles of  $20 \mu\text{m}, 40 \mu\text{m}, 80 \mu\text{m}$  and  $140 \mu\text{m}$ , each size representing  $10\%$  of the suspension volume. The star symbols corresponds to the relative effective wetting viscosity of the same suspensions obtained from the Cox-Voinov law (Zhao *et al.* 2020).

126

### 3.2. Tanner's effective viscosity

127 We start by discussing the impact of adding particles on the factor  $A$ . In the main graph  
 128 of figure 1(b), the normalised factor,  $A^* = A(\eta_f/V_0^3)^{1/8}$ , is plotted for all the experiments.  
 129 This normalisation eliminates any  $V_0$ -dependence for a given fluid. We see that the value  
 130 of  $A^*$  is smaller for suspensions than for the pure continuous phase and its value is  
 131 roughly independent of  $d$ . The viscosity of the continuous phase  $\eta_f$  used to compute  $A^*$   
 132 underestimates the viscosity of the suspensions. Seeking to compare the effective viscosity  
 133 inferred from Tanner's law with its bulk counterpart, we write the Tanner suspension viscosity  
 134 as  $\eta_T \eta_f$ . To shift the suspension data onto the solid line corresponding to the PEG data in  
 135 the figure 1(a), the relative viscosity  $\eta_T$  should be computed as  $\eta_T = (\rho g / \eta_f V_0^3) (k/A)^8$ ,  
 136 where  $k = 0.61 \pm 0.02$  has been inferred from the pure PEG data (open symbols) using  
 137  $A_{PEG}^* = k(\rho g)^{1/8} = 1.94$ . This relative viscosity extracted from Tanner's law is reported  
 138 in the inset of figure 1(b) for the different suspensions and drop volumes. It is found to be  
 139 independent of  $d$  and  $V_0$  with a value  $\eta_T = 2.2 \pm 0.5$ .

140 This relative Tanner viscosity  $\eta_T$  which describes the global dynamics of the drop differs  
 141 from the relative wetting viscosity  $\eta_w$  extracted from the Cox-Voinov law found in the  
 142 viscous–capillary region corresponding to the very close vicinity of the contact line, see the  
 143 star symbols on the inset of figure 1(b). In contrast with the constant behaviour of  $\eta_T$ ,  $\eta_w$   
 144 decreases with increasing  $d$  and reaches a value of one (that corresponding to the pure fluid)  
 145 for a cut-off size ( $\approx 100 \mu\text{m}$ ) above which particles are too large to affect dissipation in this  
 146 region close to the contact line (Zhao *et al.* 2020; Pelosse *et al.* 2023). This difference between  
 147  $\eta_w$  and  $\eta_T$  may not come as a surprise as it derives from two different energy balances at the  
 148 local and global scales, i.e. from a balance of capillary and viscous forces at the local scale of  
 149 the contact line and of gravity and viscous forces at the global scale of the drop.

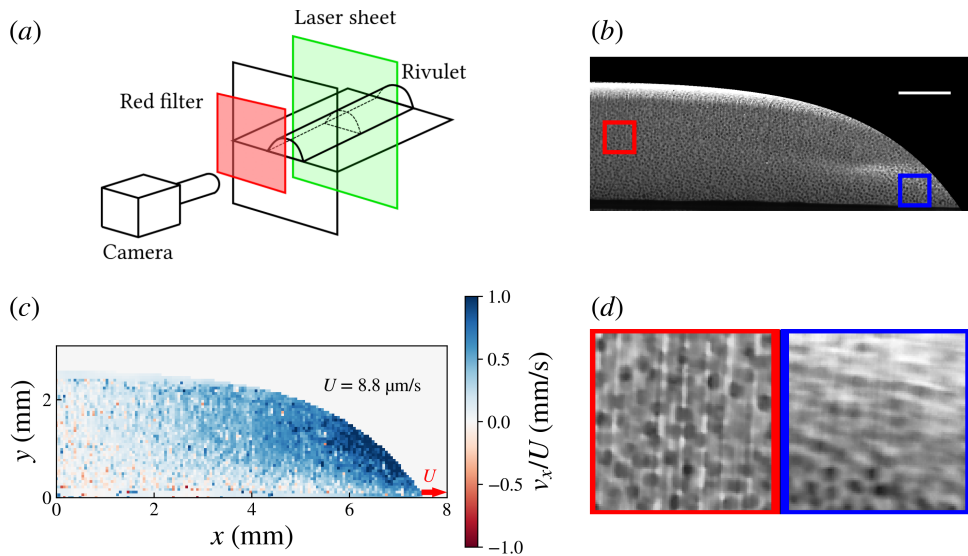


Figure 2: (a) Sketch of the apparatus used to visualize the particle flow in a spreading rivulet with a laser sheet. A fluorescent dye suspended in the fluid is excited by the green light and fluoresces red light collected by a side view camera. (b) Imaging for a suspension made of  $60\text{-}\mu\text{m}$  particles at  $\phi = 40\%$  (scale bar: 1 mm). See accompanying movie 1 in the supplementary material. (c) Horizontal speed  $v_x$  from PIV analysis of the particle flow in (b). The speed is normalised by the contact line velocity  $U$ . (d) Sum of raw pictures between  $t$  and  $t + \Delta t$  with  $\Delta t = 2d/U$  in the corresponding red (left) and blue (right) squares in subplot (b).

150 The Tanner viscosity happens also to be much smaller than the bulk viscosity of the  
 151 suspension as the relative bulk viscosity is  $\eta_s \simeq 10$  at the same  $\phi = 40\%$ . This may be  
 152 due to particle slip along the very smooth substrate. Another difference may come from  
 153 the suspension microstructure which certainly is dissimilar in the present non-viscometric  
 154 flow and in a pure shearing flow. Besides, confinement by both the substrate and the mobile  
 155 interface may create layering that could span over 10 particle diameters at  $\phi = 40\%$  (Gallier  
 156 *et al.* 2016) and end up in a strong dip in dissipation (Ramaswamy *et al.* 2017). However,  
 157 experiments with polydisperse suspensions do not support this explanation as suspensions  
 158 consisting of a mixture of 20, 40, 80, and 140  $\mu\text{m}$  particles (meant to hinder crystallisation)  
 159 do not exhibit a significant increase in dissipation, see red line in the inset in figure 1(b). To  
 160 discriminate between possible causes of this smaller value of  $\eta_T$ , we examine in detail the  
 161 particulate flow during spreading in the following § 3.3.

### 162 3.3. Visualisation of the particulate flow in the rivulet configuration

163 To gain further insight into the particle motion during spreading, particle imaging is  
 164 performed in a more convenient rivulet configuration, i.e. for a non-isotropic spreading  
 165 along only one direction, using a density and index-matched suspension described in § 2. As  
 166 sketched in figure 2(a), imaging without optical distortion is achieved using two orthogonal  
 167 microscope slides. A green laser sheet (Coherent lasers,  $\lambda = 532 \text{ nm}$ , 50 mW) illuminates  
 168 the fluid rivulet through the bottom slide and a side view camera images the flow. The  
 169 laser sheet is not deflected across the flat walls nor in the index-matched suspension and  
 170 induces fluorescence of Rhodamine 6G in the illuminated slice (about  $30 \mu\text{m}$  thick) which  
 171 is excited by the green light and emits red wavelengths. This light is filtered by a high-  
 172 pass red filter (Thorlabs,  $\lambda > 550 \text{ nm}$ ) and collected by the monochrome camera (Imaging



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173 Source, DMK33UX174, 2.3 MP) mounted with a zero distortion lens (Opto Engineering,  
 174 MC100X), and orthogonal to the laser sheet. The spatial resolution is of  $3\ \mu\text{m}/\text{pixel}$  and the  
 175 acquisition frame-rate of  $1\ \text{s}^{-1}$ . With this apparatus, the non-fluorescing particles appear dark  
 176 in the illuminated slice as seen in figure 2(b) and the accompanying movie. The horizontal  
 177 speed coming from the PIV analysis of a frame is displayed in figure 2(c). The analysis is  
 178 performed with the trackpy module of Python on two consecutive frames. The algorithm uses  
 179 an interrogation window size of 16 pixels, a search area size of 18 pixels, and an overlap of  
 180 9 pixels. The velocity is normalised by the speed of the advancing contact line (red arrow).  
 181 In figure 2(d), the sum of the pictures between  $t$  and  $t + \Delta t$  is shown for  $\Delta t = 2d/U$  in  
 182 two specific regions. With this time interval, particles moving at a speed close to that of the  
 183 contact line should not appear as fixed dark dots but should be blurred.

184 Visualisation of the particulate flow during the spreading of the suspension rivulet unveils  
 185 several significant elements. Importantly, figure 2(b) and its accompanying movie do not  
 186 exhibit any clear development of layering or ordering during spreading, meaning that  
 187 particle self-organisation cannot explain the strong decrease in viscosity as that reported  
 188 by e.g. Ramaswamy *et al.* (2017). However, PIV measurements presented in figure 2(c) and  
 189 the sum of consecutive pictures in figure 2(d) reveal two crucial features of the particle flow.  
 190 First, a strong particle slip develops along the wall near the advancing contact line as shown  
 191 by the velocity signal in figure 2(c) or by the smearing of dark dots near the rivulet edge in  
 192 figure 2(d) (right). Second, in the centre of the rivulet, particles barely move as illustrated  
 193 by the weak velocity signal from the PIV in figure 2(c) or by the sum of the pictures in  
 194 figure 2(d) (left). In this later figure, sharp dots at the centre of the rivulet (i.e. on the left)  
 195 indicate that the particles have not moved within the time interval  $\Delta t = 2d/U$ , in contrast  
 196 with the observations near the contact line (i.e. on the right) where the particulate phase  
 197 undergoes a strong flow.

198 These observations of the structure of the particulate flow can explain the low value of  
 199 the effective spreading viscosity displayed in the inset of figure 1(b). In rheometers, slip  
 200 velocity of the particles leads to a smaller stress on the particles and results in an effective  
 201 smaller viscosity (Jana *et al.* 1995; Yoshimura & Prud'homme 1988; Ahuja & Singh 2009).  
 202 In spreading experiments, particles close to the tip of the rivulet exhibit a strong slip at  
 203 roughly the contact line speed. This slip is not surprising under such strong confinement  
 204 and given the very smooth surface of the solid substrate. Yet, slip by itself cannot account  
 205 for such a large decrease in effective viscosity. This decrease could also be the result of the  
 206 almost frozen particulate phase at the centre of the rivulet such that particles act more like  
 207 a porous medium than like a flowing suspension in this region. Interestingly, the effective  
 208 viscosity of a granular bed is found to be independent of particle size and to possess a  
 209 value close to the Einstein viscosity ( $\eta_s = 1 + 2.5\phi = 2$  at  $\phi = 0.4$ ), i.e. a value much  
 210 smaller than that of a sheared dense suspension due to the screening of the hydrodynamic  
 211 interactions (Vowinkel *et al.* 2021). This bears a striking resemblance to the behaviour of  
 212  $\eta_T$  in figure 1(b). In summary, slip at the tip of the drop and porous-like behaviour at the  
 213 centre of the drop/rivulet could both account for the observed  $\eta_T$ .

#### 214 3.4. Long time behaviour: validity of Tanner's law

215 We now turn to the slowdown of the spreading dynamics observed above 200 s for suspensions  
 216 made of the largest 550- $\mu\text{m}$  particles. In figure 3, top view pictures of drop spreading evidence  
 217 different patterns for suspensions having different particle size of 40  $\mu\text{m}$ , 250  $\mu\text{m}$  and 550  $\mu\text{m}$   
 218 at  $\phi = 40\%$ . For the largest particles, top-views at a higher volume fraction, namely  $\phi = 47\%$ ,  
 219 are also presented on the last row (d). Clearly, the particle phase initially spreads with the fluid  
 220 but then does not expand further beyond a critical radius which decreases with increasing  
 221 particle size, as depicted on the first three rows (a), (b), (c), and with increasing particle

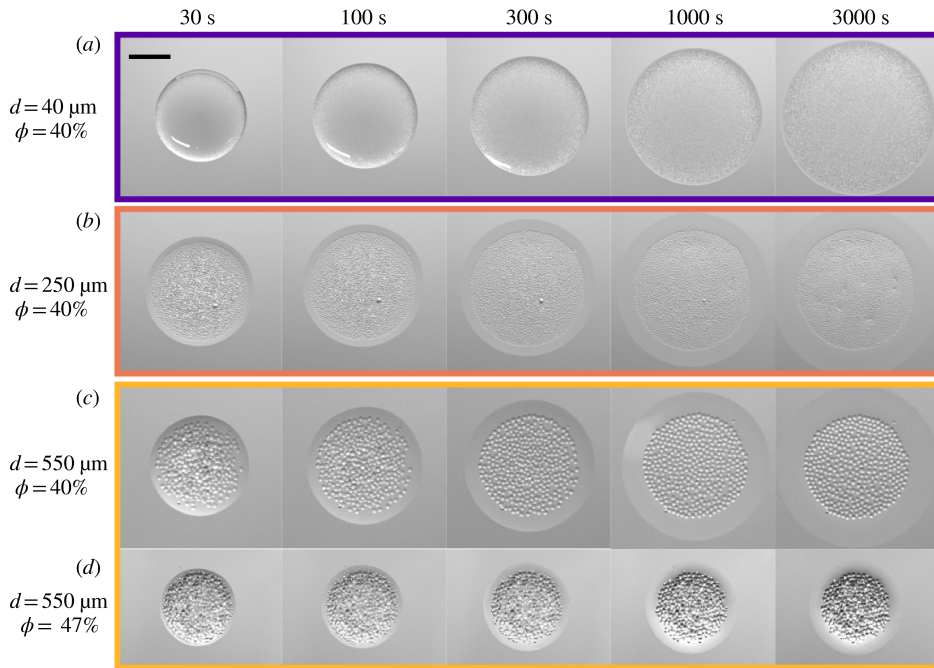


Figure 3: Top view pictures of the spreading of different suspensions. From top to bottom: (a)  $d = 20 \mu\text{m}$ ,  $\phi = 40\%$ ; (b)  $d = 250 \mu\text{m}$ ,  $\phi = 40\%$ ; (c)  $d = 550 \mu\text{m}$ ,  $\phi = 40\%$ ; (d)  $d = 550 \mu\text{m}$ ,  $\phi = 47\%$  (scale bar: 5 mm). See accompanying movie 2 in the supplementary material.

222 volume fraction, as shown on the two last rows (c), (d). There is a striking difference between  
 223 the behaviour for the smallest particles where the particle arrest is barely seen (top row) and  
 224 that for the largest particles (bottom row) where particle freeze happens at very early stage.  
 225 These observations suggest that the departure from Tanner's law can be associated with the  
 226 freezing of the particle matrix while the fluid continues to drain outward.

227 In figure 4(a), the fluid radius  $R$  of a  $100 \mu\text{L}$  drop of a suspension consisting of  $550 \mu\text{m}$   
 228 particles is plotted as a function of time (black solid line). The particle spreading radius,  
 229  $R_p$ , defined in the top left inset, is also plotted with a yellow solid line. Side-view snapshots  
 230 numbered from (i) to (iv) taken during this spreading illustrate two different regimes. First,  
 231 for pictures (i) and (ii), the drop profile is concave and  $R_p$  increases, meaning that particles  
 232 move with the fluid, in agreement with the early time dynamics in figure 3. In this regime,  
 233 the drop radius growth obeys Tanner's law and the suspension behaves like a continuous fluid.  
 234 Second, while the contact line still progresses from (ii) to (iv),  $R_p$  saturates, i.e. the particles  
 235 do not move and remain at the centre of the drop, also in agreement with observations from  
 236 figure 3. This translates into a significant slowdown of the drop radius growth  $R$  and departure  
 237 from Tanner's law (black solid line). In this second regime, the continuous approach fails  
 238 and the fluid drains out of the porous matrix consisting of the large particles. The drop  
 239 profile then takes a convex shape near the edge to connect the contact line with the centre  
 240 of the drop where the particles remain, as seen in the thumbnails (iii) and (iv). In these  
 241 pictures, the particle protrusions become more significant and one can clearly identify a  
 242 single monolayer of particles in the picture (iv). Time evolution of normalised particle radii  
 243 for drop consisting of different particle sizes and volume fractions are plotted in figure 4(b).  
 244 The same normalisation to remove volume and viscosity effects is used and the drop volumes  
 245 are kept similar at approximately  $100 \mu\text{L}$ . This graph confirms that particle freezing happens



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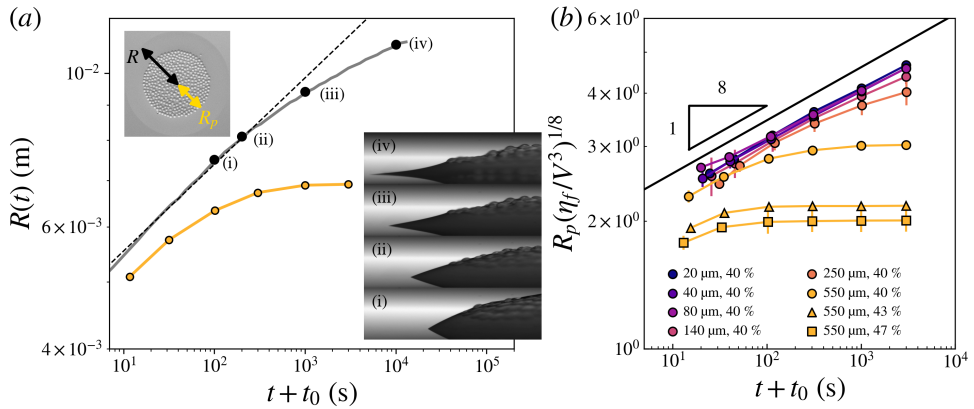


Figure 4: (a) Drop of suspension consisting of 550- $\mu\text{m}$  particles with  $V_0 = 100 \mu\text{L}$ . Main graph: fluid radius  $R$  (black) and particle radius  $R_p$  (yellow) as a function of time. The dashed line correspond to Tanner's law. Thumbnails (i) to (iv): side-view pictures corresponding to the black dots in the main graph. Top inset: top view picture with fluid ( $R$ ) and particle ( $R_p$ ) radii. (b) Particle radius  $R_p$  as a function of time for different suspensions (see legend) with a drop volume  $V_0 \approx 100 \mu\text{L}$ . Error bars are computed from standard deviations over different experiments.

246 earlier with large particles and large volume fractions. Conversely, for particle diameters  
 247  $\lesssim 100 \mu\text{m}$ ,  $R_p$  follows a power law growth with an exponent  $1/8$ , i.e. the same dynamics as  
 248 that predicted for a continuous fluid by Tanner's law.

249 To gain insight on the transition between these two regimes, the shape and thickness of the  
 250 drop have been investigated. In figure 5(a, b), plots of the time-evolution of the profile  
 251  $h(r, t)$ , of the radius  $R$ , and of the drop-centre thickness  $h_0$ , for a drop of pure fluid ( $r$  is the  
 252 radial distance from the drop centre) indicate that experimental profiles are well captured by  
 253 the predictions of Hocking (1983) for Bond numbers  $Bo = \mathcal{O}(1)$ ,

$$254 \quad h(r, t) = \frac{V}{\pi R(t)^2} \frac{I_0\left(\frac{R(t)}{\ell_c}\right) - I_0\left(\frac{r}{\ell_c}\right)}{I_2\left(\frac{R(t)}{\ell_c}\right)}, \quad (3.1)$$

255 where  $I_n(x)$  is the  $n$ -th modified Bessel function of the first kind. In figure 5(b), the radius  
 256 data (computed from top views) are fitted with Tanner's law (red dashed line) and used  
 257 to compute the drop central thickness,  $h_0$ , according to (3.1) (blue dashed line). Excellent  
 258 agreement is found with drop thickness data extracted from side views. Figure 5(a) shows  
 259 the drop profiles extracted from these side views and the comparison with (3.1) at the times  
 260 indicated by the large dots in figure 5(b). While not perfect, the agreement between the  
 261 experimental and theoretical profiles is satisfactory, especially in predicting the central drop  
 262 thickness. Side-views are therefore not required and drop profiles can be inferred using the  
 263 drop radius  $R(t)$  from top-view pictures.

264 The simplest criterion that can be proposed is that the breakdown of Tanner's law for  
 265 granular suspensions happens when the drop thickness reaches approximately a particle  
 266 diameter, as suggested by the side-view thumbnails in figure 4(a) where drop thickness is  
 267 roughly 2-diameter thick at the transition for the 550- $\mu\text{m}$  particles. This can be estimated by  
 268 evaluating the time  $t_c$  (or equivalently the drop radius  $R_c$ ) for which the central thickness  
 269  $h_0(R_c) \approx d$  with  $R_c = At_c^{1/8}$ . Figure 5(c) show that a nice collapse of drop radius versus time  
 270 can be obtained for different particle sizes and drop volumes by normalising the radius by the  
 271 critical radius  $R_c$  and the time by the critical time  $t_c$ . Spreading deviates from Tanner's law

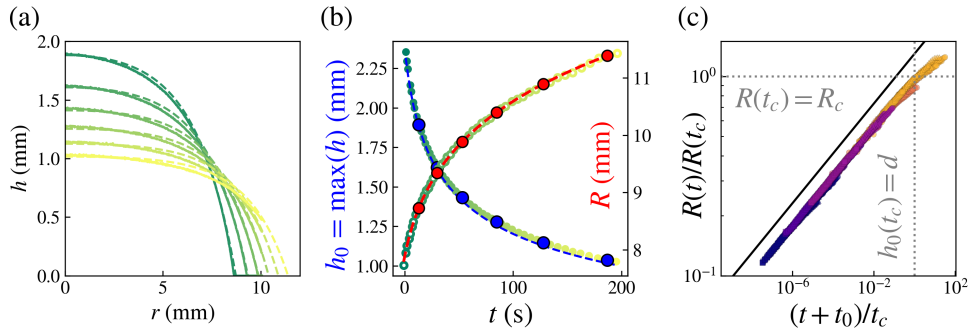


Figure 5: Spreading of a drop of pure fluid with  $V_0 = 295 \mu\text{L}$ . (a) Experimental profiles from side views (solid lines) and predicted shapes (dashed lines) from (3.1). The colour gradation indicates the time progression. The large dots in (b) correspond to the moments of the profiles plotted in (a). (b) Right axis: experimental radius  $R$  as a function of time and Tanner's law (red dashed line) with the fitted parameters  $A = 0.0059$ ,  $t_0 = 10.4$  s. Left axis: central thickness  $h_0$  as a function of time with the prediction of (3.1) (blue dashed line). (c) Radius as a function of time, normalised by  $R_c$  and  $t_c$ , respectively, with

$$R_c = At_c^{1/8} \text{ and } h_0(R_c) = d \text{ (see legend in figure 1 for the symbols).}$$

272 (solid line) for  $t \approx t_c$  for the 250 and 550  $\mu\text{m}$  particles when  $R \approx R_c$ . For the smallest particle  
 273 diameter, the experiments are too short to observe the deviation as the critical thickness  
 274  $h_0 = d$  is far from being reached.

#### 275 4. Concluding remarks

276 A global-scale investigation of the spreading of a drop of granular suspension has been  
 277 performed in the dense regime ( $\phi \geq 40\%$ ) in order to exhibit the strongest effects of the  
 278 particle phase. The first major output, developed in § 3.2, is that Tanner's law which describes  
 279 the growth of the drop radius is still valid provided one uses an effective viscosity independent  
 280 of the particle size. This so-called Tanner viscosity is much smaller than the bulk viscosity  
 281 of the suspension probably due to the combined effect of particle slip near the tip of the drop  
 282 and porous-like behaviour at the centre of the drop as shown by flow visualisation and PIV  
 283 analysis in § 3.3. The second important finding is that, when the height of the drop becomes  
 284 of the order of the particle spacing, the spreading slows down. This departure from Tanner's  
 285 law comes from particle freezing and drainage of the pure fluid out of the porous particle  
 286 matrix. In § 3.4, simply stating that this transition happens when the drop thickness reaches  
 287 a particle diameter,  $h_0(t_c) = d$ , provides a decent prediction of the breakdown of Tanner's  
 288 law at a solid volume fraction  $\phi = 40\%$ . However, a  $\phi$ -dependent criterion (which is difficult  
 289 to measure) is expected in view of the earlier particle freezing seen at larger  $\phi$  in figure 4(b),  
 290 i.e. as  $\phi$  becomes closer to a jamming volume fraction upon spreading,  $\phi_c^s$ , which is certainly  
 291 lower than the usual bulk jamming volume fraction because of confinement (Scott & Kilgour  
 292 1969; Desmond & Weeks 2009). We anticipate a weak algebraic divergence with the distance  
 293 to the jamming point similar to that found for the transition neck diameter in the pinch-off  
 294 of a viscous suspension thread (Château *et al.* 2018). A tentative criterion could then be  
 295  $h_0(t_c) = d/(\phi_c^s - \phi)^\alpha$  with  $\alpha \approx 1/3$  and could help to rationalise the earlier freezing when  $\phi$   
 296 is close to  $\phi_c^s$ . The spreading of granular drops and its transition from a continuous effective  
 297 fluid to a discrete system is therefore the signature of a confinement-dependent jamming.

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302 **Author ORCIDs.**

303 Alice Pelosse <https://orcid.org/0000-0002-4554-0604>;

304 Élisabeth Guazzelli <http://orcid.org/0000-0003-3019-462X>;

305 Matthieu Roché <http://orcid.org/0000-0002-8293-7029>.

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