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## **Postprint Version**

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## **Effects of hydrophobicity on splash erosion of model soil particles by a single water drop impact**

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### **ABSTRACT**

Raindrop impact can be a major contributor to particle mobilization for soils and other granular materials. In previous work, water repellent soils, comprised of hydrophobic particles, have been shown to exhibit greater splash erosion losses under multiple drop impact. However, the underlying principal differences in splash behavior between hydrophobic and hydrophilic granular surfaces have not been studied to date. In this study the effects of particle hydrophobicity on splash behaviour by a single water drop impact were examined using high-speed videography. Water drops (4 mm in diameter) were dropped on beds of hydrophilic and hydrophobic glass beads (sieved range: 350~400  $\mu\text{m}$ ), serving as model soil particles. The drop velocity on impact was 2.67 m s<sup>-1</sup>, which corresponds to ~30% of the terminal velocity of a raindrop of similar size. The resulting impact behaviour was measured in terms of the trajectories of particles ejected from the beds and their final resting positions. The response to the impacting water drop was significantly different between hydrophilic and hydrophobic particles in terms of the distance distribution, the median distance travelled by the particles and number of ejected particles. The greater ejection distances of hydrophobic particles were mainly the result of the higher initial velocities rather than differences in ejecting angles. The higher and longer

ejection trajectories for hydrophobic particles, compared to hydrophilic particles, indicate that particle hydrophobicity affects splash erosion from the initial stage of rainfall erosion before a water layer may be formed by accumulating drops. The ~10% increase in average splash distance for hydrophobic particles compared to hydrophilic particles suggests that particle hydrophobicity can result in greater net erosion rate, which would be amplified on sloping surfaces, for example, by ridges in ploughed agricultural soils or hillslopes following vegetation loss by clearing or wildfire.

**KEYWORDS:** splash erosion; soil water repellency; hydrophobicity; single drop impact; high-speed videography

## Introduction

Soil water repellency describes a phenomenon whereby the wetting of a soil surface is delayed for any length of time greater than immediate absorption (Scott, 2000). It occurs under natural conditions through the gradual accumulation of organic compounds with hydrophobic properties originating from plants and microorganisms (see review by Doerr *et al.*, 2000), or following the sudden accumulation of such substances during disturbances such as a wildfire (Scott, 1993; Valzano *et al.*, 1997; Doerr *et al.*, 2006) or oil contamination (Roy and McGill, 1998; Quayum *et al.*, 2002). It has been found to be a common phenomenon in soils in many regions of the world (Doerr *et al.*, 2000).

Soil water repellency has attracted considerable research interest due to its significant impact on soil hydrological and erosional processes. It can reduce or inhibit infiltration (Van Dam *et al.*, 1990; Imeson *et al.*, 1992) and consequently increase overland flow (McGhie and Posner, 1980; Crockford *et al.*, 1991; Witter *et al.*, 1991). Field observations have also shown that water repellent soils are often more susceptible to erosion than wettable soils (Megahan and Molitor, 1975; Wells *et al.*, 1979; Morris and Moses, 1987; Shakesby *et al.*, 1993) with the main reason for the enhanced erosion being an increase in overland flow caused by reduced infiltration into water repellent soils (Scott and Van Wyk, 1990; Shakesby *et al.*, 1993). The erosion processes related to water flow have received substantially more attention to date than erosion caused by rain splash. However, given that rain splash represents the initial stage of the erosive process (Blanco-Canqui and Lal, 2009), investigations into the direct effect of water repellency on the observed enhanced erosion appear to be sparsely reported.

Splash erosion is the net soil loss downslope caused by impacts of raindrops (Terry, 1992). The splash detachment process consists of three mechanisms, which are the (i) impact of a raindrop, (ii) detachment and (iii) displacement of soil particles (Terry, 1992). Although splash erosion is generally viewed as being a minor contributor to the overall water erosion loss compared to other processes, such as sheet, rill and gully erosion (Terry, 1998), it can be a dominating process in areas without overland flow such as interrill spaces (Imeson, 1977), and arid regions with little vegetation cover (Savat, 1968). In addition, the splash distance of displaced soil particles is enhanced on steep slopes (Furbish *et al.*, 2007).

Direct factors in determining splash detachment are separated into two categories in general (Terry, 1992) and these are rainfall erosivity and soil erodibility. Rainfall erosivity is

defined as the ability of rain to detach and transport soil (Epema and Riezebos, 1984) and is mainly determined by the kinetic energy of the rain (Morgan, 1995). The erodibility of soil by rain splash is determined by the particle size (Huang *et al.*, 1982; Fox *et al.*, 2007; Furbish *et al.*, 2007), aggregate stability (Farres, 1987) and shear strength of soil (Cruse and Larson, 1977; Brunori *et al.*, 1989).

Another factor that may affect soil erosion by rain splash is soil water repellency, however, to date there are very few studies that consider the effects of water repellency on splash erosion. Terry and Shakesby (1993) examined the hydrophobicity effects on rainsplash and found that hydrophobic soil had a greater splash loss with larger ejection droplets. The process was associated with the formation of a water film over the repellent soil surface, and with more detached solids being ejected. The trajectories of these tended to be shorter and lower for water repellent soil than for wettable soil. Fox *et al.* (2007) compared splash erosion loss between laboratory-burned (water repellent) and an unburned (wetable) sandy loam soils. They found that the burned soil had greater splash erosion than the unburned soil and suggested that formation of dense crusts by wetting was the main factor reducing splash detachment for the unburned soil. However, both studies above did not isolate the effect of surface water repellency from the effect of a water layer forming on the soil surface as a result of multiple drop impacts. The authors suggested that the formation of a water film or wet soil crusts by these drops accounted for the differences in the observed behaviour without considering the isolated effects of individual drop impacts on water repellent and wettable soils.

This study aimed to address this research gap by investigating the splash behaviour from a single water drop impact on beds of hydrophobic and hydrophilic spherical glass beads (serving as simple water repellent and wettable model soils respectively) under controlled laboratory conditions. Hydrophilic and hydrophobic glass particles have previously been used to provide a simple 'model' analogue to soil for particulate wettability studies (Hamlett *et al.*, 2011). This use of 'model' soil analogues allows elimination of the aforementioned water film effect, whilst keeping all other variables other than water repellency constant. Splash behaviour is often described in terms of the ejecting angle and the initial velocity of an ejecting particle (Al-durrah and Bradford, 1982; Yang, 1991; Pietravalle *et al.*, 2001), as the parabolic trajectory of an ejecting particle is determined by these two variables, assuming that the air resistance is negligible. Therefore, any difference in splash behaviour between hydrophilic and hydrophobic particles may be quantified in these two variables.

We therefore measured both ejecting angle and the initial velocity of particles using quantitative analysis of high-speed video sequences of drop impacts on beds of glass particles, and ejection distances based on resting locations of ejected particles. This allowed testing of whether or not hydrophobic particles produce a different amount and distance of splash detachments compared to hydrophilic particles for a single drop impact. We also investigated whether 1) ejecting angle or 2) initial velocity has a greater effect on any difference in particle trajectories.

## Materials and Methods

Spherical glass beads of diameter  $d$  in the range of  $350 < d < 400 \mu\text{m}$  (general purpose glass microspheres, Whitehouse Scientific, UK) were used as model soil particles with low variability in size and shape. Bead density ( $\rho_s$ ) provided by the manufacturer was  $2430\text{--}2490 \text{ kg m}^{-3}$ , and the average bead mass was  $7.6 \times 10^{-5} \text{ g}$ .

The beads were immersed in HCl (30 vol.%) for 24 hours and rinsed 3 times with distilled water and dried at  $100^\circ\text{C}$  for 12 hours to achieve fully hydrophilic particles. A subsample of these was immersed in chlorotrimethylsilane (>97% Sigma-Aldrich, UK) solution (2 vol.% in toluene; >99.8% Fisher Scientific, UK) for 48 hours and rinsed 5 times with toluene and then dried at room temperature to generate a hydrophobized sample set of particles (Hamlett *et al.*, 2011).

The water repellency of the sample sets was assessed using the water drop penetration time (WDPT) and molarity of aqueous ethanol droplet (MED) tests (Letey *et al.*, 2000). In the WDPT test, the median time required for 5 water drops ( $18 \mu\text{l}$ ) to penetrate completely into a bed of particles held in a dish of 4 cm diameter and 5 mm deep was classified according to Doerr (1998). In the MED test, the concentration of a drop ( $\sim 10 \mu\text{l}$ ) of ethanol solution required for immediate infiltration ( $< 3 \text{ s}$ ) was determined and also classified according to Doerr (1998). Three replications were made for both tests at  $20\text{--}21^\circ\text{C}$  and 30–35% relative humidity.

For drop impact experiments, the particles were held within a cylindrical cavity (diameter 1.5 and depth 0.6 cm) in the centre of a 20 cm wide polymethylmethacrylate disk (Figure 1). The combined mass of particles in the bed was  $1.63 \pm 0.03 \text{ g}$ . A warm, white 30W LED flood light, using white filter paper as a diffuser, was used as background illumination. The temperature was maintained at  $24.5\text{--}25.0^\circ\text{C}$  and the relative humidity at 40–41% throughout the experiments.

A transparent cellulose acetate sheet (inkjet transparency film) with a 1.5 cm diameter hole at its centre was used to capture particles ejected from the bed. The sheet was sprayed with water to wet the adsorbent layer on the surface to make it tacky, providing a viscous layer to immobilise the ejected particles. The adsorbent layer provided enough viscosity to arrest the landed particles, preventing them from skidding or rebounding.

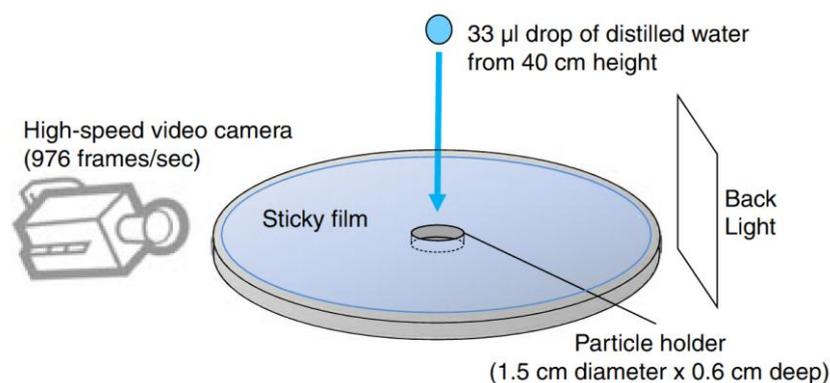


Figure 1. Schematic diagram of drop impact experimental setup.

A drop of distilled water (33  $\mu$ l) was released from a height of 40 cm above the centre of the bed. Following the impact of the drop, and ejection of particles onto the capture sheet, the sheet was air-dried and photographed on top of a black background marked with a set of concentric rings that provided a scale of distance from the centre of impact. The positions of the scattered particles in each photograph were automatically digitized using *ImageJ* software (Version 1.45c, National Institutes of Health, USA). Particles clumped around the impact site were counted by dividing the combined area by the median area of an individual particle. The total number of particles scattered, the number of particles displaced further than 1 cm distance from the centre of impact (within which similar numbers of particles are scattered for both hydrophilic and hydrophobic cases) and the median distances from the centre of impact were compared between hydrophilic and hydrophobic particles.

The splash generated by drop impact was recorded at 976 frames  $s^{-1}$  using a high-speed video camera (SVSi MVMA01-B02, Southern Vision Systems, USA) tangential to the horizontal plane (see e.g.; Yang, 1991; Furbish *et al.*, 2007). The velocity of the impacting drop was measured from the video sequences. Impacts on particle beds of both hydrophilic and hydrophobic particles were recorded and compared. The trajectories of approximately 100 particles were traced in each video sequence. The data from five replications were combined to obtain overall assessments of the effect of the impacts. The mean initial velocities and the mean ejecting angles were compared between hydrophilic and hydrophobic particles.

The initial velocity  $v_0$  and ejecting angle  $\theta_0$  of individual particles were calculated from the landing position  $x$  and the time of flight  $t$  obtained from the high-speed video frames, assuming no air resistance. The  $x$ - $y$  coordinates of particle position at time  $t$ , in terms of the initial velocity and the ejecting angle (Halliday *et al.*, 2006), are:

$$x - x_0 = v_{0x} \cdot t = (v_0 \cdot \cos\theta_0)t \quad [1]$$

(uniform motion to x direction)

$$y - y_0 = (v_0 \cdot \sin\theta_0)t - \frac{1}{2} g t^2 \quad [2]$$

(uniformly accelerated motion to y direction) where  $x$  and  $y$  are coordinates of current particle position,  $x_0$  and  $y_0$  are those of the initial position,  $v_0$  is the initial velocity along the trajectory,  $v_{0x}$  is  $x$ -axis element of  $v_0$ ,  $t$  is the time elapsed from impact,  $\theta_0$  is the ejecting angle at the initial moment, and  $g$  is the acceleration due to gravity. Substituting  $x$  and  $y$  at moment of landing, with the landing coordinates  $(x_{Landing}, 0)$ , we get

$$x_{Landing} - x_0 = (v_0 \cdot \cos\theta_0)t \quad [3]$$

$$y_{Landing} - y_0 = 0 = (v_0 \cdot \sin\theta_0)t - \frac{1}{2} g t^2 \quad [4]$$

with  $t$ , these simultaneous equations give the initial velocity  $v_0$  and the initial ejecting angle  $\theta_0$  of each particle.

In order to correct the distortion of the  $x$ -axis distance by the angle between the travel direction of an individual particle and the plane perpendicular to the view of the video camera ( $\psi$ ), all the  $x$ -coordinates were transformed by angle  $\psi$  as below.

$$x' = x / \cos\psi \quad [5]$$

The angle  $\psi$  of each particle was estimated from the departing position of the particle, assuming a radial ejection starting at a constant distance from the impact centre, which was determined as the maximum value obtained in each test.

$$\cos\psi = x_0 / x_{0\max} \quad [6]$$

Wetted perimeters generated by a drop impact were photographed, and manually digitized by the difference of brightness between wet and dry particles. The area, circularity and roundness were measured using *ImageJ* software. Circularity indicates how close the shape is to a circle and is defined here as the ratio of the area of the wetted perimeter to the square of its perimeter scaled by  $4\pi \times (\text{area})/(\text{perimeter})^2$ . The value is 1 for a perfect circle and it decreases to 0 as the shape elongates. Roundness, as the index of edge sharpness, was calculated as  $4 \times (\text{area})/\pi \times (\text{major axis of a fitted ellipse})^2$  using *ImageJ* software.

In order to determine whether or not any cohesive capillary forces (associated with local relative humidity and surface hydrophilicity) within the particle beds exerted any influence on the outcome we also examined solid plastic spheres (SP) impacts, which were similar in terms of kinetic energy to the water drop impacts. This was achieved by releasing a SP with 0.58 cm diameter and  $1.14 \text{ g cm}^{-3}$  density from 10 cm above the particle beds. The scattered particles were captured on a black card covered in a thin layer of adhesive (Craft Mount Permanent Spray Adhesive, 3M) and the card was photographed and the image digitized using *ImageJ* software. The total number and the distribution of scattered particles were determined in the same manner as for the impacts of water drops using five replications in each case under the same conditions.

The significance of any differences between hydrophobic and hydrophilic samples was tested for the following parameters using SPSS (Version 16.0, IBM, USA): (i) the shapes of the frequency distribution (Chi-square test,  $p < 0.05$ , on the basis that the responses of hydrophilic particle beds to impacts provide the control behaviour), (ii) the median ejection distance (Non-parametric Mann-Whitney U test,  $p < 0.05$ ), and (iii) all other variables including the initial velocities and ejecting angles (T-test,  $p < 0.05$ ).

## Results

### *Water repellency of particle beds*

Beds of glass beads washed with HCl were completely hydrophilic ( $\approx 0$  s WDPT and 0% MED) whereas the hydrophobized beads had a WDPT of  $> 3600$  s and 30 vol.% ethanol in water was required for immediate infiltration using the MED test. According to both tests, hydrophobized beads are extremely hydrophobic (water repellent) as defined by Doerr (1998).

## Water drop impact

### a. Particle displacements and areal scatter

Typical scatter patterns of hydrophilic and hydrophobic particle beds (Figure 2) show that more particles were detached on impact and with greater distances from hydrophobic than from hydrophilic beds. The number frequency distributions of particles as a function of distance show some similarity in shape, with consistently larger absolute numbers of particles ejected to  $\sim 3$  cm from the centre of impact from hydrophobic beds in comparison with those from hydrophilic beds (Figure 3).

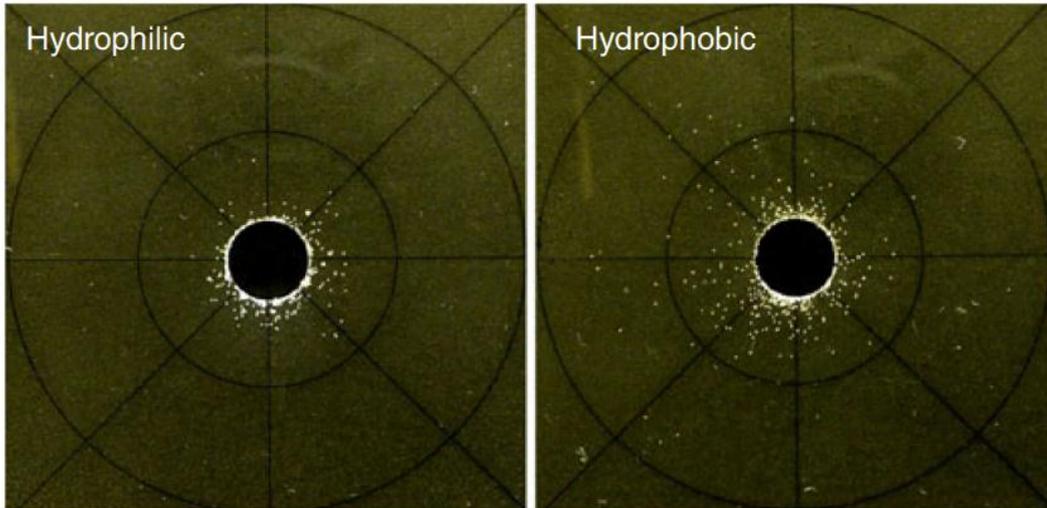


Figure 2. Splash patterns produced by an impact of a water drop ( $33 \mu\text{l}$ ) released from 40 cm height for hydrophilic (left) and hydrophobic (right) particle beds.

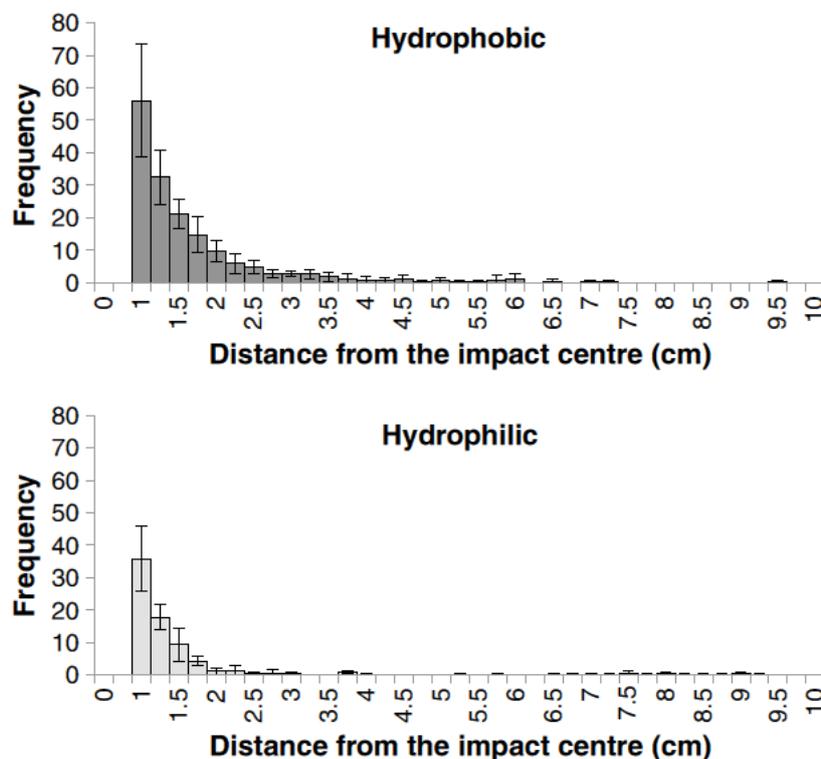


Figure 3. The number frequency distributions of the flight distance of particles, which landed further than 1 cm distance, for hydrophilic (top) and hydrophobic particles (bottom).

Using the impacts on hydrophilic particle beds as the reference to provide expected frequencies of particle displacements, a chi-squared test suggests that the distribution of displacements from the hydrophobic particle beds is significantly different (test statistic  $\sim 148$ ,  $dF = 5$ ,  $p < 0.001$ ) and much more pronounced than that found for solid sphere impacts (described below).

The numbers of particles displaced  $> 1$  cm from the centre of impact is consistently greater for all the replicate impacts on hydrophobic beds than on the hydrophilic beds. According to the non-parametric Mann-Whitney U test, the hydrophobic particles travelled significantly further (median distance, 1.44 cm) than hydrophilic ones (median distance, 1.30 cm) ( $p < 0.001$ ).

The total number of scattered particles was greater for hydrophobic particles than hydrophilic ones, but the difference was not significant (Table I). The proportion of particles that travelled further than 1 cm distance to the total number of scatters was significantly larger from hydrophobic (34%) than from hydrophilic beds (23%). The particles falling within 1 cm of the centre of impact appear to use a similar amount of the kinetic energy of impact between hydrophilic and hydrophobic cases, irrespective of whether the mode of delivery is a drop of liquid or a solid sphere of equivalent size and kinetic energy. The number of hydrophobic particles that travelled further than 1 cm was twice that for hydrophobic cases than for hydrophilic ones (Table I).

	Total number of scattered particles	Number of particles $> 1$ cm	Median distance of particles $> 1$ cm
Hydrophilic	$357 \pm 91$	$77 \pm 13$ (23%)	1.30 cm
Hydrophobic	$437 \pm 106$	$163 \pm 41$ (34%)	1.44 cm
dF and P *(significant difference)	$dF = 8$ $P = 0.24$	$dF = 5$ $P = 0.007^*$	$P < 0.001^*$

Table I. Comparison of the scatter pattern between hydrophilic and hydrophobic particles in terms of the total number of scattered particles, the number of particles that travelled further than 1 cm from the centre of impact (both by T-test at 95% confidence level) and the median flight distances of hydrophilic and hydrophobic particles by a water drop impact (by Mann-Whitney U test).

#### b. *Splashing* and *splash saltations*

Few ejections were entrained by water droplets (*splashing*) and instead most particles were dispersed without being entrained (*splash saltation*) for both hydrophilic and hydrophobic cases, as observed in the individual video frames (Figure 4).

#### c. Differences in ejecting angle and initial velocity

As shown in Figure 5a, hydrophobic particles had significantly lower mean ejecting angle ( $41.9 \pm 9.7$  degrees) than hydrophilic ones ( $46.3 \pm 10.9$  degrees) ( $p = 3.6 \times 10^{-9}$ ,  $dF = 749$ ). Also, the hydrophobic particles had a significantly higher mean initial velocity ( $38.7 \pm 16.5$   $\text{cm s}^{-1}$ ) than hydrophilic ones ( $32.4 \pm 12.1$   $\text{cm s}^{-1}$ ) ( $p = 1.45 \times 10^{-9}$ ,  $dF = 737$ ) (Figure 5b). Hydrophobic particles had both significantly higher horizontal velocity  $v_x$  ( $29.4 \pm 17.0$   $\text{cm s}^{-1}$ ) and vertical velocity  $v_y$  ( $24.1 \pm 6.0$   $\text{cm s}^{-1}$ ) than hydrophilic ones ( $22.9 \pm 13.0$   $\text{cm s}^{-1}$  and  $21.7 \pm 4.7$   $\text{cm s}^{-1}$ , respectively). The horizontal velocity showed a greater difference between hydrophilic and hydrophobic particles than did the vertical velocity (Figure 6a).

Scatter diagrams of the ejecting angle and initial velocity show a tendency for the angle to decrease with initial velocity in a similar manner for both hydrophilic and hydrophobic particles. However, the range of scatter is slightly greater for hydrophobic particles than hydrophilic (Figure 7).

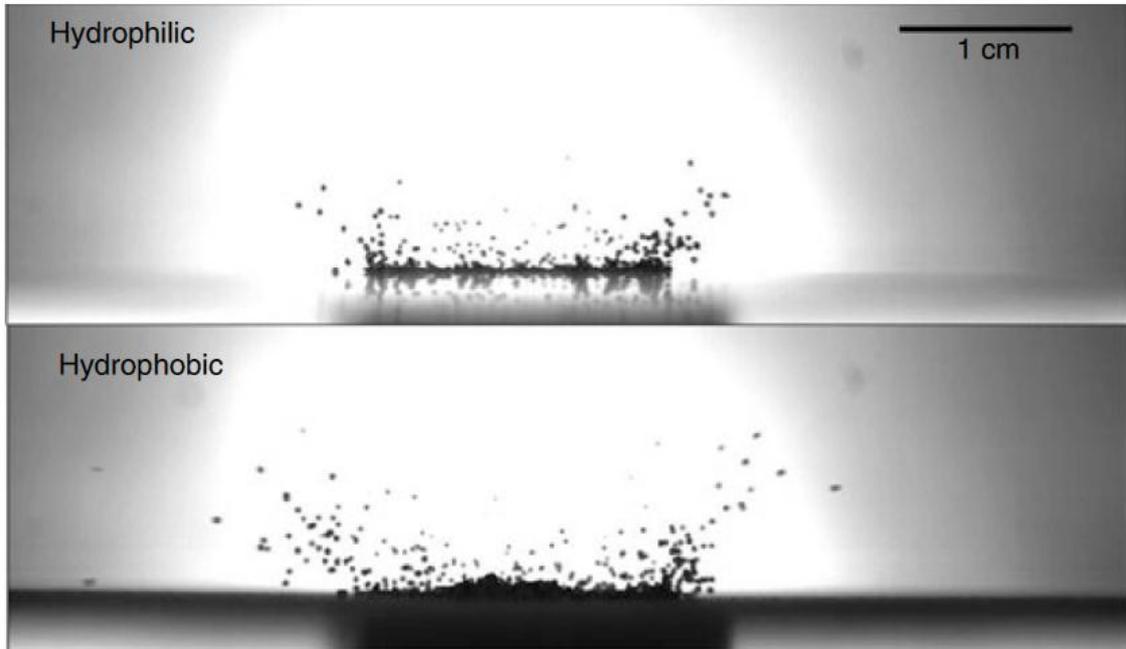


Figure 4. Ejecting particles 0.30s after a water drop impact for hydrophilic (top) and hydrophobic (bottom) particle beds.

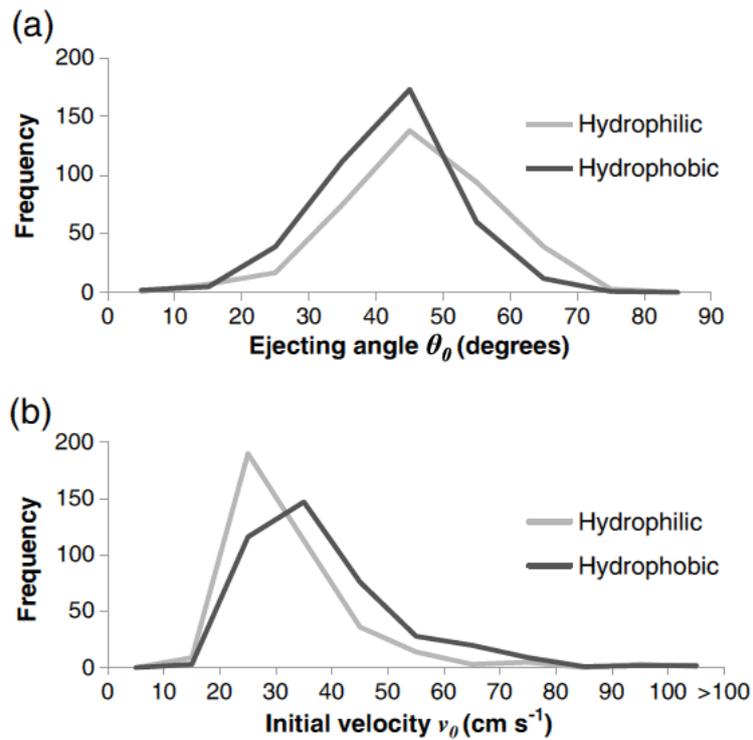


Figure 5. The number frequency distributions of (a) ejecting angles and (b) initial velocities of hydrophilic and hydrophobic particles.

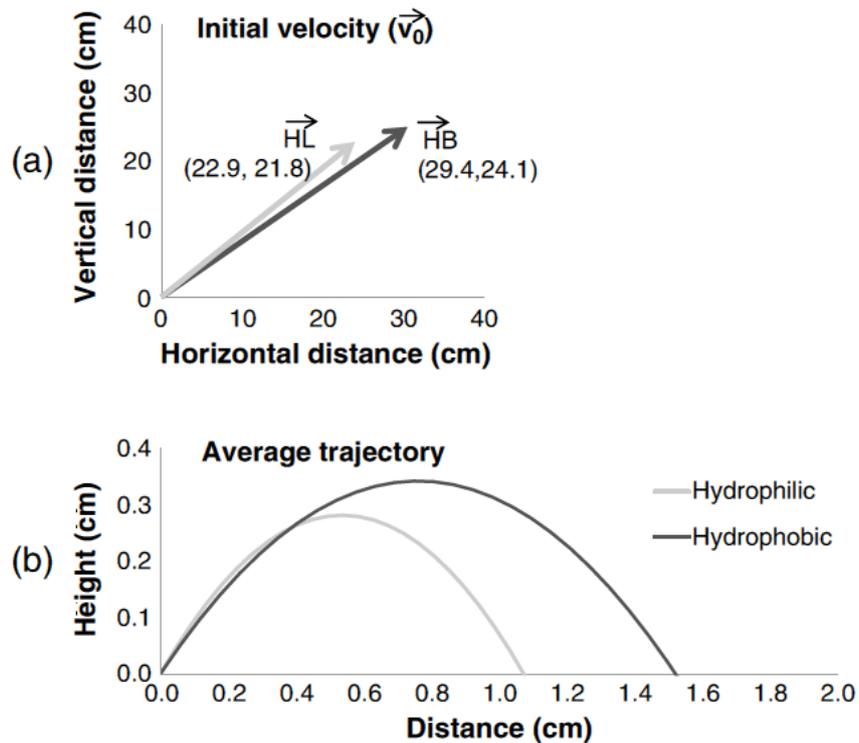


Figure 6. (a) The mean initial velocities of hydrophilic (HL) and hydrophobic (HB) particles in vector format and (b) the average trajectories of hydrophilic and hydrophobic particles landed further than 1 cm, which is calculated from mean ejecting angles and initial velocities.

#### d. Energy balance

The kinetic energy of an impacting drop (0.033 g), calculated from the velocity immediately prior to impact ( $2.67 \text{ m s}^{-1}$ ) was  $1.2 \times 10^{-4} \text{ J}$ . At a height of 40 cm the drop has a potential energy of  $1.3 \times 10^{-4} \text{ J}$  indicating an overall loss of  $0.1 \times 10^{-4} \text{ J}$  (~8%) arising from air resistance during its descent. The kinetic energy used to displace particles further than 1 cm was estimated from their number (Table I) and the mean velocity (Figure 5b) for hydrophilic and hydrophobic cases and was found to be  $3.1 \times 10^{-7} \text{ J}$  and  $9.3 \times 10^{-7} \text{ J}$  which is 0.26% and 0.78% of the kinetic energy of an impacting drop, respectively.

#### e. Shape and size of wetted perimeters

Following water drop impacts, the shape and size of the wetted perimeters formed in the particle beds, were compared. The smaller and more irregular perimeters were found in the hydrophobic cases (Figure 8). Image analysis confirmed this, showing lower circularity ( $0.64 \pm 0.11$ ) and roundness ( $0.86 \pm 0.04$ ) for the hydrophobic cases than for the hydrophilic ones ( $0.85 \pm 0.04$  and  $0.97 \pm 0.02$ ). The former also covered a smaller area ( $0.50 \pm 0.02 \text{ cm}^2$ ) than the hydrophilic cases ( $0.68 \pm 0.03 \text{ cm}^2$ ). Video frames showed that the water drop flattened after the moment of impact, and immediately penetrated the pores of the hydrophilic beds, whereas on hydrophobic beds, it re-collected and remained on the surface, covering an area with an irregular ridge of particles.

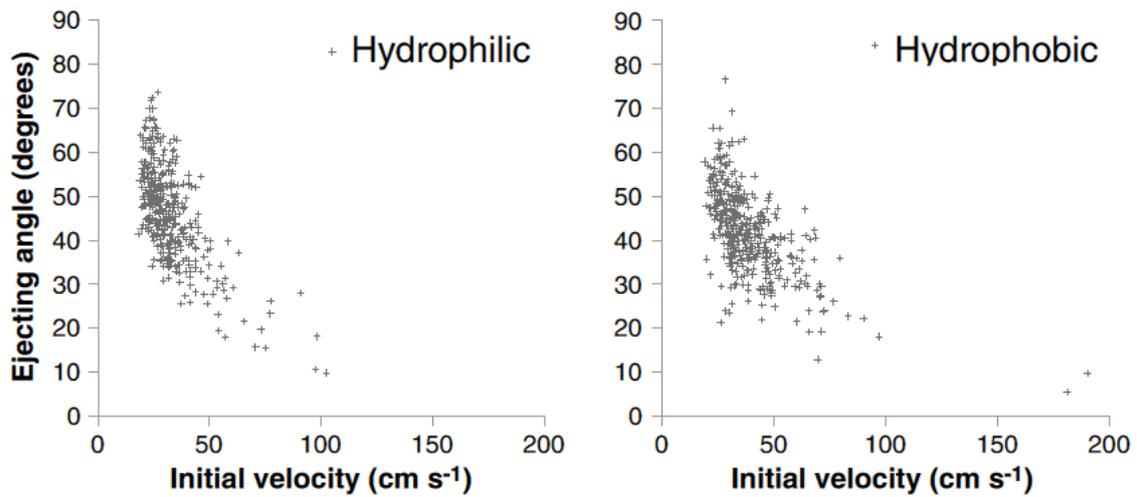


Figure 7. Scatter plots for ejecting angles versus initial velocities of hydrophilic (left) and hydrophobic (right) particles.

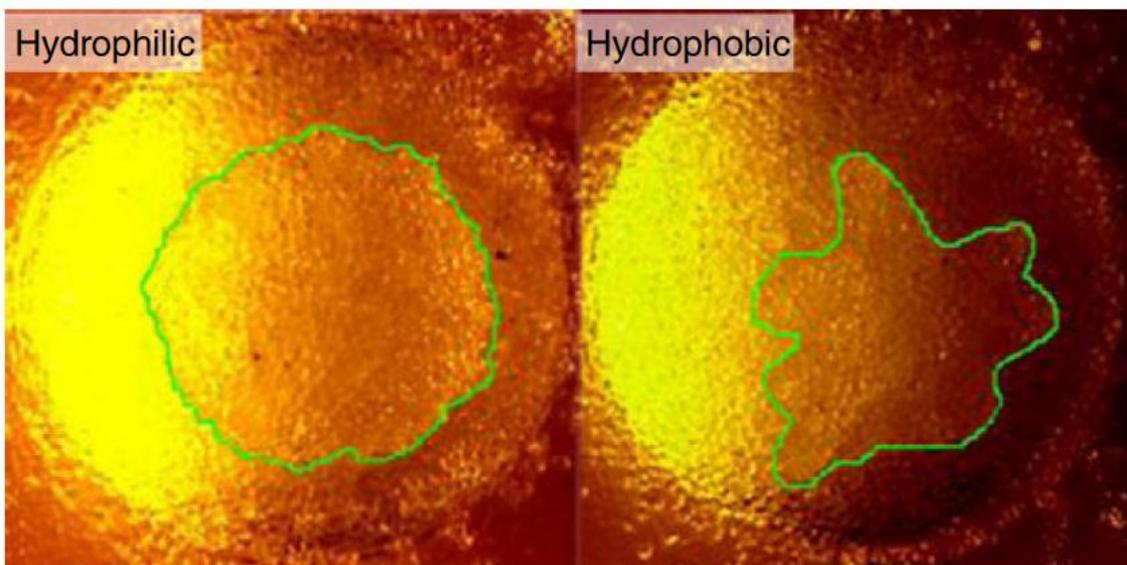


Figure 8. Shapes of wetted perimeters made by a water drop impact on hydrophilic (left) and hydrophobic (right) particle beds of 1.5 cm diameter.

### *Solid impact*

A chi-squared test of the distance distributions of particles arising from impacts with the solid plastic sphere with hydrophilic and hydrophobic beds (in which the former was taken as the reference for expected behaviour) indicated that there was a significant difference (test statistic  $\sim 53$ ,  $dF = 5$ ,  $p < 0.001$ ). It was observed that hydrophobic particles were over represented in two categories of displacement close to the impact site and were under represented in the two furthest from it. The mean numbers of particles displaced from replicate hydrophilic and hydrophobic beds were  $710 \pm 66$  and  $907 \pm 185$  respectively ( $dF = 5$ ,  $p = 0.076$ ) suggesting some evidence for a slight, but not significant difference in the numbers of particles ejected. Due to the skew in the distribution of displacements, the non-parametric Mann-Whitney U test was used. The outcomes demonstrate that there was a small, but nevertheless significant difference ( $p = 0.001$ ) in the median displacements with hydrophobic particles ejected to 1.71 cm and hydrophilic to 1.79 cm.

## Discussion

The water drops investigated in this study were of similar size (4.0 mm diameter) to that of high intensity storm within the range of natural raindrops (0.5 – 5.0 mm; Petersen *et al.*, 2011) but were of considerably lower impact velocity ( $\sim 2.7 \text{ m s}^{-1}$ ) compared to that of rainfall ( $\sim 9 \text{ m s}^{-1}$ ; Ahrens, 2007). This drop height is relevant to the effects of artificial irrigation or raindrops intercepted by a plant canopy at a similar height. The total energy used in transporting particles further than 1 cm, from the centre of the hydrophilic and hydrophobic beds  $\sim 0.26\%$  and  $0.78\%$  of the kinetic energy of the water drop respectively), falls within the range (0.1 to 10% of the impact energy of a solid projectile) previously reported for such ejections (Hartmann, 1985).

It is not presently known how water repellency may influence impacts of rain drops at terminal velocities where the larger kinetic energy may force enhanced interaction of the drop with the water repellent surface (e.g. see Reyssat *et al.*, 2006). In a similar context, it may not be straightforward to predict the impact of the typical rain drop size ( $\sim 2 \text{ mm}$ ) by its kinetic energy, as the size ratio between such a smaller water drop and the particles comprising the water repellent surface may also affect its interaction.

One factor which may have contributed the variation in the result is the shape of a falling drop at the moment of impact. Before reaching terminal velocity, a water drop oscillates between oblate (flattened) and prolate (elongated) shapes, whilst it tends to remain as oblate shape at terminal velocity (Epema and Riezebos, 1984). As prolate drops are 2-3 times more erosive than oblate ones (Riezebos and Epema, 1985), variation in the drop shape at the moment of impact can lead to variation between replications.

Ejecting angles of splashes has been often measured as a constant value obtained from the 'crown' shape in splash studies (Al-Durrah and Bradford, 1982; Pietravalle *et al.*, 2001) or from the 'ejecta cone' in planetary or powder sciences (Hartmann, 1985) rather than a distribution of individual splash angles. However, our results show that ejecting angles have a normal distribution in both hydrophilic and hydrophobic cases (Figure 5a). The initial velocities of ejections have previously been reported to have a gamma-distribution (Allen, 1988; Yang *et al.*, 1991; Pietravalle *et al.*, 2001), whilst the results of the present study show no clear evidence of such a distribution (Figure 5b). This difference on outcome may be due to the experimental conditions applied here (e.g. lower kinetic energy of impacting drop and use of homogeneous glass beads) or the limited sample excluding ejections within 1 cm distance.

The long and high trajectories of hydrophobic particles reported here (Figure 6b) seem to contradict previous research by Terry and Shakesby (1993), who observed higher trajectories for a hydrophilic soil than a hydrophobic soil. However, in their study, a layer of water formed on the top of the soil surface by accumulation of multiple drops, resulting in *splashings*, which consisted of water droplets and soil particles. In the present study, the consequences of the impact of an individual drop avoided the formation of a water layer and most particles were displaced in the form of *splash saltation* (Figure 4). This difference suggests that the mechanism of particle erosion may change once sufficient precipitation

has fallen to form a thin and continuous water layer – a transition whose duration may vary considerably with meteorological conditions and where a protracted increase in relative humidity may serve to enhance soil repellency to liquid water (Doerr *et al.*, 2002). On slopes or irregular soil surfaces, topography could discourage the formation of substantial and continuous surface water films.

The efficiency of splash erosion may be described in terms of numbers of ejected particles, their mass and the distances over which they are transported, in a similar manner as wetness is described in terms of liquid splashes (Allen, 1988). The results of this study suggest that particle hydrophobicity seems to increase the distance of ejections to a greater extent than it increases the number of particles ejected. As shown in Table I, the difference in the total number of scattered particles were insignificant between the two groups, whilst the number of particles travelled further than 1 cm were significantly different, resulting in the significant difference in the median distances.

The number of particles ejected (i.e. the total amount of splash loss) at a given kinetic energy is affected by shear strength (Al-Durrah and Bradford, 1981; Brunori *et al.*, 1989) and/or the aggregate stability (Farres, 1987) of the soil. These mechanical characteristics were standardized in this study, using glass beads of the same size and shape. This may explain the reason why total amount of displaced particles was less affected by hydrophobicity (Table 1).

As the distance of an ejection is influenced by both the initial particle velocity ( $v_0$ ) and angle of ejection ( $\theta_0$ ), the question arises as to how important either of these parameters may be in reflecting the differences observed between these particle beds. The results suggest that the initial angle does not seem to play a major role here. The longest trajectory arising from  $\theta_0 = 45^\circ$  is more within the range measured for the hydrophilic particles ( $\theta_0 = 46.3 \pm 10.9^\circ$ ) than for the hydrophobic ones ( $41.9 \pm 9.7^\circ$ ). However, despite the lower angles for the latter, their higher  $v_0$  ( $38.7 \pm 16.5 \text{ cm s}^{-1}$ ) resulted in longer and higher mean trajectories (Figure 6b). Therefore,  $v_0$  seems to be the main factor contributing to the relatively long displacements and may reflect the dynamic behaviour of the drop.

The remaining concern is the mechanism causing the difference in  $v_0$  between these two types of particles. Is it possible that hydrophilic particles transport a surface film of water, drawn from the impacting drop, which enhances their inertia, whereas the hydrophobic particles do not? This question warrants some discussion:-

a. Effective (wet) particle mass,  $m$

As the chemical modification of the particles to render them hydrophobic is unlikely to cause a significant change in their masses, the mass of water ( $m_w$ ) necessary to provide a hydrophilic particle with an equivalent kinetic energy to that of a (dry) hydrophobic particle of mass  $m_{HB}$  is:-

$$m_{HL} = m_{HB} + m_w \quad [7]$$

where,  $m_{HL}$  is the mass of the wetted hydrophilic particle.

Assuming the same kinetic energy for an ejection of each particle type, and as the mean initial velocity  $v_0$  is  $32.4 \text{ cm s}^{-1}$  for hydrophilic and  $38.7 \text{ cm s}^{-1}$  for hydrophobic particles, the following ratio between  $m_{\text{HL}} : m_{\text{HB}}$  is obtained:-

$$m_{\text{HL}} : m_{\text{HB}} = v_{\text{OHb}}^2 : v_{\text{OHL}}^2 = 1.43 : 1 \quad [8]$$

This means, that to account for the difference in trajectories, a hydrophilic particle requires an adsorbed water layer of water equivalent to 43% of its dry mass. Assuming that this water forms a uniform spherical shell around the particle, its comparatively low density ( $1 \text{ g cm}^{-3}$ ), in relation to that of the particle ( $2.46 \text{ g cm}^{-3}$ ), increases the particle radius by 30% ( $\sim 57 \text{ }\mu\text{m}$ ). If all the ejected particles ( $357 \pm 91$ ) are imparted with an adsorbed water layer, then a substantial mass and proportion of the impacting water drop ( $0.012 \pm 0.003 \text{ g}$ ,  $36 \pm 9\%$  respectively) is displaced with the particles. This enhanced size was not readily detectable within the video images (Figure 4) and it is therefore unlikely that this mechanism is significant, though it could make a contribution. However, this may be difficult to quantify as this water may readily evaporate and/or partially separate from an ejected particle during flight.

#### b. Dissipation of kinetic energy, $KE$

When a drop impacts on the surface of a particle bed, the adjustment in shape as it squashes into an oblate spheroid (or pancake) consumes energy as its surface area is enlarged (de Gennes *et al.*, 2003). Water may penetrate the pores of a hydrophilic bed more readily than those of a hydrophobic bed. The drop on the latter tends to reform as a sphere and so recovers some of the energy expended in the temporary expansion of its surface to a greater extent than the drop on the former (where particle surfaces may remain wetted). Both mechanisms discussed above could contribute to the difference in  $v_0$  observed here, however, this explanation has to remain speculative until confirmed experimentally in future work.

The relationship between the initial velocities and ejecting angles (Figure 7) shows that there is a wider range in the distribution of hydrophobic particle ejection velocities (Figure 5b) and a cluster of high velocities in the region  $50 < v_0 < 100 \text{ cm s}^{-1}$  corresponding with  $20 < \theta_0 < 40^\circ$  in the hydrophobic case. A notable finding is that the highest velocities were only observed at low  $\theta_0$  in both hydrophilic and hydrophobic cases. As the drop hits a hydrophobic bed, it will not penetrate the pores but tend to spread laterally driving particles ahead of its expanding boundary, so, although it may recover some energy on contraction of its surface, the initial repulsion from the surface and pores may account for the predominance of low values of  $\theta_0$ .

The ability of the drop to vertically penetrate the pores of the hydrophilic beds will detract from the volume available to spread laterally over the surface reducing the energy that it may impart to the particles in its path. This may contribute to the significant difference in the shape of the distributions of particle displacement (detected in the chi-squared test, but not so readily evident in Figure 3) and the fewer particles displaced from the hydrophilic beds (Table I).

The results of solid impact test show insignificant difference in the numbers of particles ejected between hydrophilic and hydrophobic particles beds, suggesting that there is no significant difference in particle cohesion within the beds under the condition of this experiment. The significant difference in the number of particles ejected caused by impact of a water drop, on the other hand, suggests an effect associated with the interactions of an impacting water drop with the individual particles and the particle bed within the impact zone.

A small, but significant, difference in the distribution of particle ejections by a solid impact between hydrophobic and hydrophilic particle suggests that the dynamic response of the water drop on collision with the surface of the beds can be, to some extent, influenced by the cohesion between the particles. Such cohesion may arise from the formation of water capillaries at or near particle/particle contacts in a humid atmosphere and would provide stronger cohesion in the hydrophilic particle beds (de Gennes *et al.*, 2003).

Although splash erosion has been considered to be minor contributor to soil transport in general, it may play an important role in the initial stages of the process (Kinnell, 2005). The results of this study suggest that initial splash detachment may be more influential in the case of water repellent soil, especially as water repellency is often associated with dry conditions (Bond and Harris, 1964; DeBano, 1971; Witter *et al.*, 1991; Ritsema and Dekker, 1994; Doerr and Thomas, 2000). This effect can be considerable where water repellent soil surfaces are exposed in areas suffering fires that remove the covering vegetation and where the (usually hydrophilic) ash layer has been removed by wind (Blong *et al.*, 1982) or previous water erosion (Wells, 1987). Another example can be seasonally dry areas where the onset of rainfall is slow and limited such that uniform continuous surface films of water may not form, but nevertheless the surface may be disrupted. Also the local topography, as formed by ploughing, may also provide opportunities for significant enhancement of splash erosion loss (Furbish *et al.*, 2007).

The ratio of median distance of ejections (hydrophilic: hydrophobic of 1:1.1) obtained in the present study suggests that the efficiency of particle transportation can be significantly higher, resulting in an increase in net erosion through the accumulation of this difference. This difference can also affect the range of redistribution of silt and clay by eluviations through macro pores, which is enhanced by drop impact (Biolders and Grymonprez, 2010).

## **Conclusions**

This study investigates the impact of hydrophobicity of particles on splash behaviour by isolating the effect of an individual drop impact. The results demonstrate that hydrophobicity significantly affects the splash detachment of particles even at the initial stage before a water film or wet soil crusts are formed by accumulation of water drops.

In comparison with hydrophilic particle beds, hydrophobic beds appear to be more susceptible to initial splash erosion arising from impacts of water drops at low kinetic energy ( $\sim 1.2 \times 10^{-4}$  J). The susceptibility can be described in terms of both greater numbers of particle ejections and enhanced distances of hydrophobic particles from their beds.

The greater distance of ejections appears to arise from larger initial velocities at angles below 45°, suggesting that hydrophobic surfaces direct a vertical drop parallel to the surface on impact providing an impulse to the particles along the surface rather than as a vertical rebound following intrusion into pore space (as likely in the hydrophilic case). The quantitative difference of splash behaviours between hydrophilic and hydrophobic particles, at the scale reported here, is ~10%.

Impact studies of solid plastic spheres delivering similar kinetic energy on impact as the water drops reveal only small differences in response of hydrophilic and hydrophobic beds, suggesting that water capillary forces provide negligible cohesion within the hydrophilic beds. This finding confirms that the main cause of the different behaviour is due to the interaction of the impacting water drop with particles.

It is likely that the fundamental findings obtained here for idealised model 'soil' material apply, at least to some degree, to loose sandy or aggregate-rich soils subjected to low velocity water drop impacts. The results suggest that for water repellent soils a greater net particle displacement can be expected. For terrain with substantial topography (i.e. hillslopes or ridges in cultivated terrain), a greater net downslope movement and hence net erosion of particles would be expected for soils exhibiting water repellency.

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