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Experimental characterisation of the impact of temperature and humidity on the breakdown of soil water repellency

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Abstract

Soil water repellency is a widespread phenomenon, the implications of which can manifest themselves as a reduction in infiltration and retention capacity as well as affecting hydrological and geomorphological processes. Water repellency is, however, a transient process which decays with time. Therefore the consequences of water repellency are only of concern during the timescale at which it persists. Water repellency and its breakdown are dependent on a number of factors including environmental factors such as temperature and humidity. With increasing concerns over future climatic changes it may become increasingly important to understand how environmental conditions impact upon water repellency. This study aimed to characterise the influence of temperature and humidity on the breakdown of water repellency as characterised by the change in apparent contact angle with time for a range of model and natural soils. The samples used in this study consisted of sands treated with stearic acid at concentrations of 0.1g kg⁻¹ - 5g kg⁻¹, two naturally repellent dune sands from Suffolk (East of England), one sampled from under vegetation and one bare soil; and two composts, one composed of green waste and one composed of a mix of green and catering meat waste. Contact angle measurements were carried out on a monolayer of each material sprinkled onto double sided tape attached to a microscope slide. A drop of water was placed onto the surface and the apparent contact angle was measured using Image J software with a drop analysis plugin. Temperature was controlled using a cooled incubator with a temperature range of 0-50°C and humidity was controlled using a purpose designed enclosed box setup in which humidity was raised by adding water to the box and lowered using silica gel desiccant.

Results showed the stearic acid treated sands decayed with time and there was a significant increase in contact angle with stearic acid concentration which persisted for the duration of the test. The model of Starov (2010) and Lee (2008) described the contact angle decay of

these samples. This could be a useful tool for assessing the consequences of repellency in the future. The stearic acid treated sands showed a significant increase in contact angle with relative humidity at 10 and 20°C. These differences diminished with increasing temperature. The changes in apparent contact angle was not as clear but again showed less difference at higher temperatures. In all samples it was of note that despite the changes in initial and final contact angles there was no significant change in the rate at which the contact angle decayed. Whilst relative humidity was kept constant in this study, the actual amount of water vapour in the air or absolute humidity is a function of temperature. Therefore results were also examined in terms of absolute humidity and showed an increase in apparent initial contact angle with absolute humidity. However, above 15g/m³ there was no significant effect of absolute humidity on any sample. The interactions of temperature and humidity are complex and so it may be more appropriate to consider absolute rather than relative humidity when trying to examine differences between laboratory or field samples.

Keywords: Soil water repellency, contact angle, absolute humidity, infiltration

Introduction

Water repellency refers to a reduction in soil wettability and is a manifestation of low solid surface free energy of the soil particles resulting in weak attractive forces between soil and water (Ojeda et al., 2010). The implications of water repellency are widespread and include a reduction in infiltration (Wallach and Graber, 2007) and water retention capacity (Naasz et al., 2008). This can have hydrological and geomorphological consequences (Cofield et al., 2007; Doerr et al., 2000) such as enhanced runoff and erosion (Leighton-Boyce et al., 2007), subsurface preferential flow and irregular wetting patterns which can lead to accelerated

leaching of solutes (Bauters et al., 2000; Doerr et al., 2000; Ferreira et al., 2000; Quyum et al., 2002), therefore, affecting biochemical processes and soil functions (Deurer et al., 2011).

Soil water repellency is a widespread phenomenon occurring in a variety of soil types, environments and climatic conditions throughout the world (Doerr et al., 2007; Deurer et al., 2011). It is, however, a transient condition and prolonged contact with water will commonly cause repellency to breakdown or disappear with time due to changes in surface properties (Doerr and Moody, 2004). Some of the reasons suggested for the breakdown of repellency include changes in surface free energy associated with either disruption of mineral and organic molecule bonds or thin films of water and the reorientation of organic molecules or reconfiguration of amphiphilic molecules (Doerr et al., 2000; Regalado and Ritter, 2005; Arye et al., 2007). Therefore, the impact of water repellency on hydrological functions is not solely determined by the degree of repellency but is also strongly dependent on its temporal dynamics. Hence the hydrological impacts of soil water repellency will be dependent on whether it persists throughout, or breaks down within the duration of a hydrological event (Doerr et al., 2000).

The water repellency of a surface is dependent on many factors including those relating to the surface itself (roughness, chemical heterogeneity, moisture content) and those relating to environmental conditions such as temperature and humidity (Dekker et al., 1998, 2001; Shirtcliffe et al., 2006). The effect of environmental conditions on water repellency becomes apparent when considering discrepancies in results obtained in the laboratory and field (Doerr et al., 2002) but may also become of increasing concern in light of future climatic changes. The seasonality of water repellency has already been noted, for example where rainfall has been found to wet some soils more readily in summer months than winter months (Doerr et

al., 2000). Increasing frequency of extreme climatic events (rainfalls and droughts) in the future could cause soils, which were previously considered wettable to exhibit some degree of repellency (Dlapa et al., 2004). Therefore an understanding of the impact of environmental conditions on water repellency and on the breakdown of water repellency needs consideration. Data on the influence of humidity on repellency is limited but Doerr et al. (2002) show that repellency increases with increasing ambient relative humidity for a wide range of dry soils even for short exposure times. Leelamanie and Karube (2008) also showed an increase in repellency with increasing relative humidity in stearic acid treated sands for exposure times of 20-22 hours. It is, however, worth noting that relative humidity, which is defined as the ratio of the partial pressure (or density) of water vapour present in the atmosphere to the saturated vapour pressure (or density) of water at a given temperature, is dependent on temperature and that relative humidity will vary with temperature even if the actual or absolute humidity in the atmosphere remains constant. Hence, relative humidity may not provide the best measure to assess the influence of humidity on repellency. Temperature has also been shown to affect soil water repellency but much of the work to date has focused on temperatures at the higher end of the ecologically relevant soil surface temperature range to characterise the effect of laboratory sample drying temperature (~30-105°C) (Dekker et al., 1998; Goebel et al., 2011) or heating temperature (several hundred °C) in studies on the impact of wildfires on repellency (Doerr et al., 2004). These studies suggest that repellency increases with temperature until repellency disappears at very high temperature (~ 250°C) due to the volatilisation or combustion of organic compounds. There is however an interaction between the effects of temperature and humidity and Diehl and Schaumann (2007) show that, under high relative humidity (99.9%) and lower ambient temperatures, repellency can decrease with temperature (5-30°C). It is therefore likely that

results on the effect of temperature will be affected by humidity if the latter is not held constant.

To address this research gap, this study aimed to characterise the influence of temperature and humidity under controlled conditions on the temporal dynamics of soil water repellency characterised by apparent contact angle changes with time for a range of model and natural soils consisting of sands treated with stearic acid, naturally water repellent soils and composts.

Materials and methods

Soil materials

Two types of soil materials were used for this study. Sands treated with stearic acid were used as a model material with a controlled degree of repellency along with some naturally repellent soil materials. Stearic acid is a fatty acid containing amphiphilic molecules which, upon contact with water, can reorientate, thus altering the wettability of the surface (Yiannos, 1962). Stearic acid was dissolved in diethyl ether and gently mixed into fine sand at concentrations of 0.1, 0.3, 0.4, 0.5, 1.0 and 5.0 g kg⁻¹ and left to evaporate in a fume hood overnight. The influence of stearic acid concentration on contact angle was investigated using all of these concentrations at 20°C and 50% relative humidity; whereas the impact of temperature and relative humidity on contact angle was examined for only three concentrations of stearic acid (0.1, 0.5 and 1.0 g kg⁻¹) as described below. The natural soils consisted of two dune sands obtained from sand dunes in Suffolk, East of England (Latitude 52.21, longitude 1.62), one obtained under dune grass vegetation and one with no vegetation (hereafter referred to as dune sand 1 and dune sand 2) and two composts, one composed of green waste and one composed of approximately 50% green waste and 50% catering meat

waste obtained from TEG Environmental Ltd, Perth, Scotland (hereafter referred to green and meat compost respectively). All samples were air dried at 40°C for 24 hours and sieved to 2 mm prior to use.

Contact angle measurement

Preparation of the samples for contact angle measurements followed the method of Bachmann et al. (2000). Air-dried sieved material was sprinkled onto hydrophobic adhesive tape attached to a glass microscope slide, to prevent drop contact with the tape and spreading, and tapped to remove excess loose material and form a monolayer of soil. A fixed monolayer of material was needed due to the complications in contact angle interpretation posed by drop infiltration into samples (Bachmann et al., 2000; Hilpert and Ben-David, 2009) and the risk of the marble effect which will impact on contact angle when loose repellent particles migrate around drops eventually encasing them as liquid marbles. Examples of this effect can be seen in Fig. 1 as well as McHale et al. (2007).

Similarly to Diehl and Schaumann (2007), the changes in apparent contact angle with time due to repellency breakdown were measured by photographing a 20 µl drop of deionised water placed on the surface of the soil at 30 second intervals for 15 minutes. In order to assess the potential influence of evaporation on contact angle during this period of time, tests were also carried out with glass beads treated with dichlorodimethylsilane for each temperature and humidity treatment. These treated glass beads had stable water repellency and thus any changes in contact angle over time were considered to be the result of evaporation only. The changes in contact angle were less than 2% and the influence of evaporation was therefore considered to be negligible. The photographs were analysed using Image J software adapted with a specific contact angle drop plugin (Stalder et al., 2010). In

brief, the images were converted to grayscale images and the scale of the images was set by using the known size of the tip of the pipette used to deposit the water drop as a reference. A line was then matched to the drop profile before optimisation by the software to determine the contact angle. This uses an axisymmetrical drop shape approach, as opposed to a spherical cap assumption, where the influence of gravity and inertia is included, and a Young Laplace profile is fitted to give contact angle values (Stalder et al., 2010).

In this study, all apparent contact angle measurements were carried out on triplicate samples.

Temperature and humidity control

Temperature was controlled using a cooled incubator with a temperature range of 0-50°C. Humidity was controlled using an enclosed box setup (Fig. 2), whereby humidity could be raised by adding water to the bottom of the box or lowered using a silica gel desiccant. A hole was machined in the side of the box and the camera lens fitted with an o-ring to prevent changes in humidity during the experiments. The pipette used to deposit the drop on the sample was held by a clamp at a standard height of 1 cm above the sample surface. The sample was held on a laboratory jack. Temperature and relative humidity were monitored throughout all tests using a Klima Logger thermo-hygro station (TFA Dostmann). Tests were carried out at 10° C, 20° , 30° C and 40° C (all $\pm 2^{\circ}$ C) at relative humidity values of 20% ($\pm 5\%$), 50% ($\pm 6\%$) and 98% ($\pm 2\%$) for the sand samples treated with stearic acid at concentrations of 0.1, 0.5 and 1.0 g/kg, the two composts and two dune sands. As mentioned above, relative humidity is equal to the ratio of actual vapour pressure to saturated vapour pressure. Since the saturated vapour pressure is temperature dependant, relative humidity does not provide a direct measure of changes in the amount of water vapour in the gas phase (water vapour density) unless temperature is constant. Hence, in order to be able to compare the influence of

humidity under the different temperature regimes, absolute humidity was also used in this study. Absolute humidity is expressed as the mass of water vapour per unit volume of the gas phase and is therefore a measure of the water vapour density. Absolute humidity was calculated using British standards BS1339-1 (2002) and BS1339-3 (2004) and values for each temperature and relative humidity regime can be seen in Table 1.

Statistical analysis

Statistical analysis was carried out using Statistica 9 (Statsoft, 2010). A one way ANOVA was used to assess the effect of particle size and factorial ANOVA was used to analyse the effect of particle size distribution. The breakdown of water repellency was analysed using a combination of repeated measures ANOVA to assess the impact of time and factorial ANOVA to assess the impact of initial and final contact angle and the breakdown model parameters. The effect of temperature and humidity was considered using factorial ANOVA's. The differences in the timescale of breakdown in the soil material prevented the use of repeated measure ANOVA for the soil materials and so only factorial analysis was carried out for the soils.

Modelling of apparent contact angle decay during the breakdown of repellency

Few studies have assessed the breakdown of water repellency in soils through measurements

of changes in apparent contact angle with time (Wessell., 1988; Diehl and Schaumann, 2007).

However, these studies and others on various types of porous media such as chemically

hydrophobised sands (Leelamanie and Karube, 2009), clay colloids (Shang et al., 2010)

pharmaceutical powders (Muster and Prestidge, 2002) and polystyrene (Wang et al., 2005)

have all shown a clear functional relationships between apparent contact angle and time. As

shown in some of these studies and in the results presented hereafter, this relationship can be

represented by an exponential decay of the cosine of the advancing apparent contact angle of the form $cos\theta(t) = A + B e^{-Ct}$, where A, B and C are positive constants. This can be written as:

$$\cos\theta(t) = \cos\theta_{\infty} + (\cos\theta_{0} - \cos\theta_{\infty})e^{-t/\tau}$$
 [1a]

which is the same expression as that derived theoretically by Starov et al. (2000) and Lee et al. (2008) for the spreading of aqueous surfactants solutions onto initially hydrophobic surfaces:

$$\cos\theta(t) = \cos\theta_0 + (\cos\theta_\infty - \cos\theta_0)(1 - e^{-t/\tau})$$
 [1b]

where θ_0 and θ_∞ are the initial and final contact angles, respectively; and τ is a characteristic time scale of droplet spreading. Spreading of aqueous surfactant drops onto hydrophobic surfaces is due to surfactant adsorption at both the liquid–solid and liquid–vapour interfaces and transfer of surfactant molecules onto the solid–vapour interface in front of the drop. Although these three processes contribute to spreading and a decrease in contact angle, according to Starov et al. (2010) the spreading kinetics are mainly controlled by the slow transfer onto the solid–vapour interface and changes in the instantaneous solid–vapour interfacial tension in front of the three-phase contact line. Although the current study does not deal with aqueous surfactants, the processes involved in a water drop spreading over a hydrophobic surface, which may contain amphiphilic molecules capable of overturning, are considered to be similar (Starov et al., 2010) and thus this model may provide a useful empirical tool for assessing the breakdown of repellency. Aqueous drop spreading is due to the rearrangement of the surface layer when in contact with a polar liquid, such as the

overturning of amphiphilic molecules, and the lowering of the interfacial free energy as a result of the adsorption of polar groups at the surface. It is argued, however, that this rearrangement also takes place ahead of the advancing three-phase contact line due to interaction between neighbouring amphiphilic molecules and that this is the limiting process of the spreading kinetics whereas under the main surface of the drop equilibrium occurs rapidly (Starov et al., 2010).

Results and discussion

Effect of stearic acid concentration on water repellency breakdown

This study found that for all concentrations of stearic acid used (0.1 to 5.0 g/kg) the apparent contact angle, at 20°C and 50% relative humidity, showed a clear continuous decay with time (Fig. 3). There was an increase in contact angle with stearic acid concentration which was apparent for the duration of the tests and resulted in significantly larger initial (p<0.03) and final (p<0.04) contact angles between the sand treated with 5.0 g kg⁻¹stearic acid compared with all other concentrations. For the sands treated with stearic acid at concentrations below 1 g kg⁻¹the initial and final contact angles were not significantly different from one another with the exception of the sand treated with 1.0 g kg⁻¹of stearic acid which had a significantly larger initial and final contact angle than the sand treated with 0.1 g kg⁻¹(p<0.04). In soils the structural composition and arrangement of molecules and functional groups is thought to be more important than the amount or concentration of organic substance in determining the degree of repellency (Diehl and Schaumann, 2007). The results of the current study, in part, reflect this with there being little significant alteration in repellency up to 1 g kg⁻¹concentration of stearic acid. However, above 1 g kg⁻¹the alteration in repellency becomes significant.

The transient nature of water repellency makes characterisation difficult. The model of Starov et al. (2010) and Lee et al. (2008) could describe contact angle decay (Fig. 3) for all stearic acid concentrations. This simple equation gives an indication of the nature of water repellency decay with time and could be a useful tool when assessing the potential consequences of repellency. The parameters used to fit this contact angle decay function (Equation 1) indicated a significant increase in the initial contact angle between the sand treated with stearic acid at 5.0 g kg⁻¹ and all other concentrations (p<0.01) except 0.5 g kg⁻¹ ¹(Table 1). Unlike the measured initial contact angle there was a significant increase in the fitted initial contact angle of the sand treated with 1.0 g kg⁻¹ and all other concentrations (p<0.03) except the sand treated with 0.4 g/kg. Furthermore, there was a significant increase in the fitted initial contact angle between 0.5 g kg⁻¹ and all other concentrations except 5.0 g/kg. The final fitted contact angle was significantly larger in the sand treated with 5.0 g kg ¹of stearic acid compared to the 0.1, 0.4 and 1.0 g kg⁻¹concentrations (p<0.04). The equation parameter, τ , which gives an indication of the time for overturning of amphiphilic compounds at the three phase contact line, increased from 183 seconds in the sand treated with 0.1 g kg⁻¹ ¹to 707 seconds in the sand treated with 5.0 g kg⁻¹but the differences were not significant. The increase in τ with stearic acid concentration could indicate an increase in the time for amphiphilic molecules to overturn in these samples. As such this could indicate that with a greater degree of repellency, persistence is increased. The temporal dynamics of repellency are crucial in assessing the risk posed by hydrological events (Doerr et al., 2000). Therefore, if the degree of repellency causes an increase in the time taken for repellency to decay then the time in which the consequences of repellency are a concern is increased in those soils which are more repellent.

These results are similar to those of Leelamanie and Karube (2008), who found an increase in contact angle with stearic acid concentration and an exponential decay in contact angle with time. However, the maximum decay in contact angle observed in this study over 15 minutes was 20° which was half that observed by Leelamanie and Karube (2008) over the same period of time. This difference could be the result of environmental conditions as their experiments were carried out at the higher temperature and relative humidity of 25°C and 80%, respectively.

Effect of temperature and humidity on water repellency breakdown

Environmental conditions play a complex role in the repellency of soils and the implications of this could be extensive. With global temperatures expected to rise in the future and an increased likelihood of extreme climatic events (Goebel et al., 2011; IPCC, 2013, the distribution and degree of repellency may be altered.

In the stearic acid treated sands there was a significant increase in contact angle with relative humidity at 10°C and 20°C for all samples (p<0.01). At 30°C the effect of relative humidity had diminished and was only significant between 20% and 50% relative humidity and between 98% and both 20% and 50% relative humidity (all p<0.03). By 40°C this effect had diminished further and was only significant between the 20% and 98% relative humidity (p<0.05). This temperature and relative humidity effect was most apparent in the 0.1 g kg⁻¹ treated sand (Fig. 4) but showed a similar trend in the 0.5 g kg⁻¹ and 1.0 g kg⁻¹ treated sands (data not shown).

In addition to the stearic acid treated sands, several soils and composts were considered. The effect of temperature and relative humidity on the breakdown of repellency in the soils and

composts was less clear. Fig. 5 shows changes in apparent contact angle for the meat compost. Similar trends were observed in the two dune sands whereas in the green compost repellency broke down very rapidly (data not shown). As with the stearic acid treated sands there was a significant increase in the initial contact angle with increasing relative humidity at 10 and 20°C, however, there was no significant increase in final contact angle. At 30 and 40°C the effect of relative humidity was negligible in all four soil materials and showed no significant differences.

Despite the significant differences observed in contact angles at different relative humidity and temperature, the rate of decay of the contact angle remained unaffected by the different treatments. This was true for all samples except the green compost for which the rate of decay in apparent contact angle decreased with increasing relative humidity. However, the rate of contact angle decay in the green compost was much higher than in the other materials.

Water repellency has been suggested to increase with increasing relative humidity (Wallach and Graber, 2007; Doerr et al., 2002; Leelamanie and Karube, 2009). This was initially attributed to increasing microbial activity (Jex et al., 1985). However, it was shown that an increase in repellency could also be observed when equilibration at high relative humidity was carried out over short periods of time (Doerr et al., 2002) but this increase in repellency did not continue when the equilibration time was increased as would be expected if microbial activity was the cause (Wallach and Graber, 2007). The reduction in interfacial tension between the solid and vapour phases and the release in energy associated with water vapour adsorption (Bachmann and Van der Ploeg, 2002), the expansion of organic matter into the pore space obstructing the passage of water in the soil (Doerr et al, 2002), the presence of thin films of water and thus reduction in surface energy of the solid (Goebel et al., 2004) and

immobilisation of certain fatty acids (Graber et al., 2009) with increasing relative humidity have been suggested as potential causes of the increasing in repellency. However, if adsorption continues to such a point that a continuous film of water is established over the entire surface, repellency may decrease at higher relative humidity (Shang et al., 2010).

It is also important to note that, if water vapour adsorption plays an important role, relative humidity may not provide the best measure of the influence of water vapour present in the atmosphere. In this study, while relative humidity is kept constant, the actual amount of water vapour present in the atmosphere or absolute humidity is increasing with increasing temperature. Fig. 6 shows the initial and final apparent contact angles plotted as a function of absolute humidity for the sand treated with stearic acid at a concentration of 0.1 g/kg. Very similar results we observed for the concentrations of 0.5 and 1 g kg⁻¹(data not shown). There was a clear initial increase in apparent contact angle with increasing absolute humidity; however, the initial and final apparent contact angles reached their maximum rapidly and remained approximately constant for absolute humidity >15 g/m³. In contrast, there was no significant effect of absolute humidity on the initial and final apparent contact angles of the four soil materials. This reflects the limited influence of humidity and temperature on the dynamics of repellency breakdown in these soils, which tended to be only significant for low relative humidity and low temperature.

It is not possible to separate the effect of temperature on the breakdown of repellency because absolute humidity is a function of temperature. However, the results obtained for the treated sands (Fig. 4 and Fig. 6) illustrate the confounding effect of temperature when compared to relative humidity. Increases in temperature tended to result in a decrease in apparent contact

angle and therefore repellency. Hence, whereas there was a clear effect of relative humidity on contact angle at lower temperatures, the effect at 40°C was negligible.

Studies of the effects of temperature on soil water repellency have typically focussed on higher temperatures associated with wildfires (Doerr et al., 2004; DeBano, 2000) or laboratory sample drying temperatures (Dekker et al., 1998; Goebel et al., 2011). Studies at lower temperatures have received less attention. Nevertheless, Diehl and Schaumann (2007) found repellency to decrease with increasing temperature between 5-40°C at high relative humidity. In contrast, Goebel et al (2011) found repellency to increase between 25-60°C. However their study was carried out to assess the effect of sample drying temperature and therefore repellency would have been affected by changes in moisture content as well as temperature. The apparent decrease in repellency with increasing temperature has been discussed in several studies and is suggested to be the result of a reduction in liquid surface tension with temperature or increased reactivity of molecules which can overturn or reorganise quicker at higher temperatures (Bachmann and Van der ploeg, 2002; Doerr et al., 2000; Graber et al., 2009). The discrepancies between the studies mentioned above is likely to be due to the compounding effects of temperature, humidity and soil moisture content and therefore all need to be controlled.

Conclusions

The breakdown of repellency in stearic acid treated sands and some soils and composts was characterised by an exponential decay of the cosine of the apparent contact angle with time.

There was an increase in apparent contact angle with increasing stearic acid concentration but the effect of concentration on the rate of repellency decay was not significant. This simple

exponential model could provide a useful tool to assess the persistence of water repellency and the environmental implications associated with it.

In general, there was an increase in repellency with increasing relative humidity. However, the influence of relative humidity decreased with increasing temperature and was not always significant at higher temperatures. This was because increases in temperature tended to have a confounding effect, resulting in a decrease in apparent contact angle. As a result absolute humidity had either no effect on repellency or its influence was limited to the low humidity range. Absolute humidity appears to be a more relevant indicator than relative humidity when assessing the influence of humidity over a range of temperatures.

It is also interesting to note that, although relative humidity and temperature had a significant effect on the apparent contact angle, it had, in general, little influence on the rate at which this contact angle decayed with time.

The alteration of the amount of water vapour in the air with increasing temperature, even at comparable relative humidity, may be of consequence to the underlying mechanisms of water repellency. Whereas an increase in water vapour and adsorption may result in an increase in repellency, if it is associated with a large increase in temperature, the degree of repellency may remain unchanged or even decrease. Humidity is thought to increase ahead of a rainfall event (Doerr et al., 2000). If the frequency of intense rainfall events increases in the future (Goebel et al., 2011; IPCC, 2013), the effect of humidity and temperature on water repellency and its breakdown, could become an important consideration.

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Table 1: Parameters of Eq. 1 used to model apparent contact angle decay in sands treated with stearic acidic

Stearic acid concentration (g/kg)	Initial contact angle (°)	Final contact angle (°)	τ (seconds)
0.1	88.7	76.9	183
0.3	90.9	77.8	334
0.4	92.2	73.5	706
0.5	94.9	73.1	454
1.0	96.8	75.4	706
5.0	105.2	85.3	707

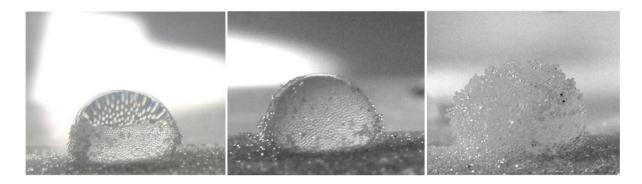


Fig. 1: Formation of a liquid marble on loose dichlorodimethylsilane-treated glass beads

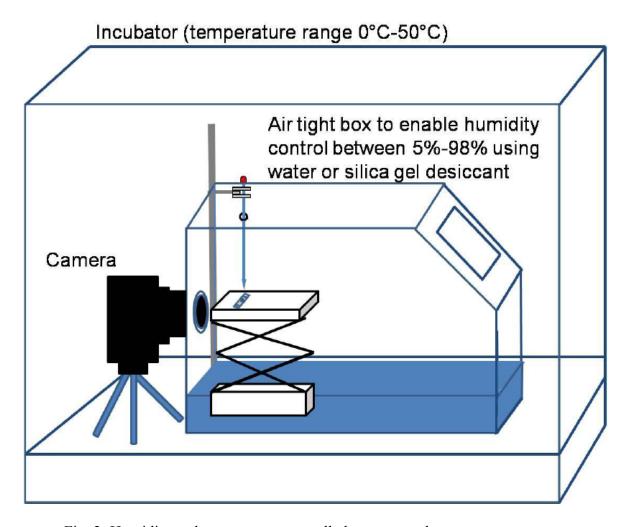


Fig. 2: Humidity and temperature controlled contact angle measurement setup

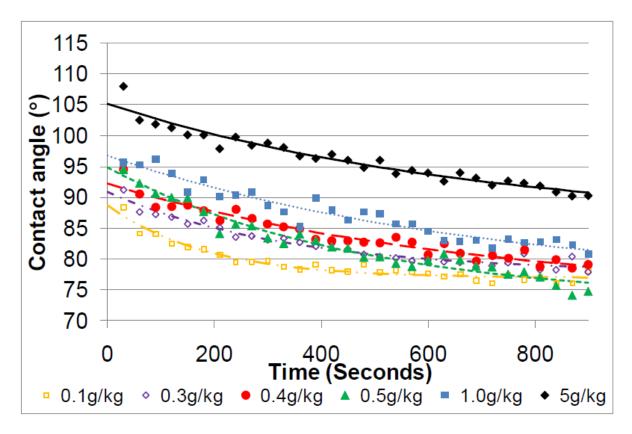


Fig. 3: Breakdown of repellency in sand treated with stearic acid

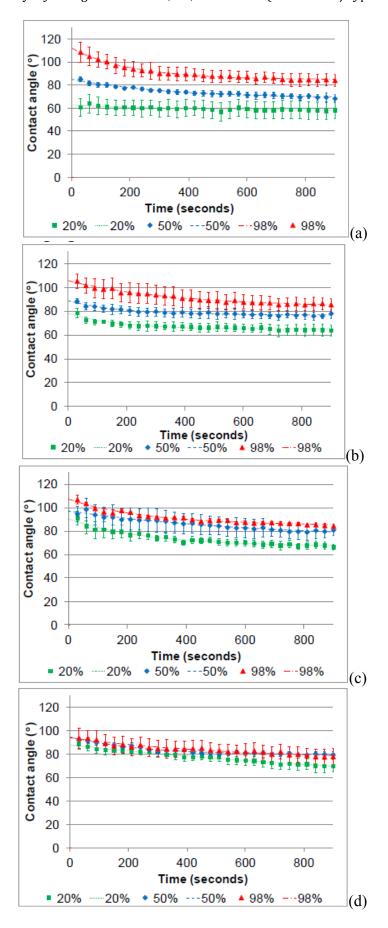


Fig. 4: Influence of relative humidity on contact angles at (a) 10, (b) 20, (c) 30 and (d) 40 °C in the sand treated with stearic acid at a concentration of 0.1 g/kg

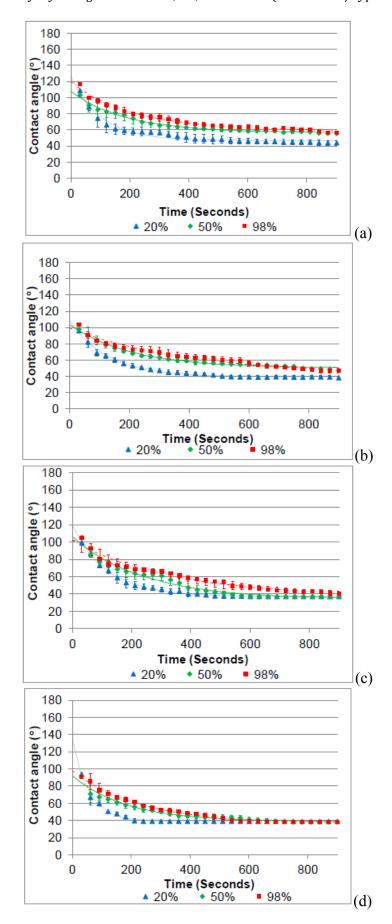


Fig. 5: Influence of relative humidity on contact angles at (a) 10, (b) 20, (c) 30 and (d) 40 °C in the meat compost

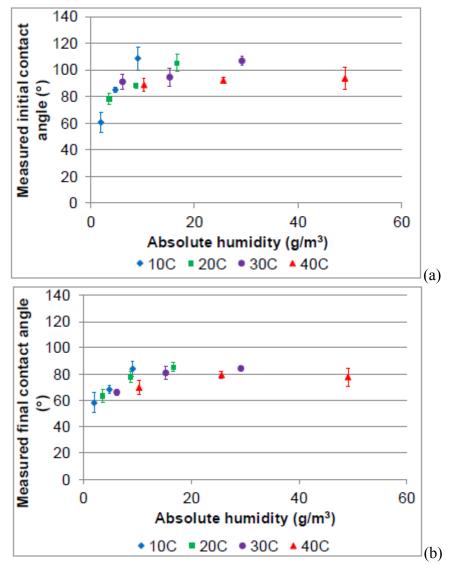


Fig. 6: (a) Initial and (b) Final contact angles measured as a function of absolute humidity for the sand treated with stearic acid at a concentration of 0.1 g/kg. Error bars represent standard error of the mean.