



Sessile drop contact angle as affected by the particle size and the drop size

Leelamanie, D.A.L.¹, Samarawickrama, U.I.¹ and Jutaro, K.²

¹Faculty of Agriculture, University of Ruhuna, Mapalana, Kamburupitiya, Sri Lanka.

²Faculty of Agriculture, Ibaraki University, 3-21-1 Chuo, Ami-machi, Ibaraki-ken 300-0393, Japan.

✉ leelamanie@yahoo.co.uk

Abstract

Soil water repellency is an important phenomenon that primarily affects the water infiltration rates in soils. The magnitude of soil water repellency can be expressed in terms of soil-water contact angle, in degrees. The purpose of this study is to examine the effects of particle size and drop size (drop volume) on the contact angle of a sessile drop. Silica sand and Glass beads were used to assess the effect of particle size, whereas Acryl, Vinyl tape, Adhesive tape, and hydrophobized silica sand were used to assess the effect of drop size, on sessile drop contact angle. Contact angle was directly measured using a digital microphotograph and calculated using equations using drop parameters as functions. Sessile drop contact angle decreased, from 60° to 20° in silica sand and from 76° to 45° in glass beads, with the increase in particle size from 19 to 250 µm and from 50 to 400 µm, respectively. Calculated contact angle of all the samples decreased continuously with increase in drop size from 5 to 50 µl, which was attributed to the changes in drop parameters with the change in drop size. Measured sessile drop contact angle decreased to a minimum with the increase in drop size up to 15 µl in Acryl, 20 µl in Vinyl tape, and 30 µl in Adhesive tape, and both hydrophobized silica sand samples. This was considered as a result of the decreasing importance in linear tension of a liquid drop with increasing volume. Measured contact angle increased again with increasing drop size up to 50 µl in all the samples, which might be because of the gravity interference to the contact angle after some level.

Keywords: sessile drop method, contact angle, water repellency

Introduction

A water repellent or hydrophobic soil is defined as “a soil that does not wet spontaneously when a water drop is placed on the surface” (Feng *et al.*, 2001). The primary effect of soil water repellency is the reduction of the water infiltration, which will increase the runoff and top soil erosion ultimately resulting in soil and land degradation. Water repellency affects soil moisture dynamics and leads to preferential flow through less repellent patches and macro pores (Wallis *et al.*, 1991; Wallis and Horn 1992; Feng *et al.*, 2001).

Water repellency appears on low-energy surfaces where the attraction between the molecules of solid and liquid interface is weak (Heslot *et al.*, 1990; Roy and McGill, 2002). Under natural conditions, high-energy soil mineral surfaces are often covered by films of low-energy organic compounds (Doerr *et al.*, 2000; Goebel *et al.*, 2004), forming water repellent surfaces. However, the presence of hydrophobic compounds does not necessarily always cause the repellency (Doerr *et al.*, 2005; Leelamanie and Karube, 2007).

Water repellency in soil can be expressed in terms of the degree, i.e., how much repellent, and the persistence, i.e., how long repellency would persist (Buczko *et al.*, 2002, 2006; Lachacz *et al.*,

2009). Many different techniques have been developed to measure the soil water repellency.

Persistence of soil water repellency can be measured using Water Drop Penetration Time (WDPT) test, which consists of measuring the time taken for the complete penetration of a water drop placed on the soil surface. Degree of soil water repellency can be expressed in terms of contact angle, surface free energy, ninety degree surface tension etc. The most commonly used two indices of characterizing the magnitude of water repellency are the liquid-solid contact angle and the WDPT, where the former is related to the degree and the latter is related to the persistence of water repellency.

Assessing the surface properties of soil particles is considerably difficult compared with that of plain and smooth surfaces. A direct measurement of the contact angle on soil is usually not possible due to the nature of soil surfaces. Bachmann *et al.* (2000a, 2000b) developed the modified Sessile Drop Method (SDM) for the direct measurement of the contact angle on soil. The SDM basically consists of measuring the contact angle of a small water drop placed on a monolayer of air dried soil. The range of contact angles that can be measured by the SDM is extended to values higher than 90°, even if pure water is used as the test liquid (Bachmann *et al.*, 2000b).

Sessile drop contact angle is known to be affected by various factors such as ambient relative humidity (Leelamanie *et al.*, 2008b) and soil texture (Leelamanie *et al.*, 2010). Soils contain different sizes of particles while different sizes of drops are been used for contact angle measurements. However, the effects of the particle size of sample and the size of the test drop on sessile drop contact angle has not been tested so far. The objective of this study is to examine the effects of particle size and drop size on sessile drop contact angle using different sizes of glass beads and hydrophobized silica sand.

Materials and Methods

Experimental setup

The model soils were prepared using fine silica sand (Tohoku Keisha Co., Yamagata, Japan) and glass beads (Thoshinriko Co., Ltd Tokyo, Japan) were used to assess the effect of particle size, whereas Acryl, Vinyl tape, Adhesive tape, and hydrophobized silica sand were used to assess the effect of drop size. The particle size distributions of silica sand and glass beads are given in Table 1. The experiment was conducted in a constant temperature room, with 25°C and 75±5 relative humidity. All the prepared samples were kept in a sealed container under 75% relative humidity maintained using saturated sodium chloride (NaCl) solution for 48 hours before the contact angle measurement.

Table 1: Particle sizes (diameter) of glass beads and silica sand used in the experiment

Glass beads (µm)	Silica sand (µm)
50 - 70	<38
70 - 100	38 - 53
100 - 150	53 - 75
150 - 200	75 - 106
200 - 400	106 - 150
	150 - 250

Sample preparation

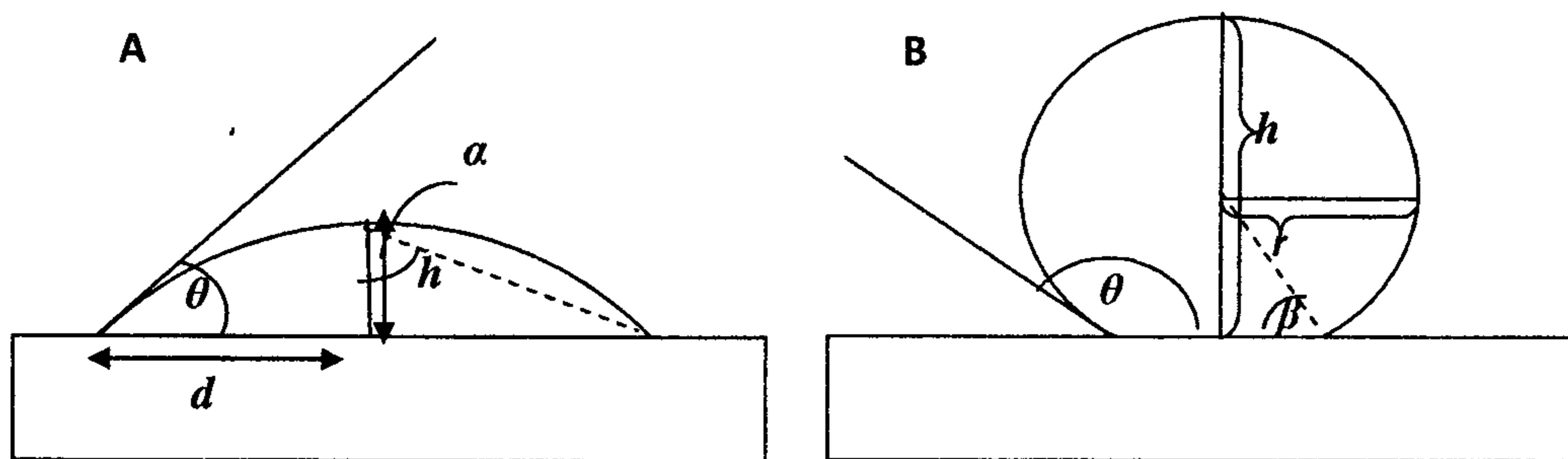
From each glass bead category 10 to 15 g were taken in to separate 100 ml beakers. Those were first washed using 0.5% neutral detergent for 30 minutes in an ultrasonic cleaner, and second using de-ionized water for several times to remove the detergent completely. The glass beads were allowed to be drained and dried in an oven at 105°C for 8 to 10 hours.

Silica sands were hydrophobized with 1 and 5 g kg⁻¹ of stearic acid. Stearic acid was dissolved in diethyl ether and mixed with the sand in a fume hood. Samples were left for 2 hours in the hood to allow the complete evaporation of diethyl ether and then kept for one day before fixing of the samples for the sessile drop contact angle measurement.

Sessile drop method

For the contact angle measurement of glass beads and hydrophobized silica sand, each sample, in three replicates, was sprinkled on a double-sided 1.5 cm × 1.5 cm adhesive tape pasted on a glass slide. Glass beads were pressed to the tape with a 400 g weight for 30 s, whereas silica sand samples were pressed to the tape with a 100 g weight for 10 s. The slide was tapped gently to remove the surplus particles. This procedure was repeated twice. The Vinyl tape and the adhesive tape were pasted on glass slides for the measurement of the contact angle. For contact angle measurement of acryl, ‘Acryl lenses’ were used.

Each sample was placed on the stage of a digital microscopic camera (Leelamanie *et al.*, 2008a) and a digital micro-photograph of the horizontal view of a drop of deionized water placed on the sample surface was taken within 1 s. For the assessment of the effect of particle size, a drop of deionized water with 10 µL volume was placed on the soil surface using a micro-pipette. For the assessment of the effect of drop size, drops of deionized water with 5, 10, 15, 20, 30, 40, and 50 µL were applied using two adjustable micro-pipettes. Using the micro-photographs, contact angle (θ) of each sample was directly measured and calculated as describe in the equations 1 – 4 and Figure 1.



(Annaka 2006)

Figure 1: Parameters for calculating contact angle (θ) when (A) $\theta < 90^\circ$ and (b) $\theta > 90^\circ$

For $\theta < 90^\circ$: $\theta = 180^\circ - 2\alpha$ (1)

$\tan \alpha = d / h$ (2)

For $\theta > 90^\circ$: $\theta = \beta + 90^\circ$ (3)

$\sin \beta = (h - r) / r$ (4)

Results and Discussion

Effect of particle size on sessile drop contact angle

Effect of particle size on sessile drop contact angle for glass beads and silica sand is presented in Figure 2. Contact angle obtained on the surface of glass slide was about 16°. The sessile drop contact angle for silica sand mixture was about 11°. However, the minimum contact angles observed for separated particle sizes in glass beads and silica sand were 45° and 20°, respectively. For both glass beads and silica sand, contact angle decreased with increasing particle size. In glass beads, contact angle decreased from 76° to 45° with increase in particle diameter from 50 to 400 μm . In silica sand, contact angle decreased from 60° to 20° with the increase in particle diameter from 19 to 250 μm .

When the particle size increases, the total surface area of the particles also increases. This was expected to increase the total soil surface area in contact with water drop, making the conditions of the monolayer more similar to the conditions on a flat solid surface. This was considered to be the reason for the decrease in contact angle with the increase in particle size.

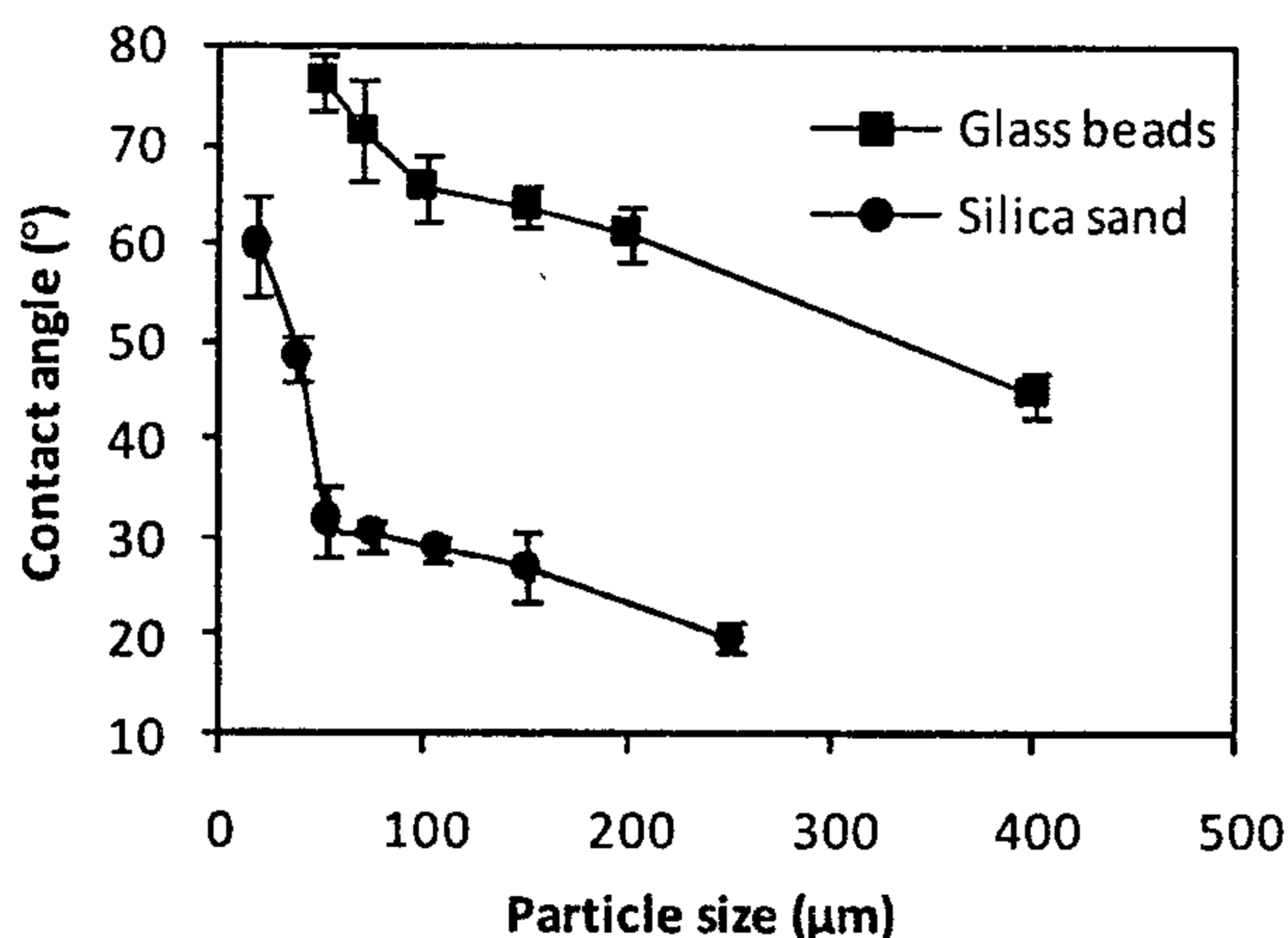


Figure 2: Effect of particle size on sessile drop contact angle for glass beads and silica sand. Error bars indicate \pm standard deviation.

Effect of drop size on sessile drop contact angle

The effect of drop size on sessile drop contact angle of Acryl, Vinyl tape, Adhesive tape, hydrophobized silica sand with 0.1% stearic acid content, and hydrophobized silica sand with 0.5% stearic acid content are respectively shown in Figure 3 (A), (B), (C), (D), and (E). With increasing drop size from 5 to 50 μL , the calculated contact angle continuously decreased. In contrary, the measured contact angle first decreased and then increased.

With the increasing drop size from 5 to 50 μL , the calculated sessile drop contact angle for acryl decreased from 70° to 52°, for vinyl tape from 90° to 70°, for adhesive tape from 106° to 88°, for hydrophobized silica sand with 0.1% stearic acid content from 98° to 86°, and for hydrophobized silica sand with 0.5% stearic acid content from 112° to 101°.

The equation for contact angle calculation was developed assuming that the shape of the water drop would be always spherical. This assumption can be justified for smaller drop sizes. However, with increasing drop size, the spherical shape gradually changes to be flattened in shape, showing

lower drop height, (h in Fig. 1) than the expected value. The decrease in drop height would result in lower contact angle as explained in Eqs.1, 2, 3, and 4. Accordingly, the decreased calculated contact angle with increasing drop size was considered to be a result of the lowered drop height compared with the expected value.

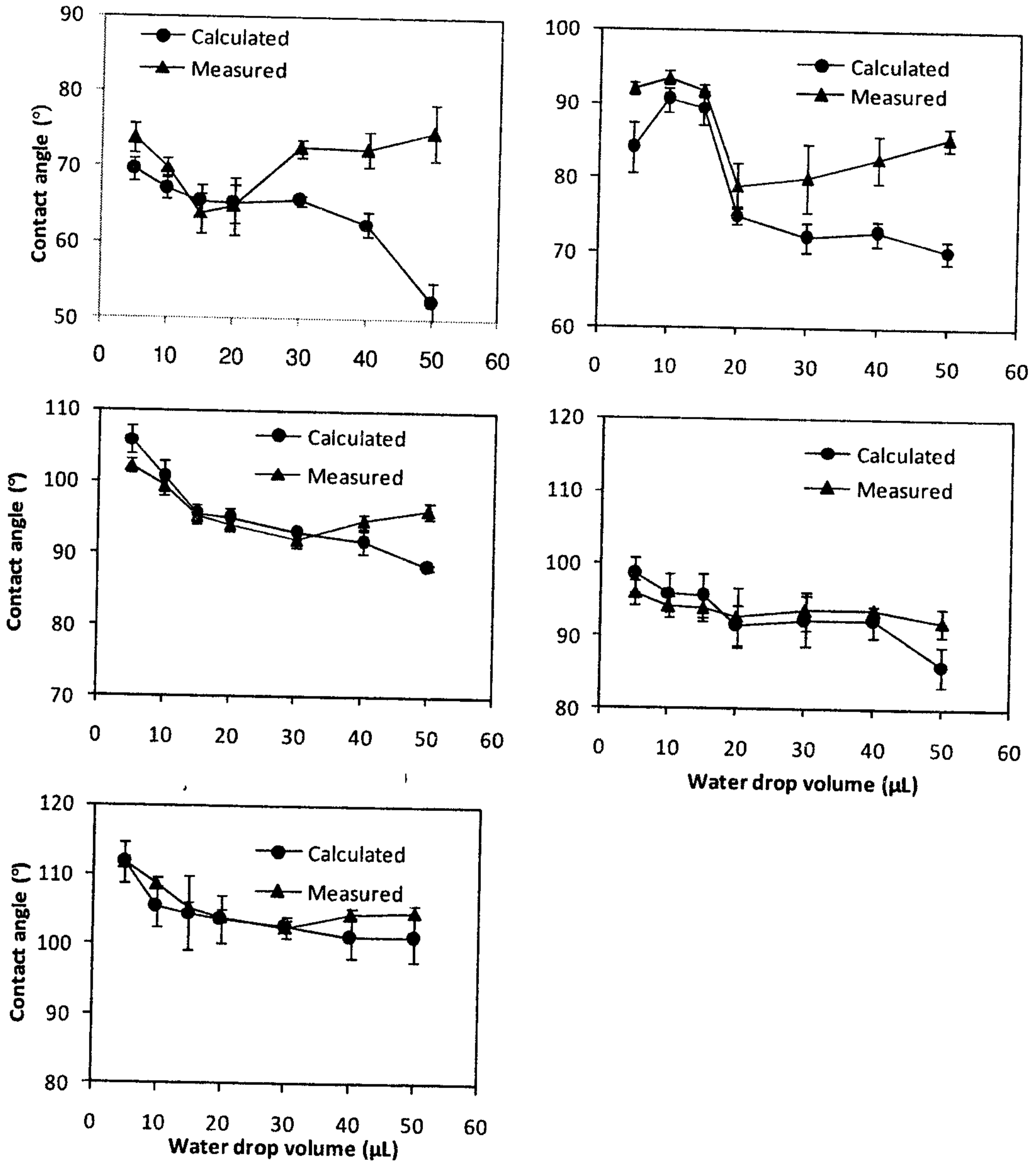


Figure 3: Effect of drop size on sessile drop contact angle of (A) acryl (B) Vinyl tape (C) Adhesive tape (D) Hydrophobized silica sand with 0.1% stearic acid (E) Hydrophobized silica sand with 0.5% stearic acid. Error bars indicate \pm standard deviation.

The measured sessile drop contact angle for acryl first decreased from 74° to 64° with increase in water drop size from 5 to 15 μL and then increased up to 74° with increasing drop size from 15 to 50 μL . For vinyl tape, the measured contact angle first decreased from 93° to 79° with increase in water drop size from 5 to 20 μL and then increased up to 85° with increasing drop size from 20 to 50 μL . For adhesive tape, the contact angle first decreased from 102° to 92° with increase in water drop size from 5 to 30 μL and then increased up to 96° with increasing drop size from 30 to 50 μL . For hydrophobized silica sand with 0.1% stearic acid content, the measured contact angle first decreased from 96° to 92° with increase in water drop size from 5 to 20 μL and then remained more or less uniform with increasing drop size from 20 to 50 μL . For hydrophobized silica sand with 0.5% stearic acid content, the measured contact angle first decreased from 112° to 103° with increase in water drop size from 5 to 30 μL and then increased up to 105° with increasing drop size from 30 to 50 μL .

Increase in drop size would decrease the contact angle due to the effect of linear tension as explained by Adamson (1990). This is applicable under the assumption that the effect of gravity on the drop shape would be negligible. However, it was clear that the drop shape was affected by the gravity for larger drop sizes. Therefore, the increase in measured contact angle after reaching a bottom value can be suggested as resulted by the effect of gravity.

Our results are in contrast with Good and Koo (1979) who reported that water–teflon contact angle decreased in about 8° with decreasing drop diameter from 4 to 1 mm (33 to 0.5 μL), whereas *n*-decane–teflon contact angle did not change. This shows that different results can be expected with different materials. However, the findings by Good and Noo (1979) are on advancing and receding contact angles and difficult to be related with the initial contact angle of sessile drops, which was considered in the present study.

Conclusion

With increase in particle size from 19 to 250 μm in silica sand and 50 to 400 μm in glass beads, the contact angle decreased by about 30° and 40°, respectively. With increase in drop size, the calculated contact angle gradually decreased while the measured contact angle initially decreased to reach a minimum at about 20 – 30 μl drop size and then increased possibly due to the gravity effect on the sessile drop.

The study revealed that smaller water drops, for example those used in mist or fog irrigation systems, will make higher contact angles and reduce the water entry to the surface soils. Therefore, pre-assessment of the initial soil-water contact angle would be essential before choosing irrigation systems for fields.

References

- Adamson, A.W. (1990). *Physical Chemistry of Surfaces*, 5th ed., John Wiley and Sons, New York, pp. 291–328.
- Annaka, T. (2006). Wettability indices and water characteristics for sands of mixed wettability. *Journal of the Japanese Society of Soil Physics*, 102, 79-84 (in Japanese with English summary).
- Bachmann, J., Ellies, A. and Hartge, K.H. (2000a) Development and application of a new sessile drop contact angle method to assess soil water repellency. *Journal of Hydrology*, 231–232, 66–75.
- Bachmann, J., Horton, R., Van de Ploeg, R.R. and Woche, S. (2000b). Modified sessile drop method for assessing initial soil-water contact angle of sandy soil. *Soil Science Society of America Journal*, 64, 564-567.

- Buczko U., Bens O. and Durner W. (2006). Spatial and temporal variability of water repellency in a sandy soil contaminated with tar oil and heavy metals. *Journal of Contaminant Hydrology*, 88(3-4):249–268.
- Buczko U., Bens O., Fischer H. and Hüttl R. F. (2002). Water repellency in sandy luvisols under different forest transformation stages in northeast Germany. *Geoderma*, 109, 1–18.
- Doerr, S.H., Shakesby, R.A. and Walsh, R.P.D. (2000). Soil water repellency: Its causes, characteristics and hydro-geomorphological significance. *Earth Science Review*, 51:33–65.
- Doerr, S.H., Llewellyn, C.T., Douglas, P., Morley, C.P., Mainwaring, K.A., Haskins, C., Johnsey, L., Ritsema, C.J., Stagnitti, F., Allinson, G., Ferreira, A.J.D., Keizer, J.J., Ziogas, A.K. and Diamantif, J. (2005). Extraction of compounds associated with water repellency in sandy soils of different origin. *Australian Journal of Soil Research*, 43, 225–237.
- Feng, G.L., Letey, J. and Wu, L. (2001). Water ponding depths affect temporal infiltration rates in a water repellent sand. *Soil Science Society of America Journal*, 65, 315–320.
- Goebel, M-O., Bachmann, J., Woche, S.K., Fischer, W.R. and Horton, R. (2004). Water potential and aggregate size effects on contact angle and surface energy. *Soil Science Society of America Journal*, 68, 383–393.
- Good, R.J. and Koo, M.N. (1979). The effect of drop size on contact angle. *Journal of Colloid and Interface Science*, 71(2), 283–292.
- Heslot, F., Cazabat, A.M., Levinson, P. and Fraysse, N. (1990). Experiments on wetting on the scale of nanometers: Influence of the surface energy. *Physical Review Letters*, 65, 599-602.
- Lachacz A., Nitkiewicz M. and Kalisz B. (2009). Water repellency of post-boggy soils with a various content of organic matter. *Biologia* 64(3), 634-638.
- Leelamanie, D.A.L. and Karube, J. (2007). Effects of organic compounds, water content and clay on water repellency of a model sandy soil. *Soil Science and Plant Nutrition*, 53, 711-719.
- Leelamanie, D.A.L., Karube, J. and Yoshida, A. (2008a). Characterizing water repellency indices: Contact angle and water drop penetration time of hydrophobized sand. *Soil Science and Plant Nutrition*, 54, 179-187.
- Leelamanie, D.A.L., Karube, J. and Yoshida, A. (2008b). Relative humidity effects on contact angle and water drop penetration time of hydrophobized fine sand. *Soil Science and Plant Nutrition*, 54, 695–700.
- Leelamanie, D.A.L., Karube, J. and Yoshida, A. (2010). Clay effects on contact angle and water drop penetration time of model soils. *Soil Science and Plant Nutrition*, 56,371–375.
- Roy, J.L. and McGill, W.B. (2002). Assessing soil water repellency using the molarity of ethanol droplet (MED) test. *Soil Science*, 167, 83–97.
- Wallis, M.G. and Horn, D.J. (1992). Soil water repellency. *Advances in Soil Science*, 20, 91-103.
- Wallis, M.G., Scotter, D.R. and Horne, D.J. (1991). An evaluation of the intrinsic sorptivity water repellency index on a range of New Zealand soils. *Australian Journal of Soil Research*, 29, 353–362.