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### **Paper:**

Hallin, I., Douglas, P., Doerr, S. & Bryant, R. (2015). The effect of addition of a wettable biochar on soil water repellency. *European Journal of Soil Science*, 66(6), 1063-1073.

<http://dx.doi.org/10.1111/ejss.12300>

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1 **The effect of addition of a wettable biochar on soil water repellency**

2

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14 *Running title: Effect of biochar on WDPT*

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16 *Keywords: soil amendment, hydrophobicity, WDPT*

17

18 **Summary**

19 The potential of biochar to ameliorate soil water repellency has not been widely studied.  
20 Previous studies have focused on the potential for biochar to induce or exacerbate existing  
21 water repellency rather than alleviate it. This study investigates the effect of adding wettable  
22 biochar to water repellent soil by comparing the water drop penetration times (WDPT) of  
23 control and biochar-amended soil.

24 The potential of wettable biochar to act as a physical amendment to water repellent soil was  
25 evaluated by mixing coarsely ground biochar (CGB, particle size range 250–2000  $\mu\text{m}$ ) or  
26 finely ground biochar (FGB, particle size range  $< 250 \mu\text{m}$ ) with one strongly and one  
27 severely naturally water repellent soil in various quantities, and measuring the WDPT for  
28 each mixture. When biochar particles did not fall within the range of existing soil particle  
29 diameters, an initial increase in both mean WDPT ( $\text{WDPT}_M$ ) and variation in WDPT was  
30 observed with small additions of biochar. These effects possibly resulted from increased  
31 surface roughness and inhibition of infiltration by suspension of drops above the average soil-  
32 air interface at a few hydrophobic points. Both CGB and FGB reduced soil water repellency,  
33 FGB more effectively than CGB. Adding 10% w/w FGB reduced soil WDPT by 50%, and  
34 25% FGB eliminated repellency. Direct absorption of water by biochar and an increase in soil  
35 surface area in contact with water are likely the predominant physical mechanisms involved.  
36 This exploratory study suggests biochar has the potential to amend water repellent soil.

37

38

## 39 **Introduction**

40 The use of biochar as a soil amendment has become an important area of research. Until  
41 recently, interests have been mainly in the effects of biochar application on carbon  
42 sequestration and nutrient and water retention to improve agricultural soil (Sohi *et al.*, 2010).  
43 More recently, biochar production and application have been evaluated for the remediation of  
44 contaminated soil. A study by Debela *et al.* (2011) found that when biochar feedstock and  
45 contaminated soil were pyrolysed together, heavy metals could be encapsulated by the  
46 biochar during pyrolysis and rendered unavailable. Similarly, Beesley & Marmiroli (2011)  
47 found Cd and Zn levels decreased significantly when leachate from a contaminated soil was  
48 filtered through biochar columns.

49 Soil water repellency (SWR) can have natural or anthropogenic causes. Plant decomposition  
50 can release long-chain fatty acids, alkanes and lipids, which coat soil particles, decrease the  
51 surface tension and restrict water infiltration at the soil surface (Morley *et al.*, 2005; Koch &  
52 Ensikat, 2008). Water repellency can also develop after an oil spill, as hydrocarbons enter  
53 pore spaces and decrease the soil surface tension (Roy & McGill, 2003). SWR occurs in  
54 many parts of the world regardless of climate or soil texture. In Australia, for example, SWR  
55 frequently develops under eucalypt stands and following wildfires. Wildfires mainly enhance  
56 existing water repellency rather than introduce it, and the degree of repellency observed  
57 depends on the severity of the fire, the vegetation community present and soil type (Doerr *et*  
58 *al.*, 2009; DeBano, 2000).

59 The reduced wettability of water repellent soil areas not only enhances the risk of overland  
60 flow and associated erosion, but it can also reduce vegetation recovery and prolong the time  
61 burned areas are susceptible to erosion (DeBano, 2000). Mitigation measures to encourage  
62 faster revegetation and decrease erodibility vary depending on the cause and the

63 characteristics of the affected site. In some areas, surfactants are used to increase soil surface  
64 tension and allow water infiltration (Barton & Colmer, 2011), whereas in others mechanical  
65 disturbance or tillage are used to disperse hydrophobic soil among more wettable soil,  
66 exposing soil to air for organic compounds to mineralize (Blanco-Canqui, 2011; Harper *et al.*,  
67 2000). Direct addition of a wettable material to the soil has also been successful, such as the  
68 addition of clay to Australian agricultural. Harper & Gilkes (1994) found SWR was reduced  
69 with a 1% increase in clay content, and eliminated with a 5% increase. Kaolin clays have  
70 been found to be most effective in rendering soils wettable (McKissock *et al.*, 1999; Lichner  
71 *et al.*, 2006).

72 Biochar, like clay, can have very large surface areas and is strongly adsorbent. Therefore,  
73 biochar could potentially amend water repellent soil in a similar way to clay. A report by the  
74 European Commission (Verheijen *et al.*, 2009) identified a research gap on the effect of  
75 biochar on water repellent soil, and several studies since have included the effects of water  
76 repellency in their research on biochar. Abel *et al.* (2013) investigated the effect of biochar  
77 (produced from maize feedstock and pyrolysed at 750°C) on soil water retention and  
78 repellency through laboratory and field trials of soil columns mixed with 0, 1, 2.5 or 5% w/w  
79 biochar. The five soils they used were all sands or loamy sands taken from the top layer of a  
80 Regosol, a Luvisol or a former sewage farm site. Their organic matter contents ranged from  
81 0.1 to 9.1%, and they were wettable before biochar application. At the end of the six-month  
82 trial, Abel *et al.* (2013) concluded that biochar had no effect on soil water repellency, as  
83 water drop penetration time (WDPT) tests revealed no increase in the time to infiltration.

84 Herath *et al.* (2013) tested whether biochar could improve soil water-holding capacity and  
85 drainage capabilities by studying the effect of two biochars on the bulk density, aggregate  
86 stability, saturated hydraulic conductivity, volumetric water content and water repellency of

87 two silt loams from permanent pastures. One soil was slightly repellent (WDPT 5–60  
88 seconds) with 41.7 g C kg<sup>-1</sup> soil and the other was strongly repellent (WDPT 1–60 minutes)  
89 with 102 g C kg<sup>-1</sup> soil. The biochars were produced from corn stover feedstock and were  
90 pyrolysed at 350°C and 550°C. They were applied to each soil at a rate of 7.18 t C ha<sup>-1</sup> and  
91 packed into columns where they were maintained at field capacity for 295 days. In terms of  
92 water repellency alone, at the end of the ~300-day study, the strongly repellent soil had  
93 become slightly repellent and the slightly repellent soil remained the same. This led to the  
94 conclusion that biochar had no significant effect on water repellency.

95 The properties of biochar depend largely on their feedstock (i.e. type of biomass used) and  
96 pyrolysis conditions. Biochar produced at temperatures < 500°C retains many organic  
97 functional groups from the original feedstock, and is therefore usually water repellent  
98 (Kinney *et al.*, 2012; Antal & Grønli, 2003). Pyrolysis temperatures > 500°C will volatilize  
99 the organic groups linked to hydrophobicity, rendering the biochar hydrophilic. Kinney *et al.*  
100 (2012) found that biochars from three different feedstocks followed the same trend: pyrolysis  
101 at 300°C produced extremely hydrophobic biochar, but hydrophobicity decreased with  
102 increasing temperature and at temperatures > 500°C the biochar produced was wettable.

103 To evaluate the potential of biochar as a physical amendment for water repellent soil, three  
104 questions were posed.

- 105 1) Does the addition of a wettable biochar have a significant effect on the water drop  
106 penetration time (WDPT) when mixed into a water repellent soil?
- 107 2) How does the addition of biochar affect soil WDPT compared to the addition of wettable  
108 soil?
- 109 3) If wettable biochar does reduce soil water repellency, is there an optimal application rate?

**Accepted manuscript version of:** Hallin, I., Douglas, P., Doerr, S.H. & Bryant, R. (2015) The effect of addition of a wettable biochar on soil water repellency. *European Journal of Soil Science*, 66, 1063-1073. (doi:10.1111/ejss.12300)

110 A laboratory experiment was designed to explore the effects of various quantities of biochar  
111 on two water repellent soils. Wettable coarsely or finely ground biochars were mixed with  
112 strongly repellent coarse sand (CS) and extremely repellent medium sand (MS) in w/w  
113 proportions of 5, 10, 25 and 40%. These ratios were chosen to gain a fundamental  
114 understanding of the effect biochar might have rather than to mimic the rates of field  
115 applications. Mixtures of wettable and water repellent soil were created as controls, and the  
116 water repellency of each mixture was tested with the WDPT test. The effect of biochar  
117 addition on mean soil WDPT ( $WDPT_M$ ) was then compared to that of wettable soil.

118

## 119 **Materials and methods**

### 120 *Naturally water repellent soil*

121 Two naturally water repellent soils were studied: CS, a coarse, relatively homogenous sand  
122 with low organic carbon content (Arenosol; IUSS Working Group WRB, 2006), taken  
123 beneath grass cover from dunes in Nicholaston, Gower, Wales; and MS, a medium sand with  
124 a similar mean texture to CS but more organic carbon (Anthrosol; IUSS Working Group  
125 WRB, 2006), taken from a Leyland Cypress stand (*Cuprocyparis leylandii* (A.B. Jacks. &  
126 Dallim.) on the Singleton Campus in Swansea, Wales (Table 1). Previous studies classified  
127 both soils as highly repellent (CS is the ‘Nicholaston sand’ characterised in Doerr *et al.*  
128 (2005); and MS is the ‘University sand’ characterised by Hallin *et al.* (2013)).

129 Both soil samples were taken from the upper 5 cm of the soil profile, and any litter layer was  
130 removed beforehand. The soil was air-dried at approximately 20°C and then sieved to 2 mm  
131 to remove large fragments of organic matter.

### 132 *Preparation of wettable analogues*

133 Mixtures of wettable and water repellent soil were created as controls to determine whether  
134 any effects observed in biochar:soil mixtures were due to the addition of a wettable fraction  
135 to a water repellent system, or to biochar-specific properties. Rather than introduce new soils,  
136 wettable analogues were prepared by stripping organic matter from CS and MS. A sample of  
137 soil was washed with 0.1 M NaOH for 30 minutes to remove organic matter and then rinsed  
138 with distilled water until the solution reached a neutral pH. The washed soil was oven-dried  
139 at 105°C for 24 to 48 hours, then allowed to cool to 20°C. This process was repeated until  
140 water drops applied to a small subsample infiltrated consistently within 5 s. The wettable soil  
141 analogues obtained from CS and MS are identified as CS<sub>w</sub> and MS<sub>w</sub>, respectively. Their mean  
142 particle diameter, range of particle diameter and total organic carbon content are given in  
143 Table 1.

#### 144 *Biochar*

145 Biochar was provided by the UK Biochar Research Centre in Edinburgh. This was prepared  
146 from a mixed softwood feedstock of pine and spruce pellets (Puffin Pellets, Banff, Scotland),  
147 pyrolysed in a 250-mm diameter rotary kiln at a peak temperature of 700°C with intermediate  
148 mean residence time. Coarsely ground biochar (CGB, particle-size range 250 to 2000 µm)  
149 and finely ground biochar (FGB, particle-size range < 250 µm) were prepared by grinding  
150 these pellets with a pestle and mortar followed by sieving through a 250 µm sieve.

#### 151 *Particle-size distributions for soil and coarse and fine biochars*

152 Soil particle size was measured by laser diffraction with a Malvern (Malvern, UK)  
153 Mastersizer 2000. Three subsamples of each water repellent soil and its analogue were  
154 analysed for particle size (µm) by volume (%). Particle-size distributions (PSD) for CS and  
155 MS are given in Figure 1a.



156 A Beckman Coulter (Brea, USA) LS 230 Dry Powder Module (Model Number DPM) was  
157 used to measure the PSD for CGB and FGB (Figure 1b), and summary statistics are given in  
158 Table 2.

159 *Particle-size distribution for biochar:soil mixtures*

160 Weighted volume fractions were used to calculate the PSD for each component according to  
161 Equation (1).

$$162 \quad V_{T_i} = \frac{(V_{B_i} \times V_{B_T}) + (V_{S_i} \times V_{S_T})}{(V_{B_T} + V_{S_T})} \times 100, \quad (1)$$

163 where  $V_{T_i}$  is the total volume (%) of particles in the mixture at any given diameter 'i',  $V_{B_i}$  and  
164  $V_{S_i}$  are the volume fractions of biochar and soil at diameter 'i', and  $V_{B_T}$  and  $V_{S_T}$  are the total  
165 volumes of biochar and soil in the mixture, respectively.

166 Total volumes of biochar and soil for each 200 g mixture were calculated with Equations (2)  
167 and (3), respectively, where  $M$  is the mass fraction of biochar or soil in the mixture (e.g. 0.05  
168 biochar and 0.95 soil), 200 refers to the mass of the mixture and  $\rho$  is the mean density of each  
169 material ( $\text{g cm}^{-3}$ ).

$$170 \quad V_{B_T} = (M_B \times 200) / \rho_B \quad (2)$$

$$171 \quad V_{S_T} = (M_S \times 200) / \rho_S \quad (3)$$

172 Densities were measured by packing each solid loosely into a  $10 \text{ cm}^3$  volumetric flask,  
173 tapping to settle until the meniscus mark was reached. Flasks were then weighed, and the  
174 entire process was repeated until the difference between two replicates was  $< 5\%$  of the  
175 sample mass. This gave:  $\rho_{CS} = 1.56 \pm 0.01$ ;  $\rho_{MS} = 0.930 \pm 0.080$ ;  $\rho_{CGB} = 0.388 \pm 0.004$ ; and  
176  $\rho_{FGB} = 0.645 \pm 0.003$ .

177 *Preparation of biochar:soil mixtures and WDPT tests*

178 Soil samples were coned and quartered (Jackson, 1958) to ensure homogeneity when  
179 obtaining the desired weight for each mixture. Soil and biochar fractions were weighed to  $\pm$   
180 0.0001 g, and all materials were then kept in a constant temperature and humidity room for  
181 24 hours prior to and following mixing at  $18 \pm 0.5^\circ\text{C}$  and  $29 \pm 3\%$  relative humidity (RH). To  
182 avoid uneven settling, soil and biochar were kept separate until they were mixed in a  $22.5 \times$   
183 31.5-cm tray. First the soil was spread evenly across the tray, then biochar was spread evenly  
184 across the soil surface. The two were incorporated manually until no distinct patches of  
185 biochar or soil were evident and the surface of the mixture appeared homogenous.

186 Previous research by Hallin *et al.* (2013) investigated the effect of drop volume in WDPT  
187 classification, and found that in general, larger drops (80, 200  $\mu\text{l}$ ) provide more of an average  
188 water repellency class for the soil, whereas smaller drops (15, 20  $\mu\text{l}$ ) better reflect surface  
189 heterogeneities. Both 20 and 200  $\mu\text{l}$  drops were used in this study to obtain information on  
190 both surface heterogeneities and overall average water repellency of the biochar:soil  
191 mixtures. Drops of distilled water (20 and 200  $\mu\text{l}$ ), equilibrated to  $18 \pm 0.5^\circ\text{C}$ , were applied  
192 from  $\leq 1$  cm height to each tray of soil following a grid pattern with a spacing of  $3 \times 2$  cm . In  
193 total, 104 drops of each volume were applied to each tray with an Eppendorf (Hamburg,  
194 Germany) Repeater Plus pipette. The time from placement to complete infiltration was  
195 recorded for each drop as the WDPT.

196 *Descriptive statistics and statistical analyses for WDPT measurements*

197 Descriptive statistics (mean, standard deviation and variance) were calculated with the  
198 equations for normal distributions. Error estimates are quoted as  $\pm 1$  standard deviation from  
199 the mean, except when only one sample was available, in which case only the sample value is  
200 reported.

201 Any irregular drops, such as those that were dispensed unevenly or those that rolled across  
202 the surface, were removed from the data sets before statistical analysis of WDPT results. The  
203 results from a one-way analysis of variance (ANOVA) indicated that there were significant  
204 differences between group means for all comparisons ( $P < 0.05$ ). Therefore, two-tailed  $t$ -tests  
205 ( $\alpha = 0.05$ ) were applied to explore the data further. Comparisons were made between results  
206 from the same mixture and between results from different mixtures made with the same grade  
207 of biochar.

208 Percentage change in water repellency ( $\Delta$ WDPT) was calculated for each mixture according  
209 to Equation (4), where  $T_S$  is  $WDPT_M$  for soil and  $T_M$  is  $WDPT_M$  for the biochar:soil mixture.

$$210 \quad \left( \frac{T_S - T_M}{T_S} \right) \times 100\% = \Delta WDPT \quad (4)$$

211

#### 212 *Microscope photographs of hydrophobized AWS:biochar mixtures*

213 Acid-washed sand (AWS), hydrophobized with octadecanoic acid according to the method  
214 described in Hallin (2013) and Mainwaring *et al.* (2013), was mixed with FGB to create a  
215 model system for the biochar:soil mixtures that would photograph well under the microscope.

216 Photographs were taken with Fuji (Tokyo, Japan) Reala colour film (ASA 100) with an  
217 Olympus (Shinjuku, Japan) BH2 microscope fitted with a DPlan 4PO 4 $\times$  objective lens and  
218 an NFK 3.3 $\times$  LD camera lens with an Olympus OM 4 camera. Samples were illuminated  
219 from the side with a Schott (Mainz, Germany) KL1500 fibre optic light through a Wratten  
220 (New York, USA) 80B filter.

221 *Preliminary studies*

222 For biochar to be considered a potential SWR amendment, the practicalities involved in its  
223 application must be considered in conjunction with its efficacy. Therefore, the wettability of  
224 different grades of biochar (pellets, CGB, FGB) and the suitability of different application  
225 techniques were evaluated before preparation of biochar:soil mixtures. Biochar wettability  
226 was evaluated by applying water drops directly to the surface of biochar pellets and dishes of  
227 CGB and FGB. All drops infiltrated immediately on contact (WDPT < 1 s).

228 Mixtures of w/w 10% biochar and 90% CS (10:90) were prepared with each grade of biochar  
229 to evaluate the application of biochar to the surface versus thorough mixing of biochar into  
230 the soil.

231 Pellets were immediately ruled out for further study because their size prevented them being  
232 mixed effectively with the soil. When applied to the surface, they created a distinct layer on  
233 top of the soil and water infiltrated the pellet before touching the soil. Both CGB and FGB  
234 could be mixed into the soil or applied to the surface, and both grades reduced WDPT when  
235 tested with nine 20 and 200  $\mu$ l drops. Surface-applied biochar resulted in uneven wetting and  
236 surface puddles, therefore the final biochar:soil mixtures were created by thoroughly mixing  
237 CGB or FGB throughout the soil.

238 Previous work by Hallin *et al.* (2013) tested mixtures of 90, 75 and 50% water repellent soil  
239 with 10, 25 and 50% w/w wettable analogue soil. The same ratios were used for biochar:soil  
240 mixtures to enable direct comparisons to wettable soil. The 50:50 biochar:soil mixtures were  
241 unsuccessful, however, as the biochar settled into a separate layer from the soil even after  
242 mixing. A trial 40:60 biochar:soil mixture did not separate, and replaced the 50:50 mixtures.  
243 An additional mixture, 5:95 biochar:soil, was also included to evaluate the effects of smaller  
244 quantities of biochar on SWR.

245 **Results**

246 *Particle-size distributions of biochar:soil and soil:soil mixtures*

247 Addition of either CGB or FGB to CS introduces a new, wide range of particle sizes to the  
248 soil, but for MS the effect of mixing on PSD is negligible (Figure 1).

249 The mean particle diameters of CS<sub>w</sub> and MS<sub>w</sub> are not much different from those of CS and  
250 MS, but the loss of organic matter and very small clay particles through the rinsing process  
251 results in a narrower PSD, especially for MS<sub>w</sub> (Figure 2). For CS, which had little organic  
252 matter or small clay particles, the effect is negligible, but for MS this narrows the particle-  
253 size distribution in the mixture.

254 *Effect of drop volume on WDPT*

255 The WDPT results for biochar:soil mixtures are given in Figures 3 and 4. General trends for  
256 200 µl indicate that WDPT<sub>M</sub> decreases with increasing biochar addition (Figure 3). This trend  
257 is also observed in the results from 20 µl drops, but not to the same extent (Figure 4). In  
258 accordance with Hallin *et al.* (2013), 20 µl drops show more variation in WDPT than 200 µl  
259 drops, likely because the smaller footprint of a 20 µl drop encounters greater surface  
260 heterogeneity than the larger footprint of the 200 µl drop. The smaller drops reflect the  
261 greater degree of heterogeneity within the 5 and 10% biochar mixtures, especially those with  
262 CGB, and as a result WDPT<sub>M</sub> shows no trend. The variation in the results is large for most  
263 mixtures with ≥ 25% biochar; the trend with 200 µl drops becomes evident at this point, as  
264 WDPT<sub>M</sub> is lowest for all mixes with 40% biochar.

265 The results of ANOVA are given in Table 3. Two-tailed *t*-tests indicate that the difference  
266 between 5 and 10% FGB mixtures is consistently significant, with 10% FGB resulting in  
267 smaller WDPT<sub>M</sub> values than 5% FGB. In contrast, the difference between 5 and 10% CGB is

268 generally not statistically significant, with the exception of CGB:CS measured with 200  $\mu$ l  
269 drops. There is no statistically significant difference between 25:75 and 40:60 mixes for  
270 either of the biochar:soil combinations or drop volumes; both mixtures have the smallest  
271  $WDPT_M$  values.

#### 272 *Effectiveness of biochar versus wettable soil analogue in reducing SWR*

273 Figure 5 shows the percentage reduction in  $WDPT_M$  with the addition of biochar or wettable  
274 soil analogue for all mixtures.

275 FGB is the more effective of the two biochars; it reduces soil water repellency by  $\geq 60\%$  in  
276 10:90 mixtures compared to an approximate 30% reduction with CGB. FGB is also equally  
277 effective in both soils:  $\geq 25\%$  FGB added to either soil eliminates SWR, whereas  $\geq 25\%$   
278 CGB is more effective in CS.

279 On a % w/w basis, FGB is also more effective at reducing water repellency than the wettable  
280 soil analogues when tested with 200  $\mu$ l drops (Figure 5a). The FGB:CS mixtures are at least  
281 25% less repellent than the corresponding  $CS_w$ :CS mixtures, and 25% FGB removes  
282 repellency in both soils, whereas the mixtures with 50% wettable analogue still show signs of  
283 water repellency. Once again CGB is not consistently effective, and results vary between the  
284 two soils. Reductions in water repellency are consistently greater for  $CS_w$  mixes than for  
285 CGB mixes, sometimes by 30% or more. However, CGB is effective in the medium sand:  
286 40% CGB reduces water repellency by approximately 96% compared to the 90% reduction  
287 achieved with 50%  $MS_w$ .

288 When tested with 20  $\mu$ l drops (Figure 5b), the wettable soil analogues appear to be more  
289 effective than additions of either CGB or FGB; 10%  $CS_w$  reduces water repellency in CS by  
290 approximately 90%, compared to a 75% reduction by 10% FGB. Furthermore, 200  $\mu$ l drops

291 show no repellency with > 25% biochar present, whereas 20  $\mu$ l drops show some repellency  
292 even with 40% CGB, but none with  $\geq$  25% wettable soil analogue.

## 293 **Discussion**

294 The following discussion focuses on results from 200  $\mu$ l drops, which best reflect the average  
295 degree of water repellency.

### 296 *Effect of particle-size distribution on WDPT*

297 Increased variation in  $WDPT_M$  is observed in results from both CGB and FGB for 5:95  
298 biochar:CS mixtures (Figure 3), but not for biochar:MS mixtures, suggesting a soil-specific  
299 change was introduced by biochar. This variation might arise from increased surface  
300 roughness: the addition of CGB and FGB to CS introduced a new range of fine particle sizes  
301 to the coarse sand, which changed the topography of the surface by increasing surface  
302 roughness. The changes to surface topography and roughness in CS through biochar addition  
303 might inhibit infiltration by the suspension of drops above the position of the average soil-air  
304 interface at a few hydrophobic points. Soil particles of comparable size to those of biochar in  
305 CGB and FGB are already present in MS, so the addition of biochar to MS does not affect  
306 surface topography in the same way as for CS (Figure 1).

307 Additional biochar would increase surface roughness further, but would also be likely to  
308 increase the probability of contact between the water drop and biochar. Photomicrographs of  
309 hydrophobized AWS mixed with biochar show that biochar adheres to AWS particles (Figure  
310 6), suggesting that it could cover the hydrophobic compounds adsorbed to a soil particle and  
311 provide a wettable surface. The controlling factor for water infiltration then becomes  
312 wettability of the biochar rather than surface roughness, similar to the mechanism proposed  
313 for clay amendment (Müller & Deurer, 2011).

314 The chemical effect of biochar on soil may also be important (Hallin, 2013), but biochar in  
315 contact with a water drop can physically decrease  $WDPT_M$  in several ways. The high porosity  
316 and wettability of CGB or FGB allows it to act like a sponge and soak up water rapidly. This  
317 is unlikely to make a noticeable difference at 5% levels because there is little biochar at the  
318 surface. Figure 7 shows theoretical contact areas between water drops and CGB:soil  
319 mixtures, and illustrates that drops are unlikely to come into contact with much biochar for  
320 5:95 mixtures. If water drop-biochar contact were made, the wettable biochar would absorb  
321 water from the drop like a sponge.

322 The wettability and porosity of biochar pellets was estimated by assessing their average water  
323 uptake. Based on the mass difference between dry and saturated biochar and the volume of  
324 water absorbed, pellet porosity was estimated to be approximately 55%. Even assuming that  
325 porosity remained at ~ 55% after grinding, the largest FGB particle would still be able to  
326 absorb only about 0.05% of a 200  $\mu$ l drop. Similarly, the largest CGB particle could absorb ~  
327 1.5% of a 200  $\mu$ l drop. The ‘sponge effect’ is, therefore, more likely to make a noticeable  
328 difference when enough biochar is present so that its particles are in contact with each other  
329 and can form preferential flow paths through the soil (Figure 7, 40:60 mixtures).

330 A rough, but useful, estimate of potential soil particle cover by biochar can be calculated  
331 from the ratio of mean particle diameters and the proportions of each component within the  
332 mixture if we make the following assumptions: (i) a biochar particle will bind to a soil  
333 particle only along one side, and (ii) soil particles are roughly spherical and biochar particles  
334 are roughly cuboid. Results of these calculations for soil particle cover by biochar are given  
335 in Table 4; even for the largest mixture ratio CGB could cover only a very small fraction of  
336 the soil particle area, whereas FGB could potentially cover a considerable fraction of both CS  
337 and MS particles.



338 Biochar might also reduce  $WDPT_M$  by increasing the effective surface area of soil in contact  
339 with water. A water drop in contact with a water repellent soil surface will be attracted to the  
340 polar functional groups present in the hydrophobic compounds coating the soil (Bachmann &  
341 van der Ploeg, 2002; Roy & McGill, 2002). As water adsorbs to the more wettable parts of  
342 the soil particle surfaces, the surface energy of the soil-water interface decreases and the  
343 initial soil-water contact angle decreases, eventually decreasing sufficiently for the drop to  
344 overcome repellency and spread across the soil surface. According to Leelamanie & Karube  
345 (2009), this process is repeated on contact with each subsequent layer of soil until the drop  
346 has infiltrated completely; the total time required is the WDPT. When biochar absorbs water  
347 from the drop or the surrounding soil, it increases the total area of soil exposed to water.  
348 Absorbed water would come into contact with subsurface soil through biochar surfaces, and  
349 contribute to the overall wetting process and reduction in  $WDPT_M$ .

#### 350 *Potential for biochar as a soil amendment*

351 Before any recommendations can be made on the potential of biochar as a SWR amendment,  
352 laboratory and field trials with soil of different textures and different degrees of water  
353 repellency are required. A range of water contents relevant to specific applications should be  
354 considered. For example, irrigated soil often shows water repellency most strongly when  
355 almost air dry, so further testing of biochar:soil mixtures at water contents equivalent to air-  
356 dry conditions should be done before any application to irrigated soil. Additionally, its  
357 longer-term efficacy needs to be evaluated and biochar needs to be compared to other  
358 amendments to determine its relative effectiveness both physically and economically.

359 While bearing these caveats in mind, these analyses indicate that both FGB and CGB could  
360 potentially be suitable amendments for water repellent soil. Biochar was generally more  
361 effective at reducing water repellency than the wettable soil analogues, which suggests that

362 biochar has an effect in soil beyond that of simply adding a wettable component to a water  
363 repellent system. The effectiveness of biochar, however, varied considerably with soil texture  
364 and biochar particle size. FGB additions  $\geq 25\%$  eliminated water repellency in both medium  
365 and sandy soil, and FGB often reduced water repellency more than CGB, which suggests that  
366 a finely ground biochar might be more effective than coarse biochar.

367 The most comparable soil amendment to FGB currently in use is probably clay (Cann 2000;  
368 Dlapa *et al.*, 2004). Both wettable biochar and clay have similar physical effects on soil, i.e.  
369 they increase the hydrophilic surface area for water infiltration within the soil (Cann, 2000;  
370 Lichner *et al.* 2006), but biochar has the added benefit of being a carbon source and of storing  
371 nutrients for slow release over time, as well as being a possible route for carbon capture of  
372 atmospheric CO<sub>2</sub>. Biochar is not inert, however, and it will eventually decompose, which  
373 might affect its ability to act as a long-term remediation strategy for water repellent soil.  
374 Residence times of biochar in soil will vary, but Woolf & Lehmann (2012) estimate a mean  
375 half-life of 1000 years for the recalcitrant portion. Clay amendments are similarly time-  
376 dependent; clay will not degrade like biochar, but natural soil development processes over  
377 similar time scales (~1000 years) can translocate clay deeper in the soil profile. Over shorter  
378 time scales clay has been shown to be effective: Van Dam *et al.* (1990) found that clay was  
379 still present in clay-amended topsoil 30 years after application, and Ward & Oades (1993)  
380 reported anecdotal evidence that claims clay was effective in reducing SWR for at least 20  
381 years after application. The lifetime of biochar within the soil and its long-term effectiveness  
382 within water repellent soil as they weather and change is another area that must be explored.

383 Rates of biochar application must also be considered. For example, golf courses in the USA,  
384 which are frequently affected by SWR (Kostka, 2000), stipulate a clay content of no more  
385 than 3% for the root zone soil (USGA, 1973). Similar limits would probably be applied here

386 to biochar to preserve the textural integrity and playability of greens, making large  
387 application rates, such as 25% FGB, unrealistic.

388 Like any amendment, the viability of applying biochar to reduce water repellency depends on  
389 its direct cost, the availability of cheaper alternatives, transport costs and on return for the  
390 investment. These factors vary considerably between potential applications, and an in-depth  
391 analysis is beyond the scope of this study. Biochar might be a feasible option for medium  
392 textured soil, such as the medium sand in this research, in areas where clay is not readily  
393 available but pyrolysis facilities can be built, or where less biochar is needed to reduce  
394 repellency, for example, localized problems with water repellency in high value areas such as  
395 in manicured landscapes and gardens. The environmental, social and potentially economic  
396 (through, for example, 'carbon credits') value of using biochar, might also become important  
397 considerations depending on the targeted application.

398 There are many unknowns about the effectiveness of biochar, including (but not limited to)  
399 the long-term effects of biochar addition on SWR and the effects of repeated wetting and  
400 drying cycles and of water repellency-inducing bacteria and fungi on biochar porosity and  
401 sorption capabilities. The results of this preliminary study, however, demonstrate that biochar  
402 has the potential to be used for this purpose.

### 403 **Conclusions**

404 Wettable biochar substantially increased the wettability of highly water repellent coarse- and  
405 medium-textured soil, more so than was achieved by adding wettable soil analogues. Of the  
406 three amendments tested (FGB, CGB and wettable soil analogue), FGB was most effective:  
407 10% FGB reduced  $WDPT_M$  more than larger quantities of CGB or wettable soil analogue.  
408 The addition of 10% FGB reduced water repellency by 50%, and 25% FGB eliminated water  
409 repellency in both the medium and coarse textured soils tested. The predominant physical

410 mechanisms likely to be involved are: direct absorption of water by biochar; a decreased, or  
411 more rapidly decreasing, soil-water interfacial energy; and an increased surface area of soil in  
412 contact with water. When biochar particles did not fall within the range of existing soil  
413 particle diameters, an initial increase in  $WDPT_M$  and variation in  $WDPT$  was observed,  
414 which might be explained by increased surface roughness and the resulting inhibition of  
415 infiltration by the suspension of drop above the position of the average soil-air interface at a  
416 few hydrophobic points.

417 The results presented and discussed here suggest that biochar has the potential to act as an  
418 amendment for water repellent soil, however, further laboratory and field trials are necessary.

419

420

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- 523

524 **Table 1.** Source locations and characteristics for naturally water repellent soil used in this study.

Sample Code	Coarse Sand (CS)	Medium Sand (MS)
Source location	Nicholaston, Wales, UK	Swansea University Campus, Wales, UK
Latitude and longitude	51°34'21"N; 4°08'2"W	51°36'34"N; 3°58'50"W
Textural class	Coarse sand	Medium sand
FAO classification	Arenosol	Anthrosol
Mean particle diameter/ mm	0.30 0.31 <sup>1</sup>	0.41 0.28 <sup>2</sup>
Particle diameter range/ mm	0.21 to 0.42 0.22 to 0.42 <sup>1</sup>	0.08 to 1.18 0.13 to 0.60 <sup>2</sup>
Total organic carbon/ %	0.57 ± 0.05 0.15 ± 0.01 <sup>1</sup>	17.25 ± 0.76 1.33 ± 0.22 <sup>2</sup>

525

526 <sup>1.</sup> Wettable coarse sand analogue (CS<sub>w</sub>)

527 <sup>2.</sup> Wettable medium sand analogue (MS<sub>w</sub>)

528



529 **Table 2.** Summary statistics for coarsely ground (CGB) and finely ground (FGB) biochar particle size  
530 analyses. Note that distribution width (standard deviation/mean) is unitless.

		Median/ mm	Mean/ mm	Standard deviation/ mm	Distribution width
CGB*	Arithmetic	0.75	0.79	0.63	0.80
	Geometric		0.69	0.02	0.03
FGB†	Arithmetic	0.17	0.19	0.28	1.43
	Geometric		0.13	0.01	0.07

531 Particle size obtained with a Beckman Coulter LS 230 Dry Powder Module with settings as follows.  
532 \*auger, 71; vibration, 16; mean obscuration, 4 to 7%; chute tilt, slightly off maximum; hopper, full.  
533 †auger, 21; vibration, 25 to 30; mean obscuration, 5 to 10%; chute tilt, full angle; hopper, 0.5 to 0.75  
534 full.  
535

536 **Table 3.** The results of ANOVA for coarsely ground (CGB) and finely ground (FGB) biochar mixtures  
 537 with medium (MS) and coarse (CS) sand. Each listing under ‘Group Compared’ refers to all mixtures  
 538 within that group (for example, CGB:MS 200  $\mu$ l refers to an ANOVA in which the 5:95, 10:90, 25:75,  
 539 and 40:60 mixes of CGB:MS, tested with 200  $\mu$ l drops, are compared). Degrees of freedom (df) and  
 540 the relevant *F* statistics are provided.

Groups Compared	df <sub>within</sub>	df <sub>between</sub>	<i>F</i> <sub>critical</sub>	<i>F</i> <sub>statistic</sub>	<i>P</i> -value
CGB:MS 200 $\mu$ l	366	3	2.63	541.4	$3.62 \times 10^{-134}$
FGB:MS 200 $\mu$ l	400	3	2.63	2548.6	$3.09 \times 10^{-260}$
CGB:CS 200 $\mu$ l	403	3	2.63	41.9	$1.43 \times 10^{-23}$
FGB:CS 200 $\mu$ l	391	3	2.63	90.7	$1.41 \times 10^{-44}$
CGB:MS 20 $\mu$ l	412	3	2.63	171.8	$3.17 \times 10^{-72}$
FGB:MS 20 $\mu$ l	410	3	2.63	257.5	$6.67 \times 10^{-94}$
CGB:CS 20 $\mu$ l	412	3	2.63	105.1	$1.57 \times 10^{-50}$
FGB:CS 20 $\mu$ l	408	3	2.63	188.6	$1.01 \times 10^{-76}$

541

542

543 **Table 4.** Ratio of coarsely ground (CGB) and finely ground (FGB) biochar to medium (MS) coarse  
544 (CS) sand soil surface areas, as calculated from the mean particle sizes.

Mixture	Ratio of biochar to soil surface area			
	CGB:CS	CGB:MS	FGB:CS	FGB:MS
5:95	0.0003	0.0001	0.87	0.31
10:90	0.0006	0.0002	1.83	0.65
25:75	0.0017	0.0006	5.50	1.96
40:60	0.0034	0.0012	11.0	3.92

545

546

547

548 **Figure captions**

549 **Figure 1** Particle-size distributions for: (a) coarse sand (CS) and medium sand (MS), and (b)  
550 coarsely ground biochar (CGB) and finely ground biochar (FGB). Note the horizontal axis is  
551 a logarithmic scale.

552 **Figure 2** Particle-size distributions for coarse sand (CS), medium sand (MS) and 50:50  
553 mixtures of wettable coarse sand (CS<sub>w</sub>) with CS, and wettable medium sand (MS<sub>w</sub>) with MS.  
554 Note the horizontal axis is a logarithmic scale.

555 **Figure 3** Water drop penetration time (WDPT) test results for 200 µl drops on coarsely  
556 ground (CGB) and finely ground (FGB) biochar mixtures with (a) medium sand (MS), and  
557 (b) coarse sand (CS).

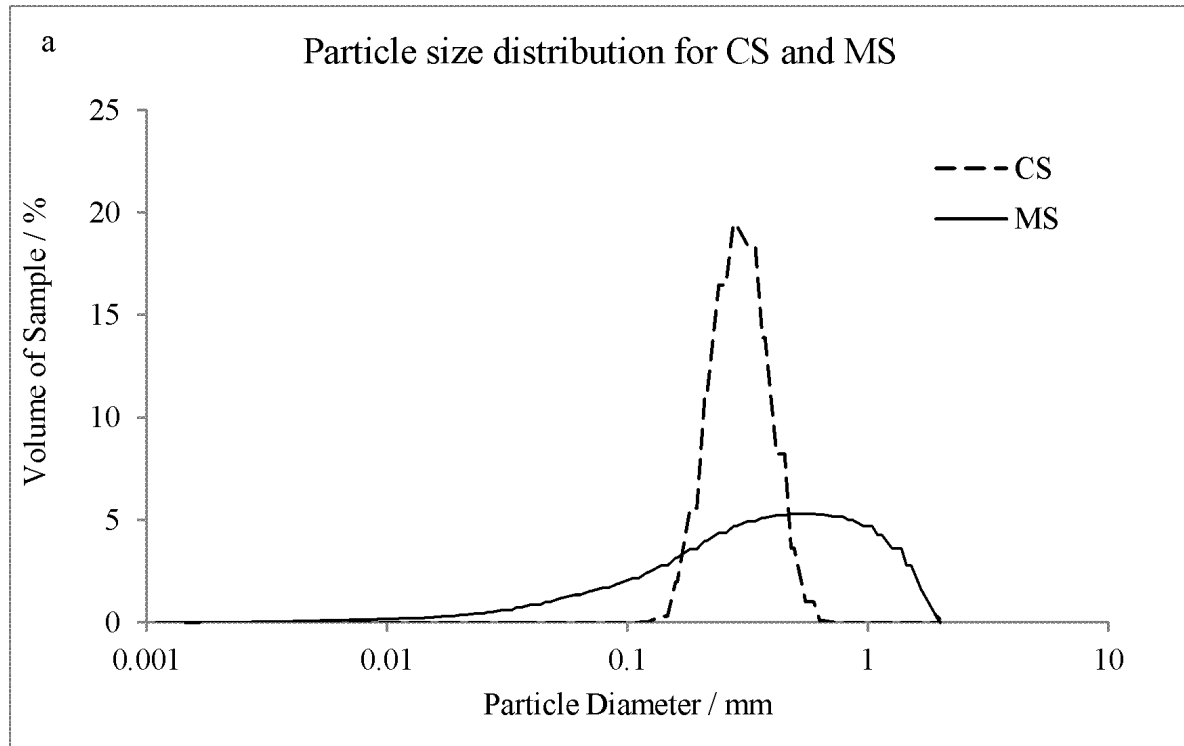
558 **Figure 4** Water drop penetration time (WDPT) test results for 20 µl drops on coarsely ground  
559 (CGB) and finely ground (FGB) biochar mixtures with (a) medium sand (MS), and (b) coarse  
560 sand (CS).

561 **Figure 5** Percentage decrease in water repellency ( $\Delta$ WDPT) of coarse sand (CS) and medium  
562 sand (MS) with increasing quantities of wettable coarse sand (CS<sub>w</sub>), wettable medium sand  
563 (MS<sub>w</sub>), coarsely ground biochar (CGB) or finely ground biochar (FGB) as measured by: (a)  
564 200 µl, and (b) 20 µl drops.

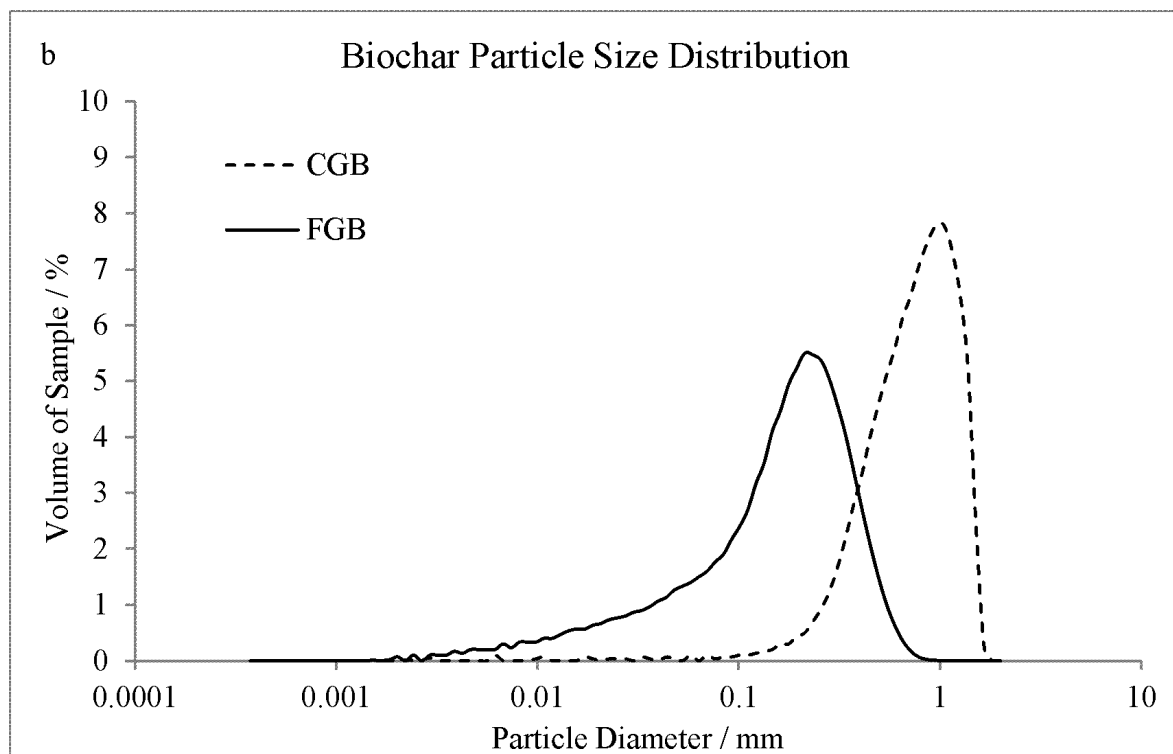
565 **Figure 6** Microphotographs of hydrophobized acid-washed sand mixed with: (a) 1% finely  
566 ground biochar (FGB) and (b) 10% FGB.

567 **Figure 7** Model view of: (a) 200 µl and (b) 20 µl drops applied to the surface of coarsely  
568 ground biochar (CGB) mixtures with coarse sand (CS) and medium sand (MS) (SpherePack  
569 1D (Farr, 2011)). In the packing arrangements spheres represent soil particles and cubes  
570 represent biochar. For both soil and biochar the particle-size distributions used are those  
571 obtained experimentally.

572

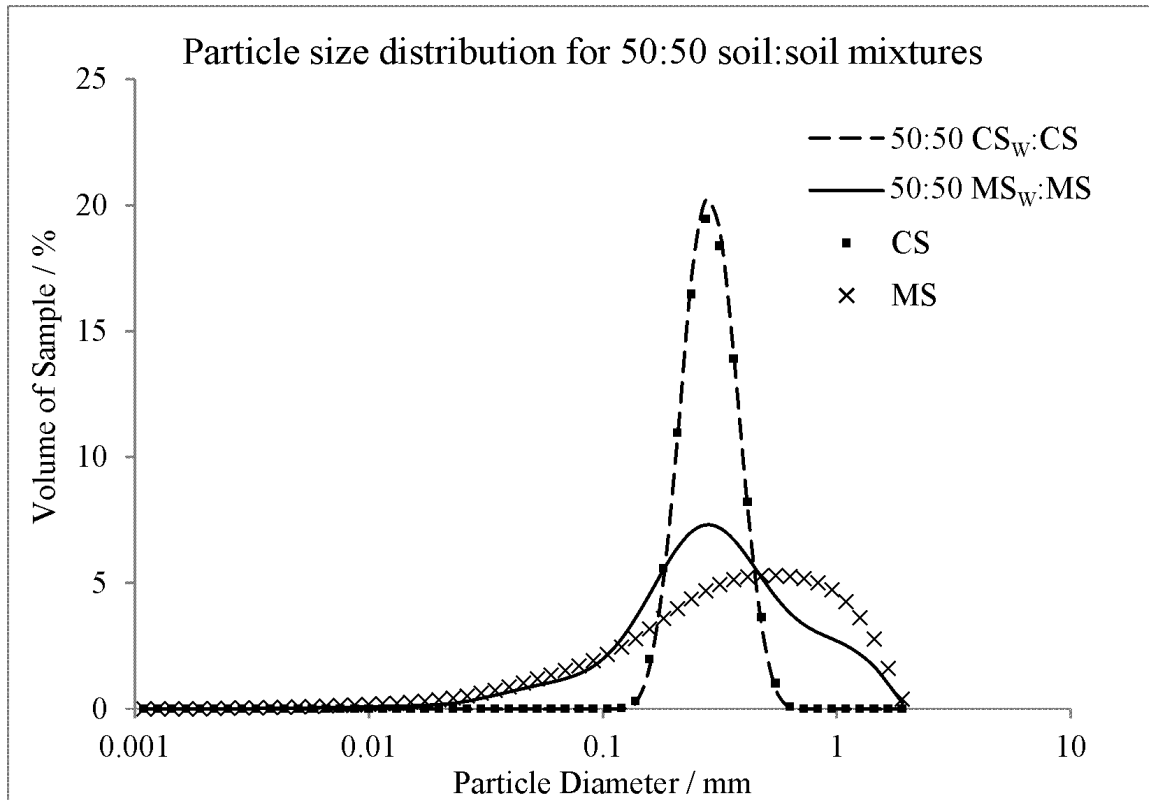


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575 **Figure 1.**

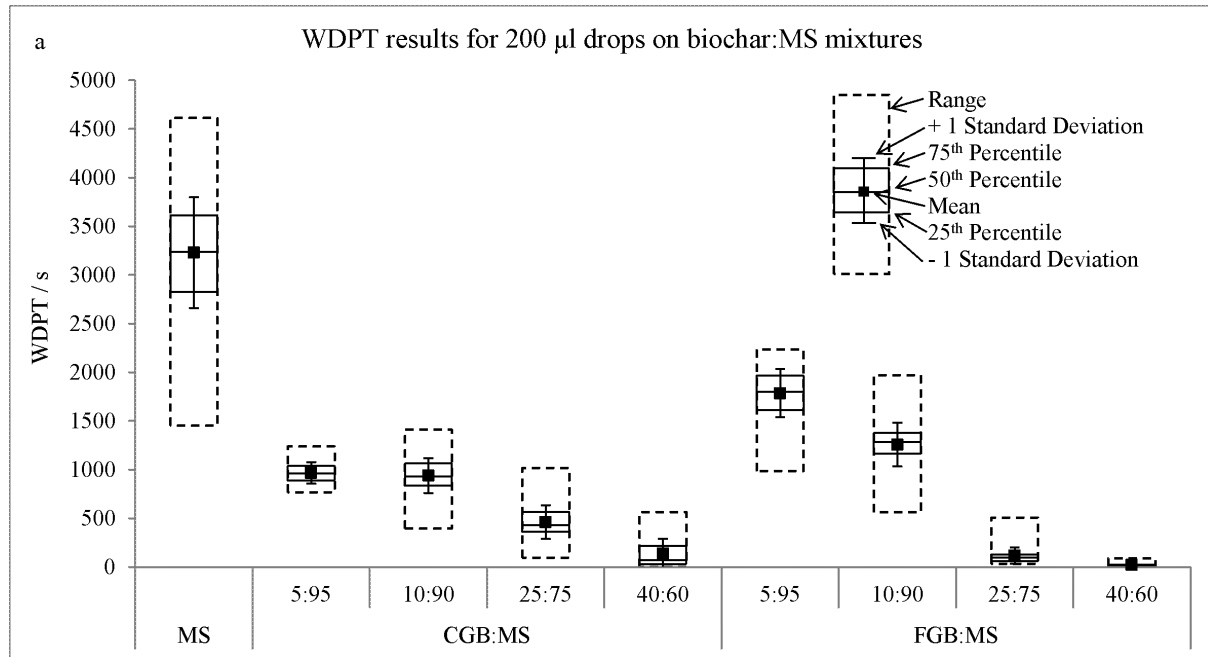


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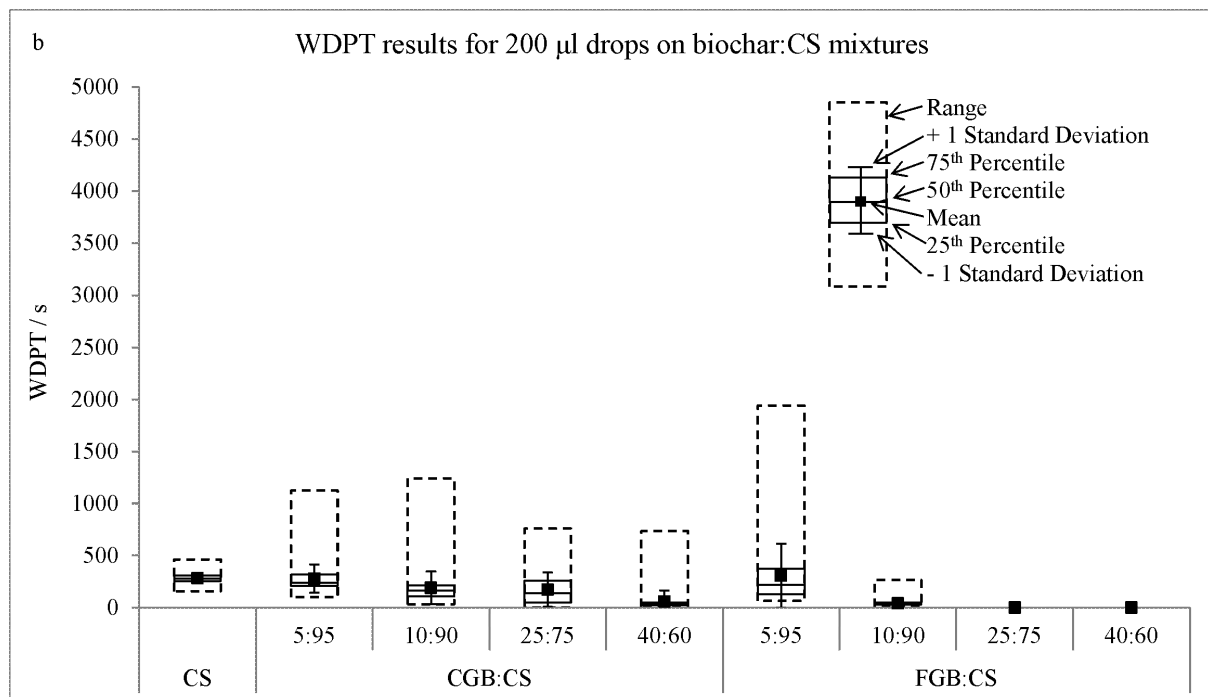
577 **Figure 2.**

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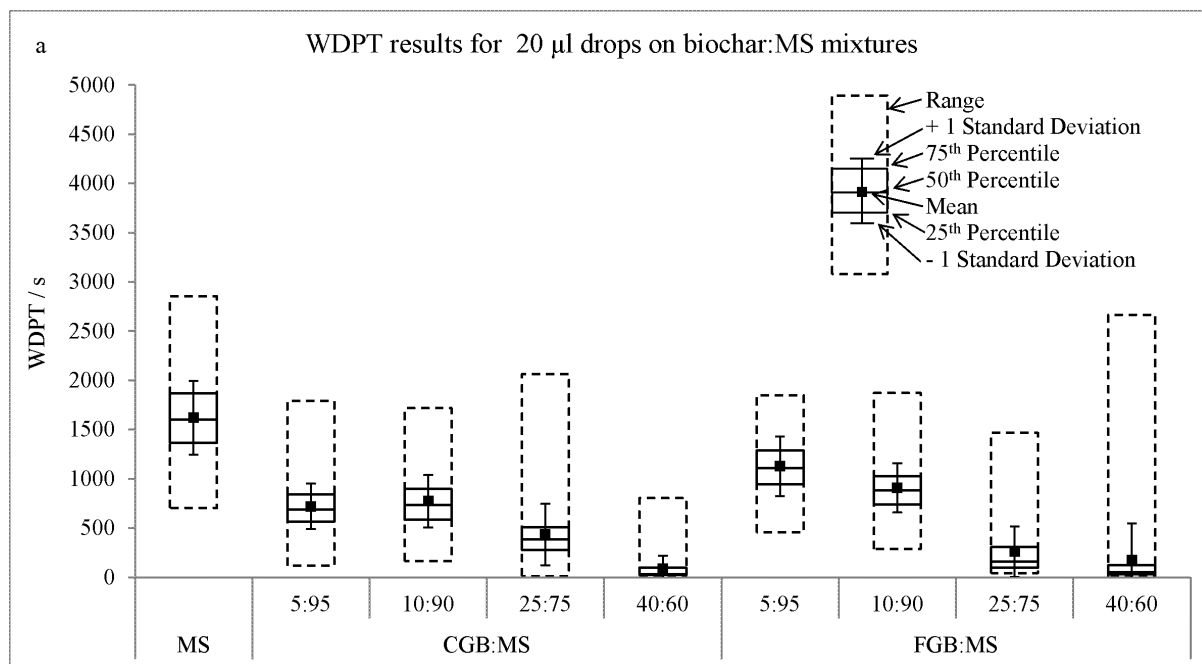
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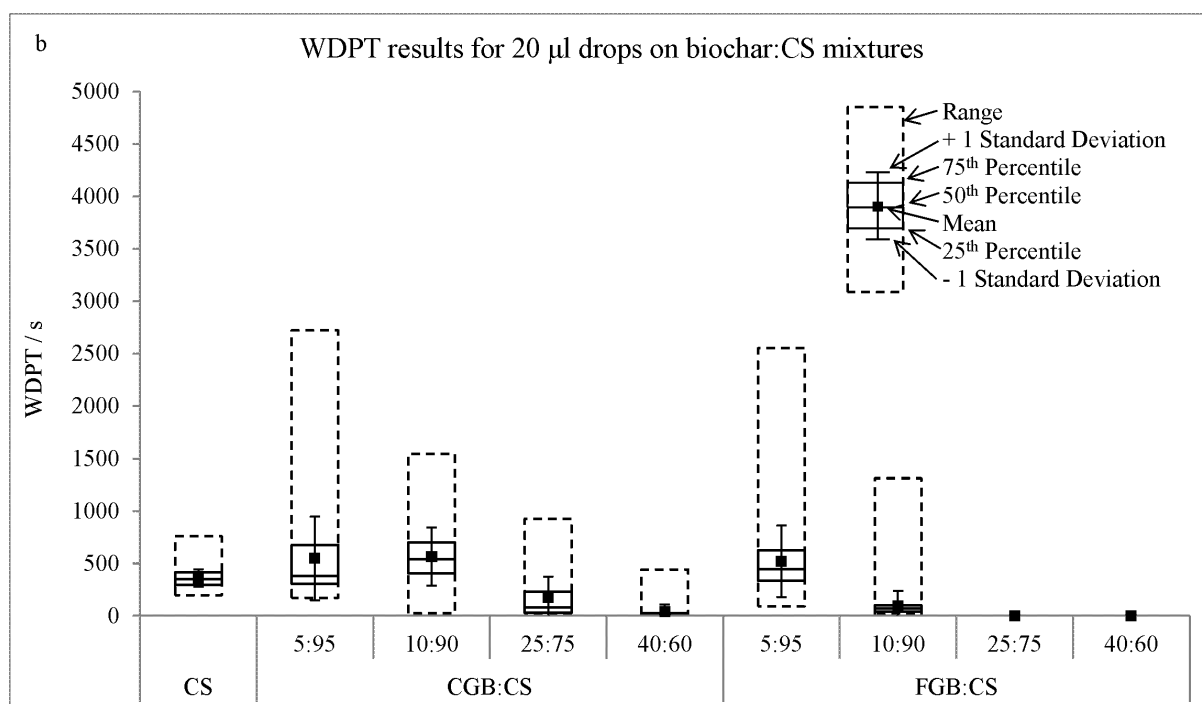
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582 **Figure 3.**

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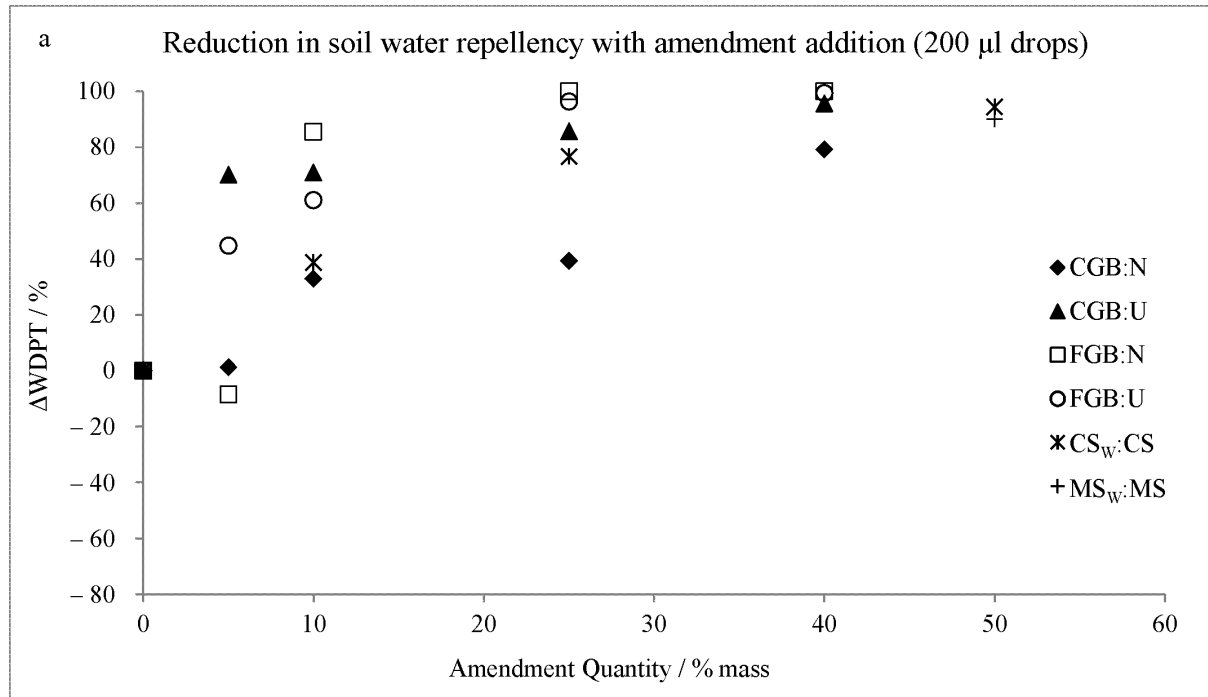


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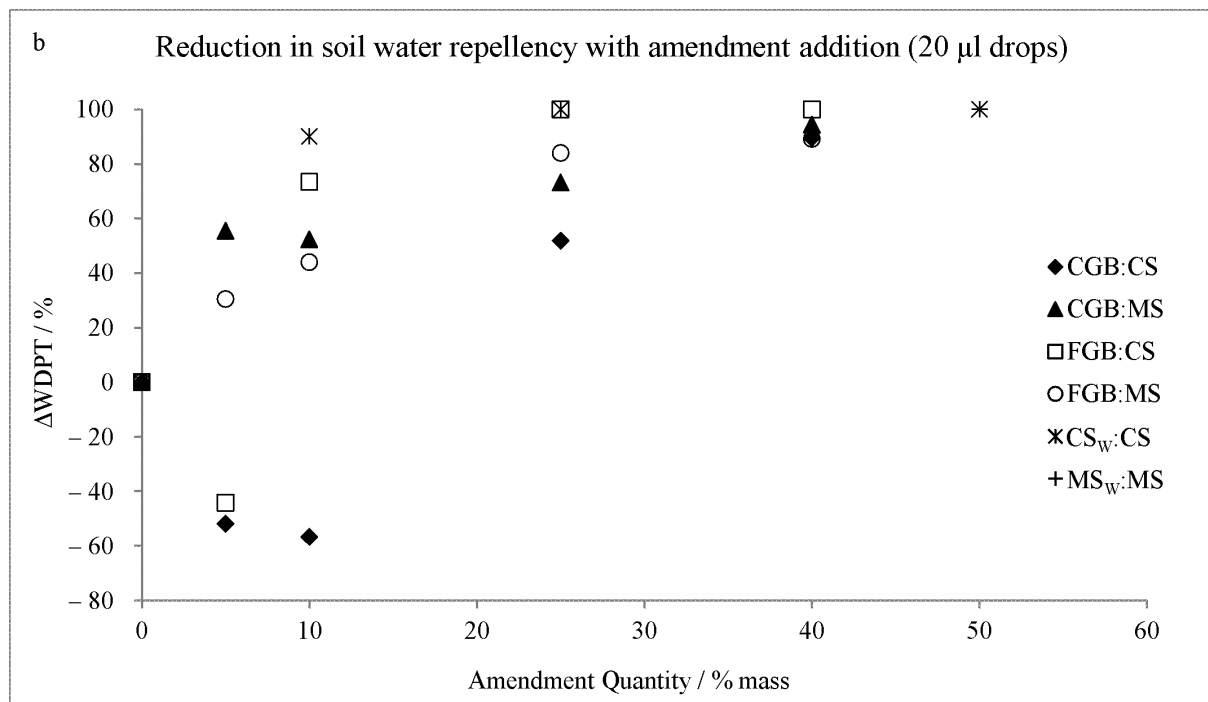
586 **Figure 4.**

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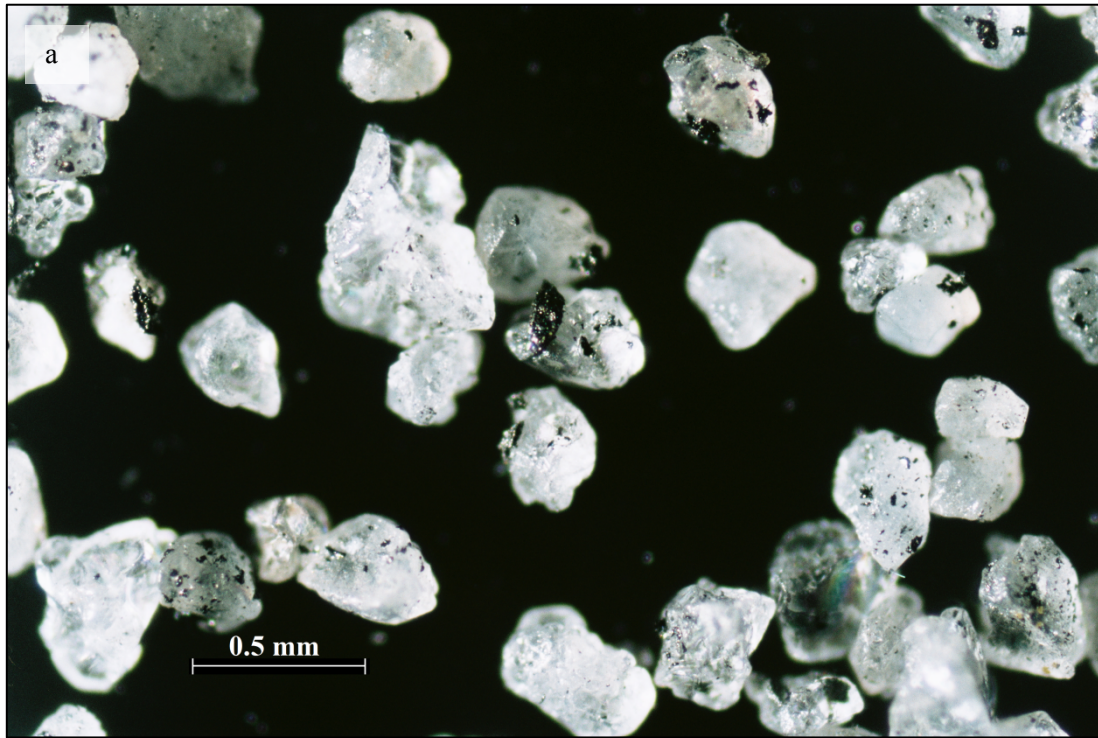


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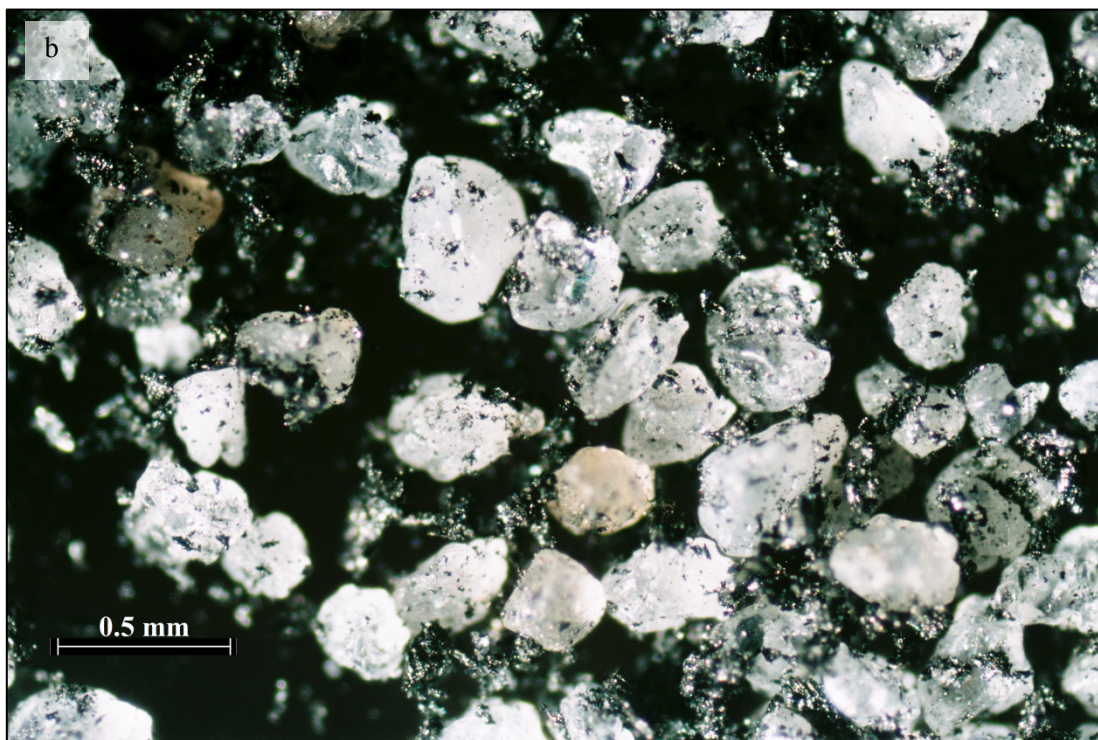


589

590 **Figure 5.**



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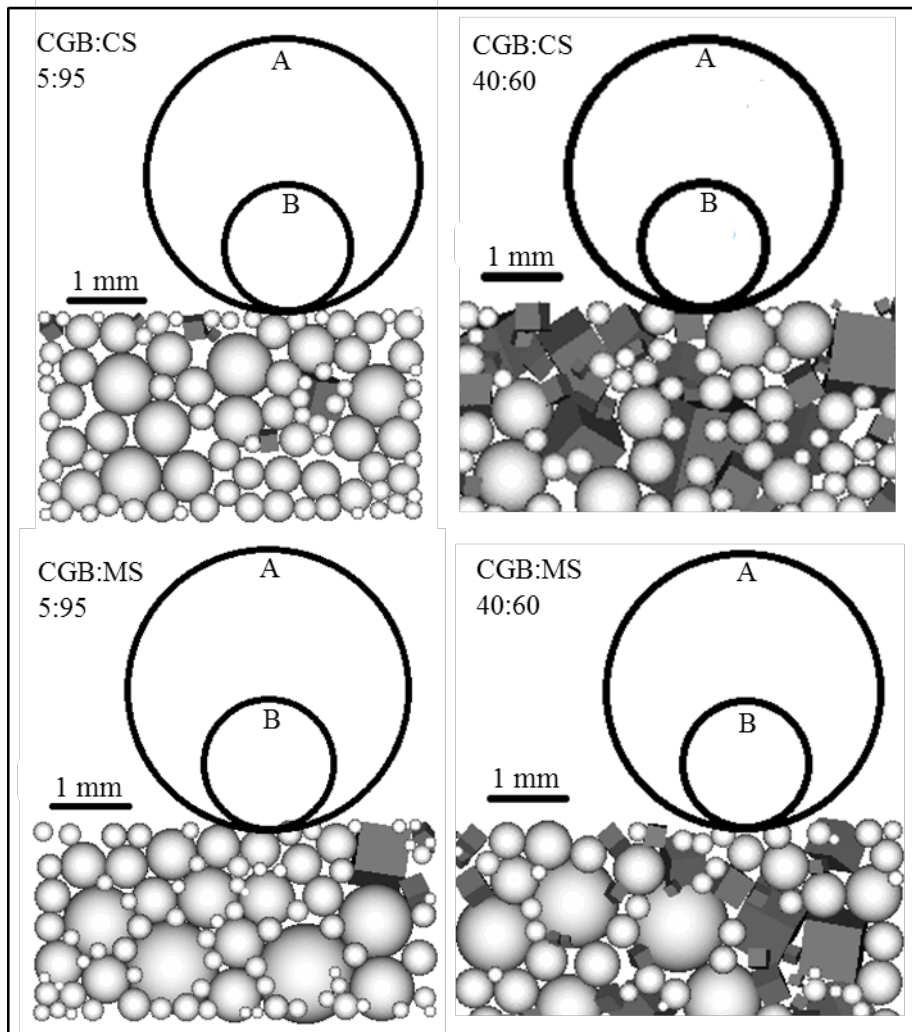
592

593 **Figure 6.**

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598 **Figure 7.**

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