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### **Paper:**

Jiménez-Pinilla, P., Doerr, S., Ahn, S., Lozano, E., Mataix-Solera, J., Jordán, A., Zavala, L. & Arcenegui, V. (2016).  
Effects of relative humidity on the water repellency of fire-affected soils. *CATENA*, 138, 68-76.

<http://dx.doi.org/10.1016/j.catena.2015.11.012>

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27 were water repellent at the lowest heating temperature and became strongly water repellent at  
28 the higher heating temperatures and near saturation (95% RH). Shrubland soils were mostly  
29 wettable at the onset and remained so at every RH level studied except being slightly SWR at  
30 95% RH. A similar trend was found after MED and CA measurements. The results demonstrate  
31 that high RH contributes to enhanced SWR also in burned soils, where high temperatures had  
32 already led to a substantial enhancement of SWR. These findings suggest that SWR levels  
33 determined for fire affected areas ambient under field or laboratory conditions may  
34 underestimate the apparent SWR levels present at the high RH levels that often precede major  
35 storm events. This in turn has implications for predicting post-fire runoff and erosion events.

36

37 *Key words:* Hydrophobicity; Relative humidity; Contact angle; Pre-heated temperatures; Forest  
38 soils; Mediterranean calcareous soils;

39

## 40 **1 Introduction**

41 Soil water repellency (SWR) may be defined as the condition of a soil which does not wet  
42 spontaneously when water comes in contact with the soil surface (Leelamanie et al., 2008a).  
43 This behaviour modifies infiltration and evaporation rates, erodibility and other hydrological  
44 processes of soils (Feng et al., 2001; Jordán et al., 2009; Wallis et al., 1991; Wallis and Horne,  
45 1992). SWR is a common property of soils under many vegetation types and is often induced in  
46 previously wettable soils or enhanced by fire (DeBano, 2000; Doerr et al., 2000; Granged et al.,  
47 2011a; Jordán et al., 2013; Jordán et al., 2014). It has been shown that environmental conditions  
48 such as ambient temperature (King, 1981; Goebel et al., 2011), drying temperature (Franco et  
49 al., 1995; Dekker et al., 1998), water content (Berglund and Persson, 1996; Bodí et al., 2013;  
50 DeJonge et al., 1999; Dekker and Ritsema, 2000) and the wetting and drying history of samples  
51 (Doerr and Thomas, 2000) can strongly affect SWR. Atmospheric relative humidity (RH) is

52 another important factor conditioning SWR. Jex et al. (1985) and Doerr et al. (2002) reported  
53 that SWR increased when soil was exposed to > 90% RH (near saturation) over a short period  
54 (< 1 day), but gradually decreased when exposed to an ambient laboratory atmosphere of ~40%  
55 RH. Leelamanie et al. (2008b) reported a positive correlation between RH and SWR at RHs  
56 between 33 and 94%. Another main factor that recently has been considered responsible of the  
57 severity of SWR is the soil surface structure (Ahn, 2014). SWR appears on low-energy surfaces  
58 where the attraction between the molecules of the solid and liquid interface is weak (Heslot et  
59 al., 1990; Roy and McHill, 2002). Under natural conditions, high-energy soil mineral surfaces  
60 are often covered by films of low-energy organic compounds (Doerr et al., 2000; Goebel et al.,  
61 2004) forming water repellent surfaces (Jiménez-Morillo et al., 2014; Leelamanie et al., 2008a)  
62 and this can be amplified by surface structure (Wenzel, 1936; Cassie and Baxter, 1944). It has  
63 been reported that the overall susceptibility to developing soil water repellency is higher in  
64 sandy and coarser textures (González-Peñaloza et al., 2013; McGuire and Posner, 1980; Roberts  
65 and Carbon, 1971) and lower in those containing clay (Crockford et al., 1991; Zavala et al.,  
66 2014). A soil surface may minimize the contact area with a water drop by its porous structure.  
67 In soils prone to develop water repellency, the upper soil layers tend to exhibit the greatest  
68 severity. This effect is usually enhanced as soil dries and water is lost from the pores, enlarging  
69 the air-solid interface, which critically increases the net contact angle (Ahn, 2014). The contact  
70 angle (CA) between the solid and water can be measured at the three-phase interface (gas-  
71 liquid-solid). According to Goebel et al. (2011) SWR occurs if  $CA > 0^\circ$ ; soils show reduced  
72 wettability with CA varying between 0 and  $90^\circ$  (i.e. infiltration of water into the soil matrix  
73 decreases); and values of  $CA > 90^\circ$  indicate extreme SWR. A zero CA occurs when the surface  
74 tension of solid and liquid are equal to each other (Bachmann and van der Ploeg, 2002).  
75 Although, numerous studies have used CA to determine SWR (Carrillo et al., 1999; Bachmann  
76 et al., 2000a, b; Leelamanie et al., 2008b; Doerr et al., 2009), and some studies exist in which

77 the effect of RH on SWR has been examined (Jex et al., 1985; Doerr et al., 2002; Leelamanie et  
78 al., 2008b). All previous studies have been carried out on unburned soils.

79 Fire is an important ecological agent, which has increasingly affected Mediterranean ecosystems  
80 in the last decades, leading to changes in chemical, physical and microbiological soil properties  
81 (Neary et al., 1999; Certini, 2005). Fire may induce or increase SWR in previously wettable or  
82 water-repellent soils (Doerr et al., 2000; Mataix-Solera and Doerr, 2004; Zavala et al., 2009a),  
83 but can also destroy it after intense combustion of organic matter (Arcenegui et al., 2008;  
84 Granged et al., 2011b; Jordán et al., 2010; Robichaud and Hungerford, 2000). The specific  
85 effect depends mostly on the duration of heating and temperatures reached (DeBano, et al.,  
86 1976; Doerr et al., 2004; Gordillo-Rivero et al., 2014), but also oxygen availability (Bryant et  
87 al., 2005) and soil water content (Robichaud and Hungerford, 2000; Zavala et al., 2010).

88 The influence of increasing temperatures and RH on SWR is of substantial importance in  
89 affecting ecosystem processes, which regulate the soil system during post-fire recovery. Given  
90 the fact that fire-affected soils often exhibit particularly high levels of SWR, the removal of the  
91 protective vegetation cover during fire can make such soils particularly susceptible to  
92 accelerated hydrological and geomorphological responses (Doerr et al., 2009). An important  
93 research gap thus exists in elucidating the effects of RH on SWR of soils that have been  
94 exposed to heating during vegetation fires.

95 The main aim of this study was therefore to explore the effect of different ambient RHs on SWR  
96 variations that might prevail under very dry surface conditions following a wildfire. We focused  
97 here on a Mediterranean calcareous soil under *Pinus halepensis* and mixed shrub vegetation,  
98 which is a common soil-vegetation combination subjected to fire in Eastern Spain (Mataix-  
99 Solera et al., 2002; Arcenegui et al., 2008; Mataix-Solera et al., 2013; Jiménez-Pinilla et al.,  
100 2015).

101

## 102 2 Material and methods

### 103 2.1 Sampling site, soil sampling and experimental design

104 The sampling site is located in a formerly cultivated area in Gorga (N 38° 43'44'', W 0°  
105 22'58''; 545 masl), province of Alicante (SE Spain), with a Mediterranean climate type and  
106 approximate annual average rainfall of 500 mm. Vegetation type is mainly composed of *P.*  
107 *halepensis* forest and an understory stratum formed by Mediterranean shrubs, including  
108 *Quercus coccifera*, *Rosmarinus officinalis*, *Cistus albidus* and *Erica arborea*. The soil is  
109 classified as a Lithic Xerorthent (Soil Survey Staff, 2014), developed over limestone with a silt  
110 loam texture (49.7% sand, 40.7% silt and 9.6% clay). Surface soil samples (0 - 2.5 cm depth)  
111 were collected beneath either well-demarcated pine or shrub areas after carefully removing any  
112 superficial litter by hand. Samples were stored in plastic bags, transported to the laboratory, air  
113 dried at room temperature (~25 °C) for one week and then carefully sieved through a < 2-mm  
114 mesh.

115 Soil samples were homogenized before the heating procedure. Then triplicate soil samples (~30  
116 g) were heated at selected temperatures under controlled laboratory conditions (50, 100, 150,  
117 200, 250, 300 and 350 °C), during 20 minutes in ceramic crucibles using a muffle furnace  
118 (Nabertherm, P320, Bremen, Germany). This range of temperatures was selected in order to  
119 simulate different potential heating scenarios of wildfires and also in agreement with previous  
120 studies which have shown notable heat-induced increases in SWR (Neary et al., 1999;  
121 Bachmann et al., 2003; Doerr et al., 2005a; Mataix-Solera et al., 2011). For each case, the  
122 furnace was pre-heated to the desired temperature and each sample was heated separately. The  
123 experiment includes also unheated control samples.

124

125 2.2 Exposure to selected relative humidities and associated water repellency measurements  
126 using WDPT and MED tests

127 For each experiment, each heated soil sample was divided into 3 subsamples (10 g,  
128 approximately), which were then put in petri dishes (5-mm diameter and 7-mm depth), and  
129 placed inside a climate chamber (SANYO Gallenkamp, model PLC CF4; range: 30-90% RH  
130 and -40 to 180 °C temperature) and subsequently exposed for equilibrium under a different  
131 prescribed atmospheric conditions of 30, 50, 70 and 95% RH at 20 °C. Each sample type, in  
132 triplicate, was kept for 48 hours inside the chamber prior to the water repellency assessments, to  
133 ensure the entire sample was fully adjusted to the selected humidity. Soil samples remained  
134 within the sealed climate chamber throughout the experiments, with manipulations and  
135 measurements conducted by using gloves attached to sealed portholes in the chamber window.  
136 This ensured that samples remained under constant environmental conditions and without any  
137 other environmental variables affecting them.

138 Persistence of SWR was assessed using the water drop penetration time (WDPT) test, which  
139 measures how long SWR persists on a porous surface. Given that SWR usually decays with  
140 prolonged water contact, it relates to the hydrological implications of reduced wettability as the  
141 amount of surface runoff is affected by the time required for the infiltration of raindrops  
142 (Wessel, 1988; Doerr, 1998). It involved placing three drops of distilled water ( $\sim 0.02 \pm 0.05$  mL)  
143 onto the soil sample surface. In each case, the time (in seconds) required for a complete droplet  
144 infiltration was recorded and a SWR persistence class assigned according to Bisdom et al.  
145 (1993) (Table 1).

146 Severity of SWR was also assessed inside the chamber using the molarity of an ethanol droplet  
147 (MED) test (expressed as % ethanol; Doerr et al., 1998) (Table 2). This test is an indirect  
148 measure of the surface tension of the soil surface and indicates how strongly a water drop is  
149 repelled by a soil at the time of application (King, 1981; Doerr, 1998). It involved placing 3  
150 droplets ( $\sim 0.02 \pm 0.05$  mL) using an applicator of water-ethanol solution (0, 1, 3, 5, 8.5, 13, 18,  
151 24, 36 % ethanol) (Table 2) onto the soil samples surfaces. The number of droplets that  
152 penetrate the soil within 5 seconds (Crockford et al., 1991) was recorded for each ethanol

153 solution and the solution that allowed more than one drop to penetrate was then taken to assign  
154 a SWR severity class (Doerr, 1998) (Table 2).

155

### 156 2.3 *Additional water repellency assessments using advancing contact angle measurements*

157 The advancing contact angle (CA) of a water droplet on a soil can also be used to determine the  
158 severity of SWR (Letey, 2000; Leelamanie, 2008a). In surface science, a CA of 90° is usually  
159 taken as demarcating water repellent and wettable conditions on flat media (Letey, 2000). For  
160 porous granular media such as soils, it has been suggested that reduced wettability is present for  
161  $0^\circ < CA < 90^\circ$ ; and values of  $CA > 90^\circ$  indicate extreme SWR (Goebel et al., 2011; Fig. 1).  
162 More specifically, Shirtcliffe et al. (2006) found that the critical contact angle for penetration  
163 was 52-61° using soil models and test liquids (pentane and hexane), but years before Bán et al.  
164 (1987) had determined CA even lower (49.5-57.7°) using ethanol and sulphur pellets,  
165 polyamide and polytetrafluoroethylene. CA measurements were included in this study as third  
166 measure of SWR (i) to provide wider comparability of the SWR data obtained here and (ii) as  
167 CA measurements have not been included in any previous study in which the effect of RH on  
168 SWR had been examined.

169 To measure the CA of a sample, a monolayer of soil particles was immobilized on a microscope  
170 glass slide (1.5 cm × 3 cm) using double-sided adhesive tape to accomplish a plain  
171 homogeneous surface (Bachmann et al., 2000b). Samples were then placed on a tray inside the  
172 sealed climate chamber and exposed to the selected RH's as in the previous experiments, except  
173 that samples were only exposed for 24 hours given that only a monolayer of soil was used.

174 We also included two different hydrophobic control 'model soil' materials in this experiment:  
175 spherical glass beads to represent smooth-spherical particles (obtained from Whitehouse  
176 Scientific, UK); and natural quartz sand (collected from fluvial deposits, South Wales, UK) to  
177 represent textured-angular particles typical of soils. Both materials were used in two different



178 size ranges: 710-850  $\mu\text{m}$  and 1120-1180  $\mu\text{m}$  and were chemically hydrophobized using  
179 chlorotrimethylsilane as described in Ahn et al. (2013), resulting in static (temporally stable)  
180 water repellency. They were then packed as closely as possible and immobilized on the glass  
181 slides as described above.

182 It is now well established that hydrophobicity can be enhanced by the micro-topography of a  
183 surface when compared to flat surfaces (McHale et al., 2005). For particle arrays as presented  
184 by a soil surface, the texture of the particle surface itself seems to have a significantly greater  
185 influence in the amplification of the CA (i.e. and hence SWR) than the coarser-scale particle-  
186 based structure or particle shape (Bachmann et al., 2006; Ahn, 2014). Hence these ‘model soil’  
187 materials, varying in surface texture (glass beads: spheres with no particle surface texture and  
188 sand: textured-angular particles), were included to explore the fundamental physical impact of  
189 RH and surface texture on SWR behaviour for a static (i.e. chemically more stable) hydrophobic  
190 coating that is expected not to be altered by microbial activities. .

191 For CA assessment, drops of distilled water were placed onto the surfaces of soils and  
192 chemically treated materials using a 1-mL syringe with a blunt tip needle (19 gauge Luer-Lock  
193 blunt ended needles, Sylmaste, UK). The advancing contact angles of sessile drops were  
194 recorded by a video camera (30 fps) while the water drop was being gradually enlarged (~10-  
195 100  $\mu\text{L}$ ). The left and right hand side contact angles of each advancing moment were  
196 determined using the software *ImageJ* (Version 1.48, National Institutes of Health, USA) and  
197 averaged, and then 3-5 advancing CAs were averaged from each sample in triplicate. All  
198 samples remained in the sealed climate chamber throughout the experiments, as described  
199 above.

200

#### 201 2.4 Statistical analysis

202 The normal distribution of SWR data was checked using the Kolmogorov-Smirnov test. As

203 normality of SWR data was rejected ( $p < 0.05$ ), WDPT data were transformed (Ln) to allow a  
204 better data display given their large variability. Mann-Whitney U test was used to check the  
205 statistical differences between SWR data at each temperature treatment regarding to unheated  
206 (control) samples for every RH studied ( $p < 0.05$ ). Statistical analyses were performed by IBM®  
207 SPSS (© SPSS Inc., 1989) version 22 for Windows software package.

208

### 209 3 Results

#### 210 3.1 Heating effects on soil water repellency at a standard humidity (50% RH)

211 Heating has a similar effect for both persistence (WDPT test) and severity (MED and CA tests)  
212 of water repellency for samples at a standard laboratory RH of 50%. For the WDPT test, an  
213 apparent slight increase in SWR for pine soils was observed at 50 °C compared to the control  
214 samples (unheated), and also an apparent decrease between 100 and 200 °C, although in both  
215 cases, no significant differences were observed ( $p > 0.05$ ) (Fig. 2a). For the highest heating  
216 temperatures (300-350 °C), a substantial increase in SWR was observed, with significant  
217 differences ( $p < 0.05$ ), with the peak occurring at 350 °C (Fig. 2a). SWR was not found at 50%  
218 RH in shrub soils for any heating treatment (Fig. 2b).

219 The MED tests showed a similar pattern with WDPT tests for both pine and shrub soils (Fig. 3a  
220 and 3b, respectively). Nevertheless, significant differences ( $p < 0.05$ ) were found on SWR pine  
221 samples from 250 °C to 350 °C compared to the unheated samples. SWR was not detected by  
222 MED tests in pine soils heated at 150 and 200 °C (Fig. 3a).

223 The results of CA measurements showed a similar pattern with the WDPT results. CA  
224 measurements at 50% RH showed a general trend of increasing SWR with heating temperatures  
225 showing the largest CA at the highest heating range, where significant differences were found  
226 from 200 to 350 °C with regarding to unheated (control) samples ( $p < 0.05$ ) (Fig. 4).

227

228 3.2 *Relative humidity effects on soil water repellency persistence (WDPT test)*

229 Persistence of SWR in pine soils increased with RH for all heating temperatures. The WDPTs  
230 were longest at the highest RH (95%) for the most of heating treatments (Fig. 2). Particularly,  
231 for the heating treatments at 300 °C, the ln WDPT increased from 5.1 s at 50% RH to 8.6 s at 95%  
232 RH, and for the treatment at 350 °C, the ln WDPT increased from 5.1 s at 50% RH to 8.4 s at 95%  
233 RH (Fig. 2a), being in both cases significantly different ( $p < 0.05$ ). For these samples for the  
234 300 °C and 350 °C treatments were ‘severely water repellent’ (ln WDPT 6.4-6.8 s in Table 1) at  
235 30% RH (Fig. 2a), whereas ‘extremely water repellent’ (ln WDPT  $> 8.2$  s in Table 1) at 95%  
236 RH (Fig. 2a).

237 Shrub soils were wettable at the onset (ln WDPT  $< 1.6$  s) and remained stable at each  
238 considered RH except at 95% RH, where the samples heated at 250 °C and 300 °C were  
239 ‘slightly water repellent’ (ln WDPT 1.8-3.4 s in Table 1; Fig. 2b), and there were statistically  
240 significant differences ( $p < 0.05$ ).

241

242 3.3 *Relative humidity effects on soil water repellency severity (MED and CA tests)*

243 For the MED test, an influence of RH on SWR was observed mainly for the pine soils, showing  
244 a consistently increasing severity of water repellency with increasing RH (Fig. 3). At 30 and  
245 70% RH, severity of SWR ranged between very wettable and strongly water repellent (classes 1  
246 to 6; Table 2), with the highest values occurring for the highest temperature treatments (Fig. 3a),  
247 and where also statistically significant differences were detected for both RHs at 250 °C and 300  
248 °C ( $p < 0.05$ ). At 95% RH (Fig. 3a), SWR ranged between moderately and extremely water  
249 repellency (classes 5 to 9; Table 2), and the increase of SWR detected at 300 °C was statistically  
250 significant different ( $p < 0.05$ ).

251 In contrast, the influence of RH on SWR was not shown from the shrub soils at low RH levels,  
252 as the shrub soils were wettable at the onset (ethanol  $< 0\%$ ), and remained so at every RH level

253 monitored except at 95% RH. Even at 95% RH, only the samples heated at 250, 300 and 350 °C  
254 (Fig. 3b) were ‘slightly water repellent’ (class 4; Table 2), and with no significant differences ( $p$   
255  $> 0.05$ ).

256 The CA measurements were only conducted on samples taken from the pine site as only these  
257 showed a resistance to wetting sufficient for conducting CA measurement ( $LN_{WDPT} > 1.6$  s).  
258 Shrub soils had rapid infiltration of water droplet, and thus, CAs were not technically  
259 measurable.

260 The CA measurements for the pine soils also showed an effect of RH at all heating temperatures  
261 at 30 and 70% RH (Fig. 4). The most noticeable increase in CA was found in the samples  
262 heated between 250 and 350 °C, particularly for 30 and 70% RH (Fig. 4), where significant  
263 differences were also detected ( $p < 0.05$ ). The highest CA was observed at 95% RH after all  
264 heating treatments (Fig.4), finding significant differences ( $p < 0.05$ ) from 200 °C forward.

265

#### 266 3.4 Relative humidity effects on ‘model soils’: glass beads and sand

267 Both glass beads and sand showed a similar pattern where CA increased with RH (Fig. 5). CAs  
268 observed on glass beads of different sizes ranged from 77.91° (66.5°, 85.6°) to 100.9° (88.5°,  
269 107.9°) (Fig. 5a), and on sands from 117.7° (100.0°, 126.0°) to 125.9° (118.3°, 128.9°) (Fig. 5b).  
270 The CAs of glass beads at different relative humidities all indicated water repellency (according  
271 to the classification of Goebel et al., 2011). The CA of the glass beads of size 1120-1180 µm at  
272 95% RH was 100.9° (88.5°, 107.9°), indicating a fully water repellent behaviour ( $CA > 90^\circ$ )  
273 (Fig. 5a). The sand of both size groups also had consistent water repellent behavior ( $CA > 90^\circ$ ).  
274 The CA was 123.2°(111.3°, 127.1°) for size 710-850 µm and 125.9°(118.3°, 128.9°) for size  
275 1120-1180 µm (Fig. 5b). Regarding the differences between two sizes of sands, we observed  
276 that CAs were slightly larger for the smaller particles (710-850 µm) compared to the larger ones  
277 (1120-1180 µm) at 30, 50 and 70% RH, and the opposite at 95% RH (Fig. 5b).

278

## 279 4 Discussion

### 280 4.1 Heating effects on soil water repellency at different pre-established laboratory relative 281 humidities

282 An overall increasing trend of both persistence (determined using the WDPT test) and severity  
283 (MED and CA tests) of SWR with increasing heating temperature was detected from pine soils  
284 at a standard RH of 50%. More specifically, SWR increased somewhat at the lowest heating  
285 temperature (50 °C) compared to unheated samples (Fig. 2a, 3a, 4). The SWR increasing can be  
286 caused by increased alignment of hydrophobic molecule chains (Doerr et al., 2005b). This was  
287 followed by a slight reduction for 100 and 150 °C treatments. The reason for this is unclear.  
288 Following heating at 200-300 °C a strong increase in SWR was observed (Fig. 2a, 3a, 4) which  
289 is in agreement with established knowledge based on previous laboratory studies, which have  
290 shown intensification of SWR when soil temperatures reach 175-270 °C (Doerr and Shakesby,  
291 2009). Amongst the suggested causes for strong increases in SWR by heating are migration of  
292 hydrophobic material from interstitial organic matter onto soil particles (Franco et al., 1994) and  
293 chemical changes such as the relative increase in long chain (<C20) fatty acids aromatic  
294 compounds (Atanassova and Doerr, 2011). Krammes and DeBano (1965) found that SWR is  
295 destroyed at temperatures between 280 and 400 °C, with the specific destruction temperature  
296 being closer to 280 °C for shorter heating durations (Doerr et al., 2004). In the current study,  
297 however, heat treatments at 350 °C seem to be remained below the threshold for SWR  
298 destruction.

299 Shrub soils remained wettable after all heating treatments, highlighting the importance of  
300 vegetation type on the development of water repellency. It has been previously reported in  
301 burned (Arcenegui et al., 2007; Mataix-Solera et al., 2008) or unburned soils (Jordán et al.,  
302 2008; Martínez-Zavala and Jordán-López, 2009; Zavala et al., 2014).

303

304 4.2 *Relative humidity effects on the persistence and severity of soil water repellency*

305 Persistence and severity of SWR were substantially enhanced at the highest RH applied in our  
306 experiments. At 95% RH, SWR in pine soils exhibited the highest values for each heating  
307 temperature (Fig. 2a, 3a, 4), and the shrub soils that were wettable at all other RHs turned into  
308 slightly water repellent at 95% (Fig. 2b, 3b).

309 A notable increase in SWR also occurred for pine soils at 70% RH compared to the standard  
310 atmosphere of 50% RH, particularly when considering SWR severity measured by MED test.  
311 This was evident in all samples irrespective of heating temperatures (Fig. 3a). These results  
312 demonstrate a considerable enhancing influence of high RHs (near atmosphere saturation: 95%  
313 RH) on SWR persistence and severity of already water-repellent soil. This fact suggests that air  
314 humidity is a key control of SWR. It supports findings from previous work on a variety of  
315 unburned soils (Jex et al., 1985; Doerr et al., 2002; Leelamanie and Karube, 2007; Leelamanie  
316 et al., 2008b), and demonstrates that this also applies to heat-affected soils. Given the relatively  
317 rapid change observed also in previous laboratory tests, Doerr et al. (2002) proposed that  
318 physicochemical rather than microbiological processes may be responsible for this soil  
319 behaviour. They suggested that the prior interactions between soil components and water vapour  
320 at high RH may be of significant influence, as a considerable quantity of energy is released,  
321 which might be disrupting the mineral and organic hydrophobic bonds within the soil. They  
322 speculated that, similar to heating effects, the condensation and adsorption of water vapor onto  
323 soil associated with an increase in RH might lead to reorientation of the hydrophobic organic  
324 molecule chains, expanding more strongly into pore spaces, thereby enhancing SWR.

325 In this study, the heated samples, in which SWR had already been increased through the effect  
326 of temperature, showed further enhanced SWR by high RHs. Considering that high temperature  
327 treatments up to 300 °C would have led to a substantial reduction in any viable soil microbial

328 population (Guerrero et al., 2005; Mataix-Solera et al., 2009), thereby limiting the growth of  
329 both bacteria (Díaz-Raviña et al., 1996) and also fungi (Widden and Parkinson, 1975), the  
330 intensified SWR by RH does not seem to be driven by increased microbial activity within a  
331 relatively short-duration of exposure to high humidities (Doerr et al., 2002).

332 It is also noteworthy that a slight increase in SWR was observed for pine soils at 30% RH,  
333 compared to the standard atmosphere of 50% RH. This was less consistent across the heat-  
334 treated soils and was evident predominantly from WDPT and MED tests (Fig. 2a, 3a), and also  
335 from CA test for control treatment (Fig. 4). This behaviour might be related to a conclusion of  
336 Moody and Ebel (2012), in where the time scale for rewetting of fire-affected soils under  
337 “hyper-dry” conditions is much slower (days to weeks) than the time scale of rainstorms  
338 (minute to hours). RHs of 30% can be considered a threshold for hot dry summer weather  
339 conditions in the Mediterranean climate (Harpaz et al., 2014), and this behaviour may therefore  
340 be relevant to the study region.

341

#### 342 4.3 Assessments of relative humidity effects on ‘model soils’: glass beads and sand

343 The observed increase in SWR severity of hydrophobic ‘model soil’ samples after a short  
344 exposure (24 h) to relatively high RHs (> 50%) (Fig. 5a, b) suggests that the enhanced water  
345 repellency with RH observed here is a generic effect by a physical mechanism. The ‘model soils’  
346 were hydrophobized with a chemically relatively stable compound, and thus there is lowest  
347 possibility for microbial activities to get involved for these materials.

348 It supports the conclusion already stated above that physical rather than microbial processes are  
349 responsible for enhanced SWR at high RH.

350

#### 351 4.4 Implications of enhanced soil water repellency for pre-heated soils with relative humidity

352 SWR is a widespread phenomenon with the capacity to condition hydrological and  
353 geomorphological processes (Whelan et al., 2015). It can reduce infiltration rates (Wallach and  
354 Graber, 2007) and water retention capacity (Naasz et al., 2005). Important environmental  
355 consequences of the reduced infiltration rate include increased soil erosion, enhanced overland  
356 flow and non uniform wetting fronts with fingered flow (Burch et al., 1989; Imeson et al., 1992;  
357 Ritsema et al., 1993; Jordán et al., 2008; Zavala et al., 2009b). These environmental impacts can  
358 affect important ecosystem processes such as soil organic matter decomposition, plant  
359 regeneration, succession of vegetation and microbial activities. The intensity of these effects  
360 depends upon the severity and spatial variability of SWR (Jungerius and de Jong, 1989; Ritsema  
361 and Dekker, 1994) and its severity and persistence (Zavala et al., 2009b). The enhanced SWR  
362 by high RH found in this study, therefore, can result in more intensive environmental change  
363 than the generic SWR under dry conditions. In nature, high RH conditions often precede  
364 precipitation events or wetting front advances in the soil, which in turn could increase SWR  
365 (Doerr et al., 2002; Zavala et al., 2009b).

366 The results of the current study suggest that this will also be the case for soils in which water  
367 repellency was enhanced by soil heating during fire. Considering that the removal of the  
368 protective vegetation cover by fire increases the susceptibility of burned hillslopes to  
369 accelerated hydrological and erosional responses (Shakesby and Doerr, 2006), the enhancement  
370 of SWR at high RH may be even more influential in recently burned compared to unburned  
371 environments.

372

## 373 **5 Conclusions**

374 To the authors' knowledge, this is the first study in which RH effects on SWR have been  
375 examined for heat-affected (burned) soils. It was found that heat-affected soils also show the  
376 enhanced SWR that has been previously reported for soils not affected by heating. This effect



377 was observed at 70% RH and was most pronounced at 95% RH. The effect of RH was shown in  
378 all heat-treated samples, and most pronouncedly in the samples heated at high temperatures  
379 (250-300 °C) that already had a substantial increase in SWR. The fact that the latter soils, as  
380 well as model soil particles treated with a hydrophobic coating, behaved in this manner suggests  
381 that physicochemical rather than microbial effects are responsible for the rapid increases in  
382 SWR induced by RH. The finding that soils in which SWR has already been enhanced by  
383 heating exhibit further increases with RH emphasizes the importance of considering RH when  
384 investigating the environmental impact of SWR in burned areas. Furthermore, a fire burns off  
385 protective vegetation and litter, and makes burned areas even more vulnerable to hydrological  
386 and erosional responses. Enhanced SWR at high RH could, therefore, be of greater significance  
387 than in unburned vegetated terrain.

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### 389 **Acknowledgments**

390 This research has been supported through the HYDFIRE (CGL2010-21670-C02-01) and the  
391 POSTFIRE (CGL2013- 47862-C2-1-R) Research Projects, funded by the Spanish Ministry of  
392 Science and Innovation and the Spanish Ministry of Economy and Competitiveness,  
393 respectively. Patricia Jiménez-Pinilla and Elena Lozano acknowledge the Spanish Ministry of  
394 Economy and Competitiveness and the Regional Valencian Government for their respective  
395 fellowships. Authors thank Jorge Moltó for his help with the initial field and laboratory work at  
396 University Miguel Hernández (Spain) and Grahame Walters for his invaluable laboratory  
397 support with the climate chamber at Swansea University (UK). They also are grateful to the two  
398 reviewers and editor for their valuable comments suggested that helped to enrich the  
399 manuscript. In loving memory of Juan Ramón. The authors are grateful to him and his family  
400 for their help in the study area.

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