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Additional Information

1 **Influence of rainfall intensity and pollution build-up levels on water quality and quantity response**
2 **of permeable pavements**

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7

8 **Keywords:** permeable pavement; road deposited sediments; deposition rate; metals; leachability.

9 **Abstract**

10 Permeable pavements are part of stormwater management practices known as sustainable urban
11 drainage systems (SUDS). This study describes the influence of several environmental variables, such
12 as the rainfall regime or the pollution build-up level, on the hydraulic and water quality performance
13 of permeable pavements. Four infiltrometers with different configurations of pavement layers were
14 used to study the influence of two rainfall regimes (Atlantic and Mediterranean) and two rainfall
15 intensities (0.5 and 2.2 mm/min). The influence of the progressive pollution build-up level was studied
16 by dry sprinkling of road deposited sediments collected with a mechanical street sweeper with a dose
17 of 5 g/m²/d. The results show that permeable pavements retained a significant rainwater volume and
18 improved the infiltrated water quality in terms of suspended solids, organic matter and nutrients when
19 compared to the corresponding surface runoff potentially generated from an impervious pavement.
20 The volume of rainwater retained inside them varied between 16 and 66% depending on the variables
21 studied. The water infiltrated from permeable pavements subjected to a Mediterranean rainfall
22 regime contained, in general, higher concentrations of organic matter (22 to 89 mg Chemical Oxygen
23 Demand/l) and nutrients (0.6 to 2.1 mg Total Nitrogen/l and 0.05 to 0.45 mg Total Phosphorus/l) than
24 those under Atlantic regime. However, the latter infiltrated higher loadings in terms of mass. Nitrogen
25 was the substance that infiltrated the most, reaching a 25% of the total mass of nitrogen deposited on

26 the pavements surface. The concentration and mass loading in infiltrated water increased as the
27 pollution build-up level did. The leachability of nutrients and organic matter was greater for high
28 rainfall intensities. The results suggest that it is essential to carry out an adequate cleaning in dry
29 conditions, especially when high intensity rainfall events are foreseen, because of its greater capacity
30 to mobilize pollutants.

31

32 **INTRODUCTION**

33 About 46% of pollution discharged to surface water bodies is associated with urban runoff, according
34 to Kamali et al. (2017). The proper management of urban runoff is therefore a key issue in the
35 maintenance of good status of surface water bodies. Sustainable urban drainage systems (SUDS) aim
36 to manage rainwater at source and they employ natural processes, thus renaturing the urban water
37 cycle. According to their definition (Woods et al. 2015), SUDS provide four key benefits to the urban
38 environment: improved water quality (1), reduced volume or speed water quantity (2), amenity (3)
39 and biodiversity (4). Water infiltration into the ground is a key aspect of their operation. However,
40 given that the accumulation of pollutants on urban surfaces is a clear hazard source, it is necessary to
41 study the potential effects of stormwater on the surrounding environment even after this enhanced
42 infiltration, especially when water passes into the groundwater.

43 Permeable pavements are one of the infrastructure features that can be included in SUDS design and
44 provide a wide range of benefits, including: reduction of surface urban runoff and peak flow
45 attenuation, reduction of combined sewer overflows, groundwater recharge (Tedoldi et al. 2016),
46 noise reduction and avoidance of aquaplaning (Stotz and Krauth, 1994) or mitigation of urban heat
47 island effect (Liu et al. 2018), among others. Therefore, they provide the first three key benefits of
48 SUDS and the fourth, indirectly, via improved quality of water released to the water environment.
49 There is an extensive list of studies on the hydraulic performance of these types of systems as well as
50 how they contribute to reduce runoff at source. However, there are few investigations into how they

51 affect the quality of the infiltrated water although the number has been growing in recent years (Drake
52 et al. 2014; Brown and Borst, 2015 and Kamali et al. 2017 among others). Several aspects that remain
53 to be studied are the influence of the rainfall regime, the rainfall intensity, and the level of pollution
54 build-up prior to rain on the hydraulic behaviour and the quality of infiltrated water. Thus, studying
55 the response of permeable pavements to high rainfall intensity events is crucial to demonstrating their
56 ability to face future challenges associated with climate change, as the Intergovernmental Panel on
57 Climate Change expects that extreme precipitation events will become more intense and frequent in
58 many regions (IPCC, 2018).

59 The type of sediments (real dust and dirt or artificially provided) used to evaluate the decrease in
60 permeability or the influence on the quality of infiltrated water varies between studies (Andrés-Valeri
61 et al. 2016; Kamali et al. 2017). When changes in water quality are evaluated, it seems more
62 appropriate to use actual dust and dirt in order to simulate more realistic conditions. Kamali et al.
63 (2017) support the idea that applying the contaminant load through surface runoff is a more realistic
64 experimental setup because pavements are frequently cleaned and the majority of pollutants are thus
65 introduced through surface runoff. In this sense, one must be aware that real-scale installations will
66 not behave in exactly the same way as experimental laboratory-scale sets. Pezzaniti et al. (2009) found
67 that a real permeable pavement became clogged at a faster rate than those evaluated under
68 laboratory conditions, whereas Razzaghmanesh and Borst (2019) did not observed surface clogging
69 issues after an eight-year period of operation in a real parking lot.

70 Studies dealing with the quality of infiltrated water show that the conditions occurring within
71 pavements, i.e. aerobic conditions and adequate pH, are suitable for the growth of nitrifying bacteria,
72 thus nitrification is the dominant process affecting the nitrogen composition in the effluent (Brown
73 and Borst 2015; Drake et al. 2014; Kamali et al. 2017; Razzaghmanesh and Borst, 2019). In addition, if
74 the outflow is regulated using underdrains or valves, it is possible to create anoxic zones that favour
75 the elimination of nitrogen through denitrification (Drake et al., 2014). In cases where denitrification

76 does not take place, infiltration of nitrates into the groundwater increases, implying a negative impact.
77 However, nitrate concentrations usually found in urban runoff are far from the established limits
78 needed to declare nitrates vulnerable zones (Brown and Borst, 2015; Andrés-Doménech et al. 2018).
79 Another important process that affects the quality of the infiltrated water is filtration: particulate
80 substances can be retained in the pavement, thus improving the quality of the filtered water, although
81 particles retained can decompose over time, slowly releasing the substances they are composed of
82 (organic matter, nutrients, metals, etc).

83 The washing of the materials that compose the different layers of the permeable pavements also
84 affects the quality of the infiltrated water. For example, Brown and Borst (2015) saw that porous
85 asphalt leached nitrogen, i.e. the total nitrogen in the effluent was higher than in runoff, while
86 permeable interlocking concrete pavement or continuous pervious concrete exported phosphates and
87 the porous asphalt reduced it.

88 The present study considers the following hypotheses: the rainfall regime and rainfall intensity can
89 significantly influence the hydraulic behaviour and the mobilization of pollutants through the
90 pavement; the degree of pollutant build-up significantly reduces the infiltration rate, having
91 consequences on the mobilization of pollutants. Knowing the influence of these factors will allow us
92 to suggest recommendations for maintenance in order to lengthen as much as possible the lifespan of
93 the permeable pavements as well as their positive impact on the receiving water bodies. Results are
94 also important for establishing several technical specifications for their design and manufacturing
95 processes. Therefore, the aim of this study is to gain knowledge about the changes in water quality of
96 runoff produced by permeable pavements and their impact in both groundwater and surface water.

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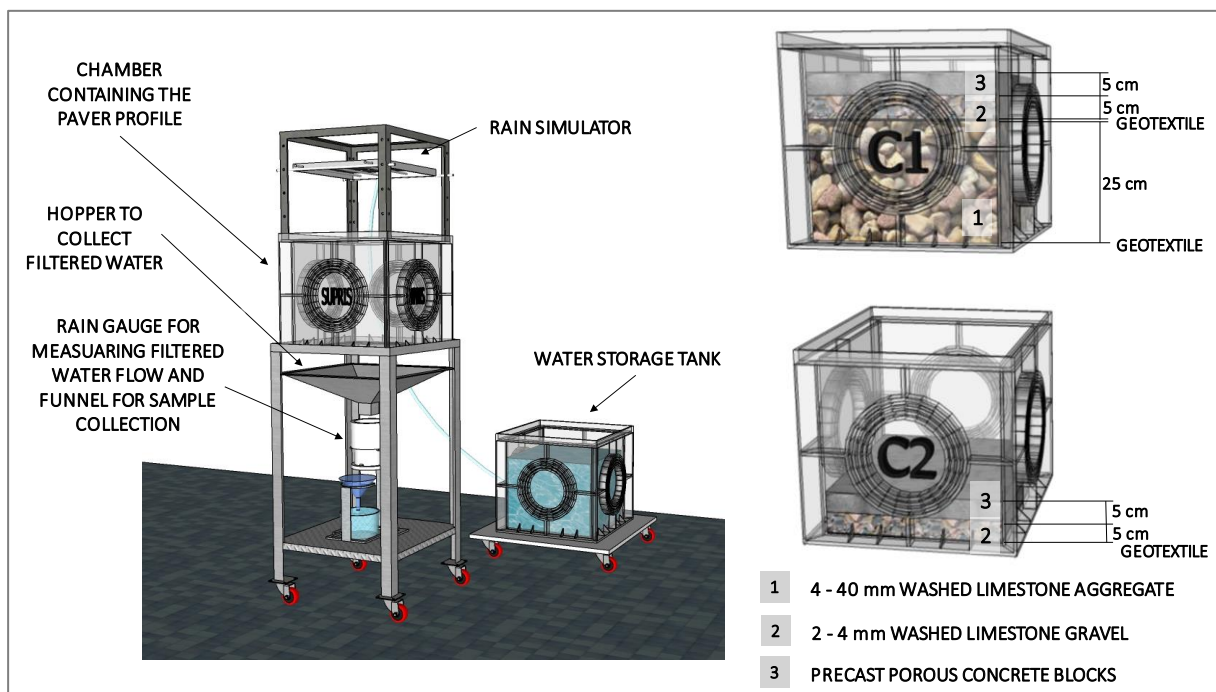
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101 **MATERIALS AND METHODS**

102 **Materials**

103 The experimental setup designed to achieve the objectives of the study consists of a four infiltrmeters
104 battery. Two configurations of pavement layers were tested: the composition of configuration C1 is
105 illustrated in Figure 1; configuration C2 has the same layers except the bottom layer of washed
106 limestone aggregate 4-40 mm size. According to the information provided by the manufacturer, the
107 components used in the concrete blocks production are the following: siliceous arid, white and grey
108 cement, plasticizer and water-repellent additives and pigments (iron oxides). The rainfall simulator is
109 based on Rodríguez-Hernández et al. (2016) and is composed of a water storage tank, a pump (Multi-
110 1300 SICCE) and a grid of drip irrigation pipes consisting of 7 rows with 13 drippers per row evenly
111 distributed and placed 50 cm above the pavement surface (see Figure 1). A rain gauge is positioned
112 below each permeable pavement to measure the infiltrated water flow, which is then collected and
113 kept in a refrigerator until chemical analysis is performed.



114

115 Figure 1. Scheme of the experimental setup.

116 Deionized water was used (Electrical Conductivity: 21 $\mu\text{S}/\text{cm}$, pH: 6.9) to simulate rainwater. The
117 material used to simulate the different degrees of pollution build-up was collected by the road cleaning
118 services of the university, thus being real dust and dirt deposited on urban roads. It was collected by a
119 mechanical sweeper in dry conditions.

120 **Methods**

121 Two rainfall regimes, Atlantic and Mediterranean, were tested. The simulated rainfall reproduces the
122 average rainfall regime of Valencia (Mediterranean) and Santander (Atlantic), both in Spain: events
123 with a rainfall volume of 16 mm and 30 minutes duration (0.5 mm/min), with a weekly frequency for
124 the Atlantic climate and biweekly in the Mediterranean climate (Andrés-Doménech et al., 2010). After
125 each rainfall event, the infiltrated water was analysed. Each rainfall regime was tested in two
126 infiltrometers (configurations C1 and C2). The experiments were planned to be long-term tests,
127 starting in January 2017. After the first year, the infiltrometers with configuration C1 continued under
128 the same conditions in order to have a longer-term experiment and those with configuration C2 were
129 cleaned with a domestic vacuum cleaner and a brush and were then used to test the performance of
130 the permeable pavements under more intense rainfall events, in particular 22 mm in 10 min (2.2
131 mm/min), which has a return period of once in 25 years in Valencia (Spain). For this test, a plastic mesh
132 was installed according to Naves et al. (2017) to break rainfall drops. If surface runoff was generated
133 it was collected and analysed.

134 The influence of progressive pollution build-up was studied by dry sprinkling sediments (real dust and
135 dirt) on the pavement surface. The applied deposition rate (5 $\text{g}/\text{m}^2/\text{d}$) was selected according to the
136 information provided by the cleaning services (mass collected, surface swept and cleaning frequency)
137 and checked against scientific references (Table 1). The deposition rate used is the same for all the
138 tests. The deposition rate was kept constant throughout the study period in order to evaluate the
139 response of permeable pavements to a surface overload of pollutants.

140

141 Table 1. Deposition rates or build-up levels after different numbers of dry days measured by other authors.

Deposition rate or Build-up level	Collection method	Reference
15 – 18 g/m ² after 3 dry days*	Vacuuming: free load.	Vaze and Chiew, 2002
15 – 20 g/m ² after 2 dry days*	Fibre brush + vacuuming: fixed load	*Total load (fixed + free)
0.1133 - 0.6525 g/m ² /d	Drain runoff with a corrugated pipe	Kim et al., 2006
2 – 2.43 g/m ² for 1 st dry day 2.85 – 5.29 g/m ² for 21 st dry day	Vacuuming (water filtration system + small circular foot with a brush)	Egodawatta and Goonetilleke, 2006
0.016 – 0.058 g/m ² /d	Capture runoff with slot drains, which drained to weir boxes	Winston et al., 2012
Particles < 0.150 mm: 1.50 – 2.45 g/m ² for 1 st dry day 1.10 – 1.50 g/m ² for 21 st dry day	Vacuuming (water filtration system + small circular foot with a brush)	Wijesiri et al., 2015
Particles > 0.150 mm: 0.25 – 0.30 g/m ² for 1 st dry day 1.10 – 3.50 g/m ² for 21 st dry day		
g/m ² after 1 st dry day:	Vacuuming with a domestic vacuum cleaner (Philips FC8264), with an air filtration system and cyclonic dustbin that effectively captures microscopic particulates.	Zhao et al., 2016
0.13 – 0.45 < 44 μm		
1.25 – 1.55 44 - 62 μm		
1.75 – 2.38 62 - 105 μm		
0.45 – 0.50 105 - 149 μm		
1.00 – 1.25 149 - 250 μm		
1.25 – 2.25 250 - 450 μm		
0.95 – 2.13 450 - 1000 μm		

Note: Maximum values usually coincide with highly populated areas. Data from literature extracted from graphs, so there is a possibility of some slight imprecision from graph reading. Free load is collected only by vacuuming and fixed load is collected by brushing plus vacuuming after the collection of the free load.

142
143 In a complementary and independently way to the infiltrometer experiments, physicochemical
144 analysis of an artificial mixture of dust in deionized water was made. The objective was to know the
145 chemical composition of the potential runoff from an impervious urban area assuming that the entire
146 volume of rainfall became runoff, and that almost all the mass of pollutant deposited on the surface
147 was washed-off. This mixture was made with the same proportions of sediments and deionized water
148 as in the infiltrometer tests (2.2 g/l for 1-week build-up level) and the same variables of water quality
149 were analysed.

150 Physicochemical analyses of sedimentary material were performed according to the following
151 standardized methods: particle size distribution (ISO 77314); organic matter as loss on ignition (LOI)
152 (550°C for 1 h) and as organic carbon (Walkley-Black method as described in Schumacher, 2002);
153 moisture (105°C for 24 h); total nitrogen (ISO 11261), total phosphorus (EN 14672) and electrical
154 conductivity (EC_{1:5}) and pH of sediment suspension in water in proportion 1:5 (V:V) (ISO 11265). For

155 metal analyses, material was milled and digested in a microwave with HNO₃ and H₂O₂ at 200°C for 15
156 minutes except for Hg, which was measured in a mercury analyser (DMA-80).

157 The total volume of infiltrated water was collected and kept in the refrigerator until analysis within 24
158 hours. The water samples were analysed for the following parameters: Total Nitrogen (TN)
159 (Spectroquant® test: ISO 11905-1 + photometry), Ammonium (Spectroquant® test: ISO 7150/1),
160 Nitrites (Spectroquant® test: EPA354.1), Nitrates (Spectroquant® test), phosphate (test
161 Spectroquant®: ISO 6878/1) and total phosphorus (test Spectroquant®: digestion + ISO 6878/1),
162 Biological Oxygen Demand - BOD (respirometry test through OxiTop® control system), Chemical
163 Oxygen Demand - COD (test Spectroquant®: ISO 15705), Total and Volatile Suspended Solids (UNE-EN
164 872 y UNE 77034 respectively) and turbidity with a TN100 Eutech turbidity-meter. WTW-Multi 340i
165 probes were used to measure: temperature, dissolved oxygen (Cellox® 325), conductivity (TetraCon®)
166 and pH (SenTix® 41).

167 Statistical analyses were performed using SPSS 16.0 software (SPSS® software). The influence of the
168 pavement configuration and the rainfall regime was analysed by comparing the temporary data series.
169 The statistical tests used were parametric if normality was satisfied (T-Student for related variables)
170 and nonparametric otherwise (Friedman's test for several related variables and Wilcoxon's test for
171 pairwise comparisons). Statistical significance was indicated by a probability of type I error of 5% or
172 less ($p \leq 0.05$).

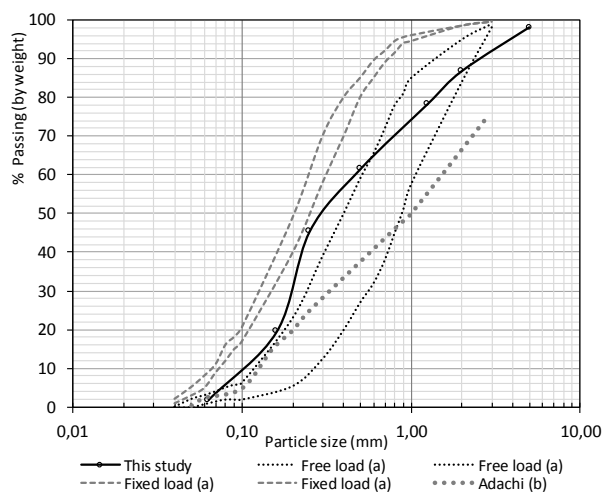
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174 **RESULTS AND DISCUSSION**

175 **Characteristics of the sediments used in the experiments**

176 The sediments used to simulate a progressive pollutant build-up over the pavement surface were
177 actual dust and dirt taken from the University roadways, as stated in the previous section. The particle
178 size distribution presents a high proportion of sands and is quite similar to the material used by

179 Sañudo-Fontaneda et al. (2014). It is also comparable to other road deposited sediments, being located
 180 between the free and fixed fraction described by other reference studies (Vaze and Chiew, 2002), as
 181 shown in **Figure 2**. The particle size distribution described by Vaze and Chiew for free load (collected
 182 only by vacuuming) and fixed load (collected by brushing plus vacuuming after the collection of the
 183 free load) encompasses the data reported in many other studies (Zafra et al. 2008; Andrés-Valeri et al.
 184 2016). The thicker particles obtained in the present study deviate a little from data on free material
 185 collected by Vaze and Chiew (2002), but if we project the particle distribution size described by Adachi
 186 and Tainosho (2005) upwards it can be supposed that these larger particles fall within this gradation.



187
 188 **Figure 2.** Particle size distribution in this study and ranges described by (a) Vaze and Chiew (2002) for fixed and
 189 free loads and (b) coarser distribution found by Adachi and Tainosho (2005). Data from literature is extracted
 190 from graphs, so there is a possibility of some slight imprecision from graph reading.

191
 192 As reported in **Table 2**, the sediments collected in this study have a moderate content of organic matter
 193 (8.23 % dry weigh-dw) compared to natural and contaminated soils or sediments (Hernandez-Crespo
 194 et al. 2012; 2017) and low moisture content (2.02 % dw). The results obtained are within the range of
 195 variation of other studies analysing street dust and dirt (**see Tables 2 and 3**). The nutrient concentration
 196 can also be classified as moderate compared to natural soils or sediments (Hernández-Crespo et al.
 197 2017), so the results indicate that the urban runoff could have a potentially negative impact on the
 198 receiving waters. The electrical conductivity of the aqueous extract is 890 $\mu\text{S}/\text{cm}$ indicating that the

199 dust and dirt material is saline according to the general classification of soils (Shrivastava and Kumar,
 200 2015) and it has a moderately basic pH (8.0). In contrast to the organic and nutrient content, the
 201 concentration of some metals (Cr, Pb and Zn) should be considered very high as they far exceed the
 202 background concentrations of sediments from the nearest watershed, i.e. the concentrations that can
 203 be found naturally in soil and sediments (Hernández-Crespo and Martin, 2015).

204 Table 2. Physical-chemical characteristics of the dust and dirt used in the tests. Average and standard deviation
 205 of replicas are shown; n is the number of replica. Results are expressed on a dry weight (dw) basis.

Physico-chemical variable	Average ± Stnd.Dev. (n)	Metal	Average ± Stnd.Dev. (n)
Moisture (% dw)	2.02 ±0.00 (2)	Cd (mg/kg)	0.32 ±0.20 (4)
LOI (% dw)	8.23 ±0.15 (2)	Cr (mg/kg)	306 ±159 (4)
Organic carbon (% dw)	1.83 ±0.11 (4)	Cu (mg/kg)	36 ±11 (4)
COD (mg/g)	48.9 ±3.1	Pb (mg/kg)	132 ±45 (4)
Total nitrogen (mg/kg dw)	1351 ±197 (3)	Mn (mg/kg)	230.0 ±48.5 (4)
Total phosphorus (mg/kg dw)	302.5 ±18.5 (2)	Ni (mg/kg)	38.2 ±15.0 (4)
Electrical conductivity (EC _{1:5}) (µS/cm)	890 ±103 (3)	Zn (mg/kg)	238 ±43 (4)
pH _{1:5}	8.0 ±0.1 (3)	As (mg/kg)	4.79 ±0.70 (3)
Particle size distribution (% dw):		Hg (mg/kg)	0.0149 ±0.0024 (4)
Gravel (> 2mm)	13.2 ±0.6 (2)	Al (mg/kg)	3450 ±140 (3)
Sand (0.063-2 mm)	85.0 ±0.5 (2)	Fe (mg/kg)	8000 ±1000 (3)
Silt and clay (<0.063 mm)	1.8 ± 0.0 (2)		

206
 207
 208

Table 3. Review of dust and dirt quality in different cities around the world.

OM (%)	COD (mg/g)	pH	TP (mg/kg)	TN (mg/kg)	Comments on methods:
2.8-18.8 ^g	107.7-253.7 ^b	6.8-12.0 ^g	529.4-1940.3 ^b	3190.5-6268.7 ^b	^a Fraction <2mm; sweeping.
2.5-13.6 ^d		7.0-8.0 ^d	346-933 ^d		^b Fraction <2mm; sweeping.
			349.7-381.2 ^f	429.6-832.4 ^f	^g Fraction <2mm; vacuum cleaner.
Cd (mg/kg)	Cu (mg/kg)	Ni (mg/kg)	Pb (mg/kg)	Zn (mg/kg)	
3.77 ±2.25 ^a	173 ±190 ^a		181 ±92.9 ^a	1450 ±869 ^a	^c Fraction <1mm; sweeping.
0.0-13.1 ^c	16.4-6688.4 ^c	0.0-636.2 ^c	0.0-199.4 ^c	81.3-164.8 ^c	^h Fraction <0.147mm; sweeping.
1.17-3.77 ^e	94.98-196.8 ^e	23-86.26 ^e	53.33-408.41 ^e	294.47-1450 ^e	ⁱ Fraction <0.050mm; sweeping.
1.3-3.4 ^g	89.8-181.8 ^g	13.4-33.0 ^g	82.0-153.1 ^g	1290-324.7 ^g	^j Sweeping.
4.00-139 ^h	102-966 ^h	15.0-69.3 ^h	157-1482 ^h	813-45675 ^h	^d Fraction <2mm; vacuum sweeper.
0-2.4 ⁱ	19-1394 ⁱ	10-2058 ⁱ	12-2388 ⁱ	70-9223 ⁱ	
0.4-3.1 ^j		12.7-151.3 ^j	20.1-96.2 ^j		
0.4-1.5 ^d	67-151 ^d	15-28 ^d	96-222 ^d	249-547 ^d	
Al (mg/kg)	As (mg/kg)	Hg (mg/kg)	Cr (mg/kg)	Fe (mg/kg)	Mn (mg/kg)
4600-11200 ^h	10.9-110.1 ^h	0.18-10.8 ^h	29.0-214 ^h	14700-65600 ^h	604-4761 ^h
	7.1-16.3 ^j	0.005-1.358 ^j	51.29-167.28 ^e	4883-54576 ⁱ	149-999 ⁱ
			19-3134 ⁱ		
			33-70 ^d		

References: ^aLi et al. (2001); ^bBurton and Pitt (2002); ^cCharlesworth et al. (2003); ^dDurand et al. (2003); ^eWei and Yang (2010); ^fBerretta et al. (2011); ^gDuong and Lee (2011); ^hOrdóñez et al. (2015); ⁱMarín Sanleandro et al. (2018); ^jWu and Lu (2018).

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The literature shows a very wide range of variation in terms of nutrients and heavy metals content in street dust (Table 3). The maximum values of metals corresponds to areas near metallurgical industries

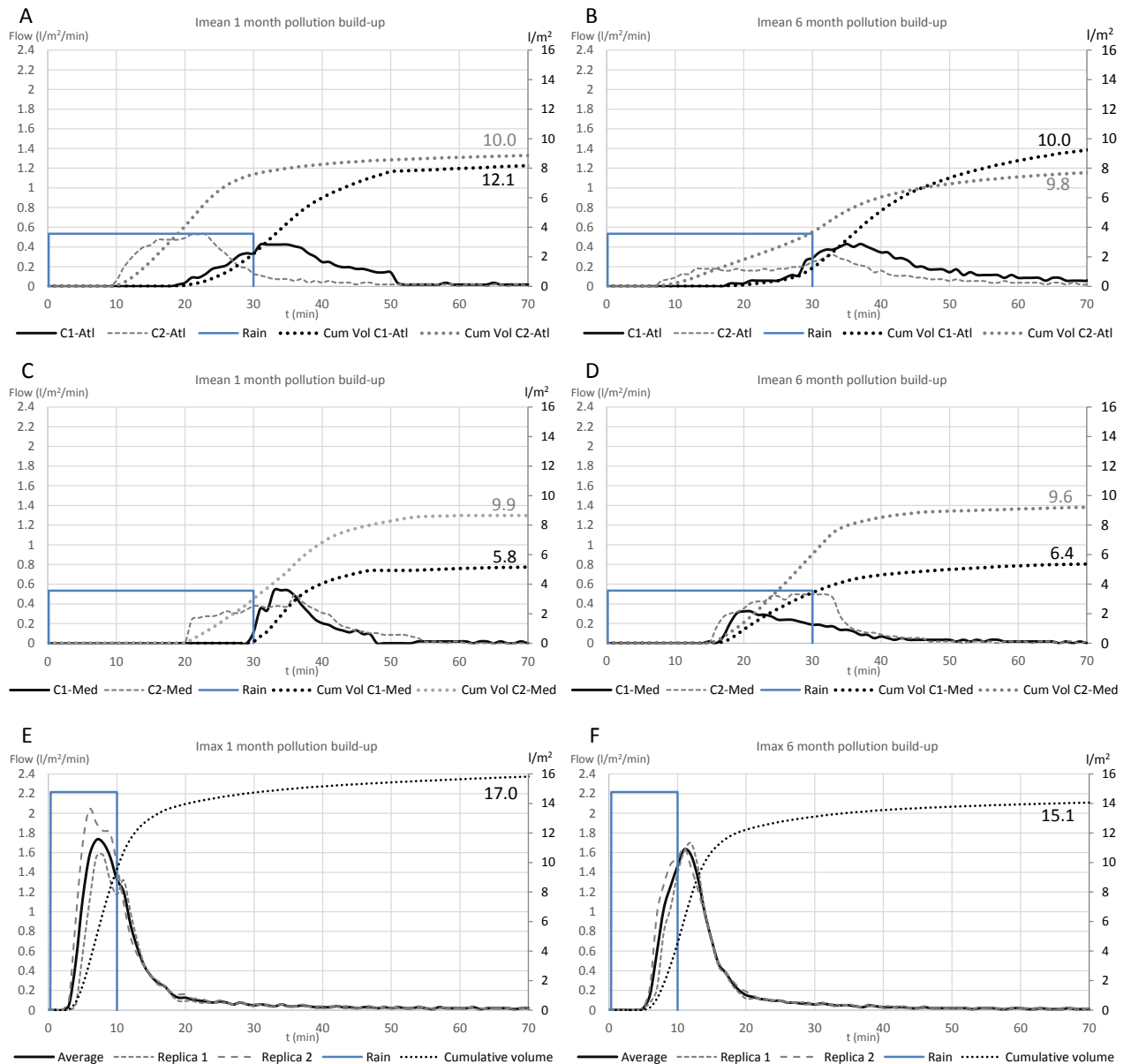
212 or heavy industrial cities (Ordóñez et al. 2015; Wei and Yang, 2010). Charlesworth et al. (2003)
213 identified a spatial pattern in the variability of metals concentrations: the more traffic with many stops
214 and starts and the higher the population, the higher concentrations. Marín Sanleandro et al. (2018)
215 also identified traffic as a classification variable; they found a high accumulation of Pb and Zn in ledges
216 of narrow city centre streets with moderate traffic; they also stated that the colour of samples can be
217 considered as a proxy for rapid diagnosis, as dark samples were the most contaminated. The values
218 obtained in the present study were in the lower limit of their range of variation, maybe because all
219 parts of the sediment were analysed together, rather than analysing only the finest fraction. This was
220 done because the entirety of the sediment was exposed to the rainfall, so we were interested to know
221 the contaminant concentration of the whole sediment sample. Another potential reason could be that
222 the sampled area is inside the university campus and the main contaminant activity is road traffic, but
223 characterized by relatively few stops and starts.

224

225 **Influence of pollution build-up level and rainfall intensity on infiltrated water quantity**

226 The hydrographs of infiltrated water obtained in the different tests performed are shown in Figure 3.
227 The maximum flow rates reached during high intensity rainfall events (around 1.7 mm/min, Fig. 3 E
228 and F), were notably higher than those simulated at low intensity (between 0.3 and 0.5 mm/min, Fig.
229 3 A to D). The duration of the peaks were lower, in accordance with the lower duration of the simulated
230 storms in the high intensity scenarios. Related to this, peak flows reached in the high intensity tests
231 were clearly below the applied rainfall intensity, whereas they were closer to the rainfall intensity in
232 the low intensity tests, due to their longer duration.

233



234
 235 Figure 3. Hydrographs of infiltrated water of mean intensity (16 mm/30 min) under different rainfall regimes:
 236 Atlantic (A, B) and Mediterranean (C, D) and different pollution build-up levels: one month (A, C) and six month
 237 (B, D). C1: complete configuration; C2: simple configuration. Hydrographs of high intensity events (22 mm/10
 238 min) under different pollution build-up levels: one month (140 g/m²) (E) and six month (840 g/m²) (F). Cumulative
 239 volume curves are displayed on the secondary axis and numerical data indicate the total drained volume. .
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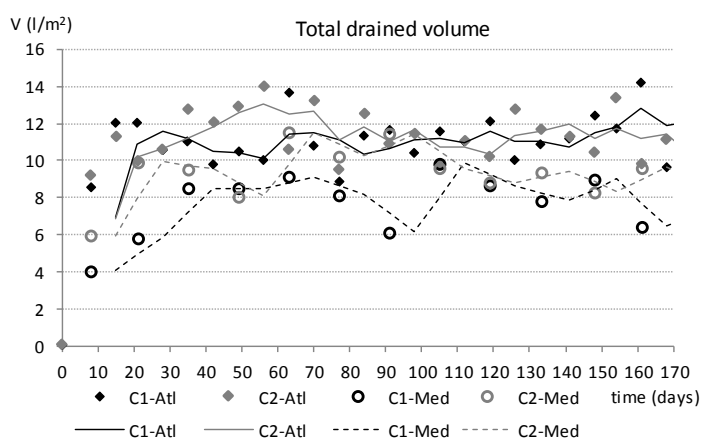
241 Regarding the influence of the pollution build-up level, it can be clearly observed in the intense rainfall
 242 simulations that the flow rate reached in the events simulated with a pollution build-up level of 6
 243 months (Fig. 3 F) was lower and was reached later than in the simulation of 1 month of build-up level
 244 (Fig. 3 E). The larger quantity of dust and dirt accumulated on the pavement surface absorbed part of
 245 the rain, thus smoothing the peak flow of filtered water. Moreover, this might also have allowed for a

246 better distribution of rainwater on the pavement surface, thus improving the distribution of flow
247 through the entire pavement pore structure. A slight decrease in the infiltration rate was also observed
248 in pavements subjected to low rainfall intensity regimes (Fig. 3 A and C versus B and D), which could
249 be due to the absorption of water by the accumulated dust and to progressive pavement clogging.

250 For simulations performed at low rainfall intensity, reasonable differences were observed between
251 the rainfall regimes when the build-up level reached one month (Fig. 3 A and C). The flow of filtered
252 water under the Atlantic regime started earlier than it did under the Mediterranean regime, because
253 the pavements under Mediterranean regime were drier due to the lower frequency of rainfall. There
254 was also an observable difference between the pavement configurations: water began to drain earlier
255 in pavements of simple configuration (C2), showing a lower capacity for water retention, and related
256 to the shorter path of this configuration. However, for the rainfall events simulated when the build-up
257 level reached six months (Fig. 3 B and D) the difference between drained water flows was not as clear.
258 Water began to filter more quickly in the pavements under Mediterranean climate and the pavements
259 with simple configuration started to drain earlier, but it did not look as clear as in the 1-month build-
260 up scenarios. This finding could be related to the capacity of the accumulated dirt to attenuate the
261 flow, as it absorbs part of water, and also associated with the increasing clogging.

262 Differences in the total drained volume can be seen in Figure 3 through the cumulative volume curves.
263 The permeable pavements were always able to retain a significant part of the total rainfall volume
264 applied (16 and 22 l/m² for the low and high rainfall intensities respectively), at least 16%. When rainfall
265 events are frequent (Atlantic climate, Fig. 3 A and B), the complete configuration (C1) does not
266 represent a great advantage in terms of water retention. In this case, the pavement with complete
267 configuration retained a lower volume fraction (16 and 24% for one and six-month build-up levels
268 respectively) than those with simple configuration (37 and 39% respectively). In contrast, in the
269 Mediterranean climate tests, the benefit of having a thicker pavement layer is very evident, since the
270 retained volume is much higher (62% of total rainfall volume applied on average) than when the

271 pavement configuration is simple (39% on average) (Fig. 3 C and D). In the case of intense rainfall
 272 events, the volume retained varied between 23 and 31% for 1 and 6-month build-up levels
 273 respectively. In relation to the retained volume, and applicable to all configurations, regulating the
 274 permeable pavements outflow, by means of discharge underdrains or valves, would allow for a greater
 275 retention of volume inside them. Temporary storage of water would favour infiltration and
 276 evaporation processes and so increasing the hydraulic and environmental efficiency of the
 277 infrastructure. Figure 4 shows the complete data series of the total drained volume in each event for
 278 the average rainfall regime tests. For infiltrimeters with C1 configuration, the total volume drained in
 279 the infiltrimeters subjected to Atlantic regime is significantly higher than that from the infiltrimeters
 280 under Mediterranean regime ($p < 0.05$). On average, there was not a significant difference between C1
 281 and C2 in the Atlantic regime ($p > 0.05$), but for the Mediterranean regime ($p < 0.05$), which is in line with
 282 the aforementioned.



283 Figure 4. Temporal evolution of total drained volume in the average rainfall regimes (C1: configuration type 1;
 284 C2: configuration type 2; Atl: Atlantic rainfall regime; Med: Mediterranean rainfall regime). Dots represent the
 285 data and lines the moving average with a period of three data.
 286
 287

288 Overall, a very important result is that, after six months of pollution build-up, permeable pavements
 289 still maintain a high permeability, being able to infiltrate the total volume of water applied at low
 290 rainfall intensity and almost the entire volume in the high rainfall intensity tests. Surface runoff was
 291 only generated for the simulation made at high intensity of rain and six-month build-up level, the
 292 volume was 167 ml (3% of the total applied volume).

293

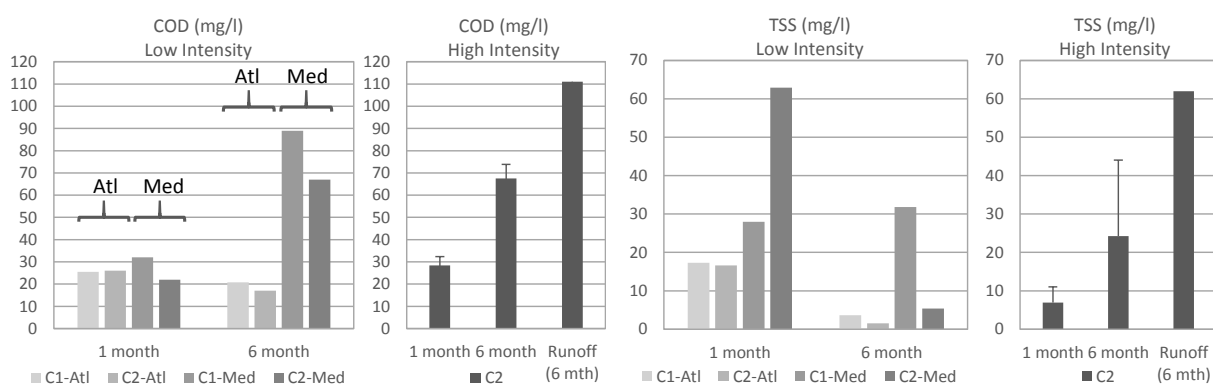
294 **Water quality results**

295 This section presents the water quality results for the permeable pavements, complete and simple
296 configurations, under both rainfall regimes (Atlantic and Mediterranean) and high rainfall intensity.
297 The water infiltrated through the permeable pavements initially presented very high values of
298 electrical conductivity and pH, which decreased as the material received more and more rainfall, from
299 values of about 3000 to 1000 $\mu\text{S}/\text{cm}$ and from 12 to 10 for pH after six months. These high values of
300 pH could negatively affect soil properties and microbiology, so it is advisable to analyse the
301 manufacturing process of the pavement to see how such an increase in pH could be avoided. Several
302 authors have also found increases in pH related to pavement components, particularly for pervious
303 concrete and porous asphalt (Drake et al. 2014; Brown and Borst, 2015). In relation to a potential use
304 for irrigation, guideline recommendations typically suggest pH values between 6.5 and 8.5. Although
305 higher pH levels may not be harmful for plants, there might be a potential risk of fouling in irrigation
306 systems especially if water also has a high alkalinity (Kazemi and Hill, 2015). In relation to these high
307 values of pH it is worth noting that this increase can be considered as a positive point in locations that
308 receive substantial amounts of acidic rainfall (Kazemi and Hill, 2015), because it contributes to
309 neutralize the acidic rainfall.

310 The influence of pollution build-up levels and rainfall intensity on infiltrated water quality can be
311 examined in Figures 5 to 8. For COD concentrations in infiltrated water, the rainfall regime and build-
312 up level seemed to be of greater influence than rainfall intensity (Fig. 5). In the rainfall events simulated
313 for one month of pollution build-up, the results were quite similar for both configurations and rainfall
314 regimes, varying between 22 and 32 mg/l. When build-up corresponded to six months, the COD
315 concentrations reached under Mediterranean climate were much higher, between 67 and 89 mg/l,
316 than those for Atlantic climate (about 20 mg/l), and so is clearly associated with the lower rainfall
317 frequency. For high rainfall intensity events, the COD is also noticeably higher for the six-month build-

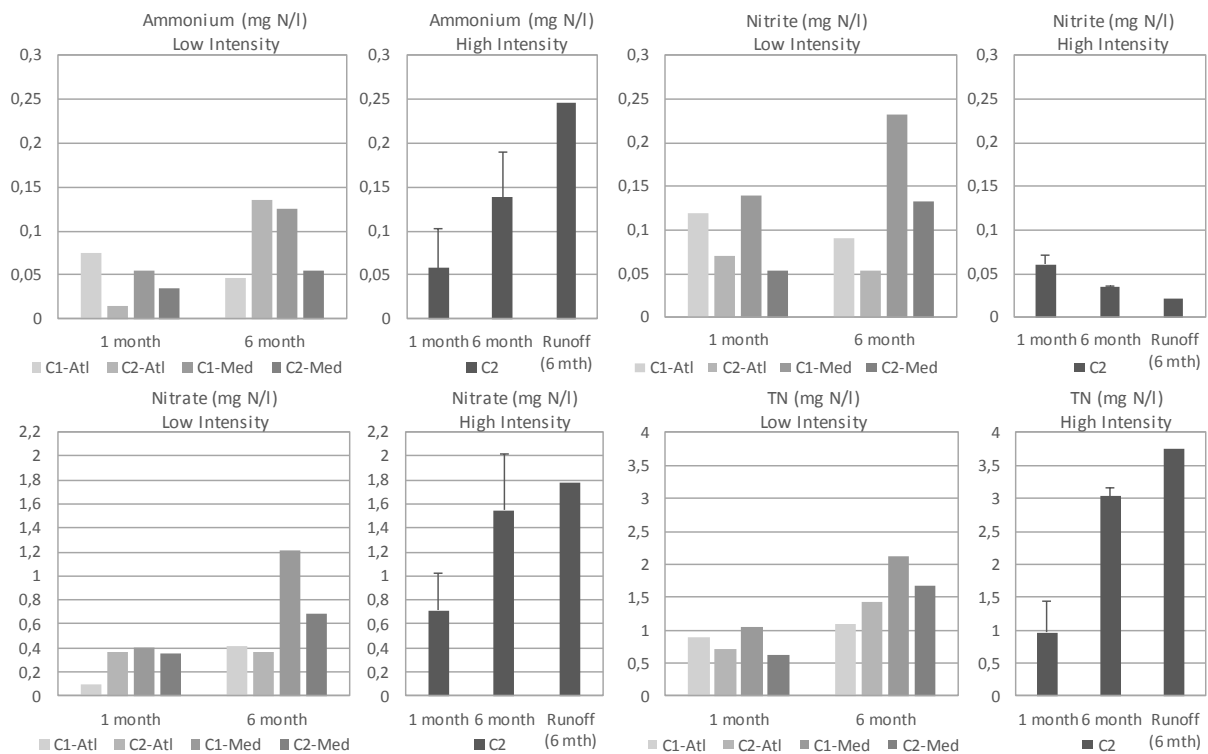
318 up level (68 mg/l) than for one month (28 mg/l). It is noteworthy that the concentration reached by
 319 the surface runoff was much higher, reaching a value of 111 mg/l. In this case, the volume of surface
 320 runoff generated was very small (168 ml) due to the zero slopes of the pavement and its high
 321 permeability, but in an impervious pavement it would have been considerably greater. Surface runoff
 322 was only generated in the test performed at high rainfall intensity and 6 month of build-up level, in
 323 which the filtration rate was slowed and the accumulation of water on the surface led to the generation
 324 of surface runoff. The results of BOD₅ test indicated that the organic matter present in the infiltrated
 325 water was slowly biodegradable, with a BOD₅/COD relationship of 0.2 on average, value similar to that
 326 found by Andrés-Doménech et al. (2018).

327 Regarding TSS, it should be noted that a first washing of the gravel that composes the pavement layers
 328 occurred, even though it was washed before the experimental set up. For this reason, the
 329 concentrations measured when the pollution build-up level reached one month were higher (17 to 63
 330 mg/l) than those for six month of dust accumulation (5 to 32 mg/l). Having a low concentration of
 331 suspended solids in infiltrated water is very important if the permeable pavement can be used as a
 332 rainwater harvesting tank for irrigation purposes, since high TSS concentrations could obstruct
 333 irrigation systems.



334 Figure 5. Chemical oxygen demand (COD) and total suspended solids (TSS) in the different assays performed (C1:
 335 configuration type 1; C2: configuration type 2; Atl: Atlantic rainfall regime; Med: Mediterranean rainfall regime;
 336 Low intensity: 16 mm/30 min; High intensity: 22 mm/10 min). All values refer to infiltrated water except when
 337 the x-axis indicates runoff, which refers to surface runoff.
 338

339 Regarding the infiltrated nitrogen, the behaviour depends on its form (Figure 6). In general, the
340 infiltrated water in the six-month build-up level events presented a higher concentration, especially in
341 the pavements under Mediterranean climate. Rainfall intensity did not influence in a very remarkable
342 way the concentrations reached under one-month build-up levels, but did at the six-month build up
343 levels, where greater nitrate and total nitrogen concentrations were recorded. Nevertheless, the
344 measured concentrations, with maximum of 1.55 mg NO₃-N/l, were well below the value established
345 to declare vulnerability to nitrate contamination (11.3 mg NO₃-N/l according to the European Directive
346 91/676/EEC). The risk of contamination by nitrates was also categorized as low or moderate by Burton
347 and Pitt (2002). Other authors have also found nitrates as the main nitrogen form in water drained
348 from permeable pavements, and values similar to those obtained in this study (Drake et al. 2014;
349 Brown and Borst, 2015). Drake et al. (2014) showed a reduction of nitrate concentrations in one of the
350 permeable systems which was temporarily saturated after moderate and large rainfalls, due to a low
351 permeability of the native soil in combination with a ball valve at the pavement outlet. Therefore, if
352 anoxic zones are created it is possible to significantly reduce nitrate concentrations. Prior to the
353 creation of these zones it would be recommendable to evaluate the concentration of sulphate in the
354 infiltrated water to avoid the generation of bad odours produced by sulphide production. The
355 concentration of ammonium in the water potentially infiltrated was below 0.16 mg N/l, which is the
356 value established to classify water bodies with high status in the Spanish legislation (RD 817/2015) that
357 develops the provisions of the Water Framework Directive (DIRECTIVE 2000/60/EC). In contrast, the
358 surface runoff generated in the high intensity rainfall simulation did exceed that value.

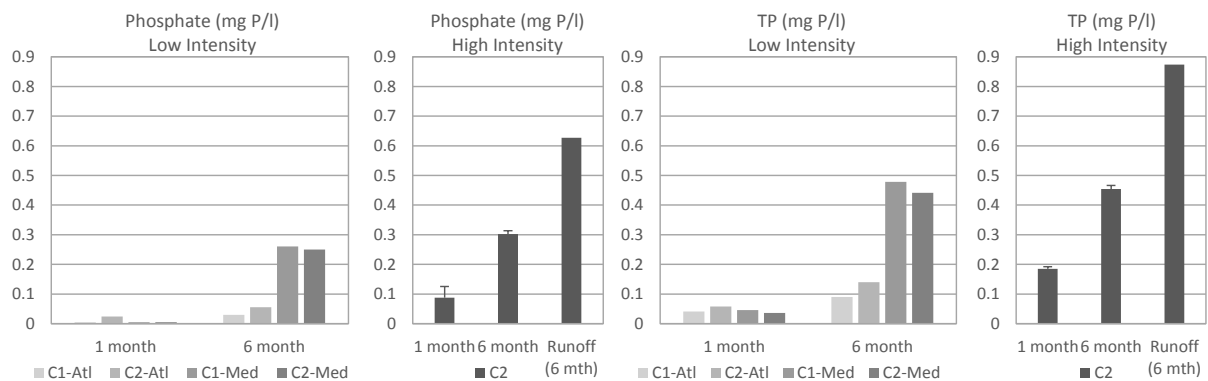


359 Figure 6. Nitrogen forms in the different assays performed (C1: configuration type 1; C2: configuration type 2;
 360 Atl: Atlantic rainfall regime; Med: Mediterranean rainfall regime; Low intensity: 16 mm/30 min; High intensity:
 361 22 mm/10 min). All values refer to infiltrated water except when the x-axis indicates runoff, which refers to
 362 surface runoff.
 363

364 The phosphorus concentrations in the infiltrated water increased with increasing build-up levels,
 365 ranging from 0.036 to 0.478 mg/l of total phosphorus (Figure 7). The effect of rainfall intensity is more
 366 noticeable for the tests performed under one-month build-up levels, i.e. for this degree of
 367 accumulated dust, the phosphorus concentration was clearly higher in the high intensity simulation
 368 (0.045 mg TP/l on average for the low intensity tests versus 0.19 mg TP/l for the high intensity tests).
 369 For the six-month build-up level, the rainfall regime also influenced the concentration of phosphorus
 370 in infiltrated water, since considerably higher concentrations were reached in tests under the
 371 Mediterranean regime and high intensity (about 0.45 mg TP/l), compared to those of the Atlantic
 372 regime. The surface runoff produced in the high intensity tests presented a concentration much higher
 373 (0.87 mg TP/l) than that of the infiltrated water (0.46 mg TP/l). This result highlights the environmental
 374 benefit of infrastructures like permeable pavements, which help to reduce the surface runoff thus
 375 protecting the receiving waters. This reduction of phosphorus loads into the receiving waters is
 376 especially important in the case of lentic fresh water bodies, since phosphorus is normally the nutrient

377 limiting the eutrophication process. The value established to classify water bodies in good status is
 378 0.16 mg P/l (RD 817/2015). This value was exceeded in simulations performed at 6-month build-up
 379 level for Mediterranean rainfall regime and, in the case of the high intensity test, both in the infiltrated
 380 water and in the surface runoff.

381



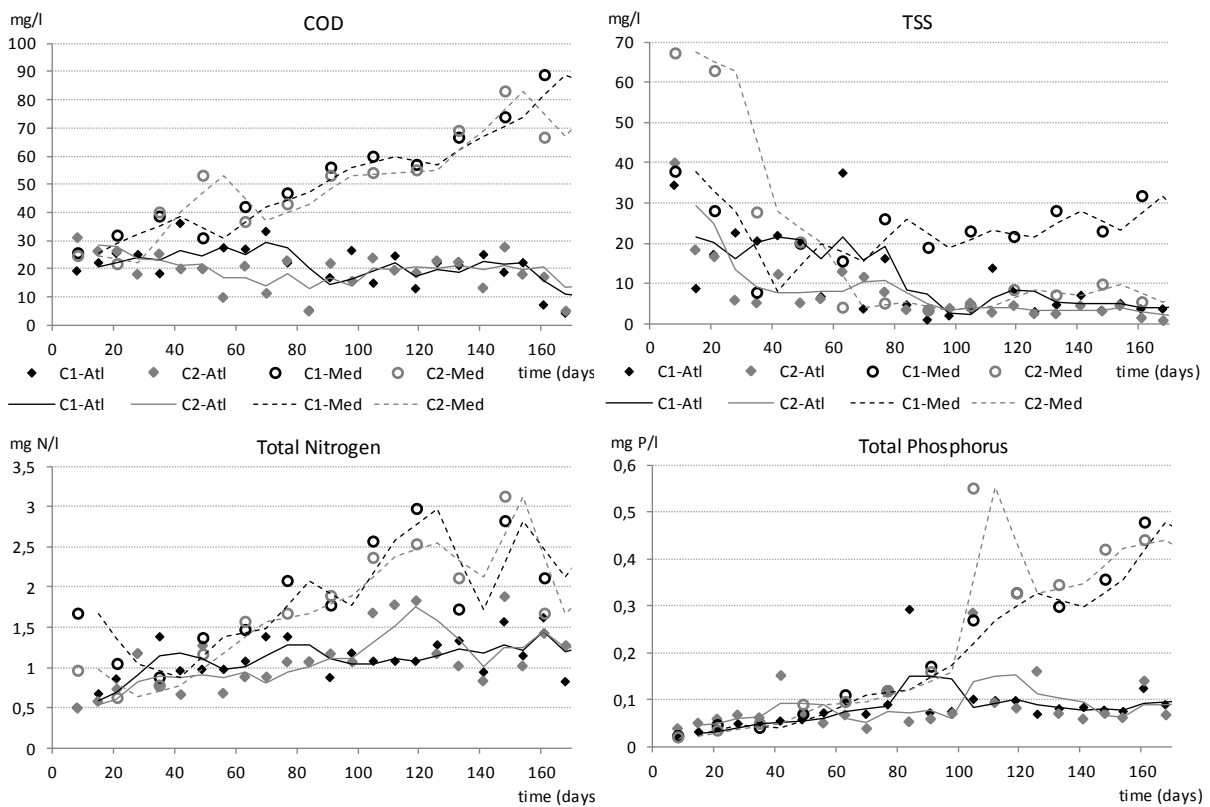
382

383 Figure 7. Phosphorus forms in the different assays performed (C1: configuration type 1; C2: configuration type
 384 2; Atl: Atlantic rainfall regime; Med: Mediterranean rainfall regime; Low intensity: 16 mm/30 min; High intensity:
 385 22 mm/10 min). All values refer to infiltrated water except when the x-axis indicates runoff, which refers to
 386 surface runoff.

387

388 The complete data series of water quality variables obtained in the average rainfall regime tests are
 389 shown in Figure 8. COD, TN and TP increased over time, especially in infiltrated water from the
 390 pavements subjected to Mediterranean rainfall regime, where the concentrations were significantly
 391 higher than those from Atlantic regime ($p < 0.05$). This increasing trend is considered to be associated
 392 with the increasing pollution build-up level on the pavements surface. The reason for the greater
 393 increase in the infiltrated water under Mediterranean regime is thought to be related to the lower
 394 rainfall frequency, so that the mobilized pollutants are concentrated in a smaller volume of infiltrated
 395 water. Conversely, TSS presented a decreasing pattern over time mainly related to the washing of the
 396 gravel that composes the pavement layers, as previously commented. For this variable, as in the
 397 previous ones, the concentration reached in the infiltrated water in pavements under Atlantic regime
 398 was significantly lower than that under Mediterranean regime ($p < 0.05$).

399 In addition, two surface runoffs potentially generated were chemically characterised: one for the
 400 Atlantic rainfall regime with a dry antecedent period of one week, and another for the Mediterranean
 401 regime with two weeks of antecedent dry period. The results respectively obtained were the following:
 402 139.8 and 344.5 mg COD/l, 154.4 and 380.3 mg TSS/l, 0.038 and 0.088 mg NH₄⁺-N/l, 0.021 and 0.027
 403 mg NO₂⁻-N/l, 0.69 and 0.80 mg NO₃⁻-N/l, 1.72 and 2.78 mg TN/l, 0.099 and 0.213 mg PO₄³⁻-P/l, 0.969
 404 and 1.720 mg TP/l. On average, these values were significantly higher than those reached in the
 405 infiltrated water (p<0.05) (Figure 8) when the complete data series is considered, even including those
 406 obtained after 6 month of pollution build-up. These results reveal the great benefit of this kind of SUDS.



407
 408 Figure 8. Temporal evolution of water quality variables in infiltrated water for the average rainfall regimes (C1:
 409 configuration type 1; C2: configuration type 2; Atl: Atlantic rainfall regime; Med: Mediterranean rainfall regime).
 410 Dots represent the data and lines the moving average with a period of three data.
 411

412

413 So far, the results obtained have been analysed for the concentration of different variables, but it is
 414 also important to analyse the results in terms of released mass load. Table 4 shows the percentage of

415 mass leached within the infiltrated water with respect to the total mass present in the dust and dirt
416 accumulated on the pavement surface. It can be seen that the mass released was relatively low, TN
417 being the most easily leached contaminant with 25% of the total mass released. This result also
418 indicates that in the pavements subjected to the Atlantic rainfall regime, with a higher rainfall
419 frequency, the percentage of pollutants released compared to the total deposited on the surface was
420 greater than in pavements under the Mediterranean regime. That is, although the concentration of
421 the different variables was greater in the water filtered in the Mediterranean regime pavements, as
422 the filtered volume was considerably lower, the total mobilized mass was lower than in the Atlantic
423 climate pavements. In general, in the tests carried out under conditions of low rainfall intensity, the
424 percentages of mass released increased as the degree of pollution increased. In the high rainfall
425 intensity tests the opposite occurred: the percentage of mass released was greater in the one-month
426 build-up level. With respect to the pavement configuration, in general the mass leached from the
427 pavements with complete configuration (C1) is lower than that from the simple configurations (C2).

428 The leaching capacity of the different types of rainfall tested was calculated by dividing the mass
429 released by the mass of dust and dirt deposited and the total volume of rainfall applied. These results,
430 also presented in **Table 4**, indicate that when the amount of dust and dirt deposited was lower (1
431 month) the rainfall was able to leach more pollutant per unit mass and volume. The results also show
432 that when it rained intensely, the mobilization and filtration of pollutants was greater than in low
433 intensity conditions, for both degrees of build-up. So, it is highly recommendable to perform an
434 adequate cleaning program with higher frequency during times of heavy rainfall.

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438

439 Table 4. Pollutants released in the water infiltrated in the different pavement configurations, rainfall regimes and
 440 intensities.

Rain intensity	Config.	Build-up (months)	TSS (%)	TSS (mg/kg/l)	COD (%)	COD (mg/kg/l)	TN (%)	TN (mg/kg/l)	TP (%)	TP (mg/kg/l)
Low Intensity	C1-Atl	1	0.4	273.2	10.8	328.9	12.1	10.2	2.5	0.5
	C1-Atl	6	0.3	35.5	12.7	64.7	25.0	3.5	8.4	0.3
	C2-Atl	1	0.5	331.1	12.2	374.4	9.7	8.2	3.6	0.7
	C2-Atl	6	0.2	24.8	12.4	63.0	24.9	3.5	8.6	0.3
	C1-Med	1	0.2	282.3	4.2	258.8	6.8	11.5	0.9	0.3
	C1-Med	6	0.2	51.3	11.8	120.0	15.6	4.4	7.3	0.5
	C2-Med	1	0.7	915.3	5.3	326.1	6.4	10.8	1.2	0.4
	C2-Med	6	0.2	47.4	13.7	139.7	17.0	4.8	9.8	0.6
High Intensity	C2	1	0.1	156.3	7.1	641.7	8.7	21.8	7.5	4.2
	C2	6	0.1	54.3	3.7	168.5	5.6	7.0	4.0	1.1

Note: the columns of percentages are the mass of pollutant released with respect to the total mass present in the dust and dirt deposited on the pavement surface; the columns expressed in mg/kg/l represent the mass of pollutant released per mass of dust and dirt deposited and volume of rain applied.

441

442

443

444 **Conclusions**

445 Permeable pavements can significantly control runoff production at source, reducing the negative
 446 impacts on sewage systems and favouring the infiltration of water into the ground. The quality of the
 447 infiltrated water is much better than that of the surface runoff generated from an impervious
 448 pavement. Therefore, the impact on the receiving water bodies is significantly reduced thanks to the
 449 reduction in the pollutant load received: lower concentration and lower volume. Nevertheless, a non-
 450 negligible load of nutrients is released with the drained water, so the installation of reactive barriers
 451 in the gravel layers would contribute to further improving the quality of infiltrated water. In this regard,
 452 the potential creation of anoxic zones by regulating outflows would contribute to a reduction in the
 453 concentration of nitrates in the effluent and favour the infiltration of water into the ground.

454 The infiltrated water gives high initial values of electrical conductivity and pH that decline as the
 455 pavement is washed repeatedly over time. Six months after installation, the characteristics of the
 456 filtered water make it suitable for irrigation in terms of salinity, pH and content of organic matter and

457 nutrients, if permeable pavements are planned to be used as rainwater harvesting systems. Therefore,
458 the manufacturing processes should pay attention not only to hydraulic and structural aspects but also
459 to aspects related to the quality of infiltrated water.

460 The results also demonstrate that an adequate cleaning program in dry conditions is essential,
461 especially when high rainfall intensities are forecasted, since the capacity for mobilization and
462 infiltration of pollutants is greater in these conditions. This is a key issue for the near future, as more
463 intense and frequent extreme precipitations are expected.

464 Finally, a complete configuration in permeable pavements provides a capacity to retain larger water
465 volumes under Mediterranean climate: the greater the thickness of the gravel layer, the greater the
466 reduction of the effluent volume achieved and the lower the contaminant load that reaches the
467 receiving environment. As the complete pavement configuration reduces the flows to a greater extent
468 it is considered that the probability of generating surface runoff will also be lower. Therefore, in cases
469 where the structural resistance requirements might not necessitate a complete configuration, such as
470 in pedestrian zones, the environmental benefits of the complete configuration are such that they can
471 eventually compensate a higher construction cost.

472

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482

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