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1 **Influence of the intrinsic characteristics of mortars on their biofouling by pigmented**
2 **organisms: Comparison between laboratory and field-scale experiments**

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14
15 **Abstract**

16 Biodeterioration of mortars by the photosynthetic microorganisms is affected by their
17 intrinsic properties such as porosity, roughness and surface pH. The influence of these
18 parameters was examined using an accelerated fouling test in laboratory and a natural fouling
19 test in the real-world (*in-situ*). Basing on color measurement and image analysis, the impact
20 of each intrinsic parameter was evaluated. The results differed from a scale to the other one.
21 No influence of porosity was measured on the algal colonization rate in the laboratory test
22 whereas, a high porosity seemed to increase slightly the bioreceptivity of the mortars exposed
23 outdoor. The roughness, in both tests, promoted the microbial colonization. However, the
24 discrimination of roughness grades was better in the laboratory test than in the *in-situ* one.

25 The surface pH influenced remarkably on the accelerated biofouling test but not on the *in-situ*
26 one. These dissimilarities resulted from the differences in experimental configurations of the
27 two tests.

28 **Keywords:** mortar, biofouling, intrinsic properties, laboratory test, *in-situ* test

29

30 **1. Introduction**

31 The colonization of building facades by microorganisms is a subject that concerns more
32 and more the civil engineering community. Indeed, this phenomenon changes the aesthetical
33 appearance of materials and, in later stage, can even compromise the durability of structures
34 by corrosion or by physical degradation induced by the microorganism (Maury-Ramirez et al.,
35 2013). The type of microorganisms colonizing cement based building facades is diverse. But
36 the analyses of *in-situ* samples show that algae are one of the initial and main colonizers
37 (Gaylarde and Gaylarde, 2005). These microorganisms allow the successive implantation and
38 the growth of other biological organisms such as lichens and bryophytes. If no solution is
39 found, the pteridophytes and higher plants may also appear (Perrichet, 1984; Deruelle, 1991;
40 Barberousse, 2006). The microbial development must be eliminated to avoid worse
41 consequences to buildings. The more the cleaning maintenance activities are repeated the
42 more expensive the cost.

43 Several factors such as the global climate, the local environment, the building design and
44 the facade materials influence the biological fouling.

45 The rainfall, the wind and the temperature consist in the climatic parameters influencing
46 together the water availability of facade materials which is an essential element to the
47 microbial metabolism. A permanently wet facade promotes the growth of algae and higher

48 plants (Ariño and Saiz-Jimenez, 1996; Loh, 2002). Therefore, the facades represent a higher
49 susceptibility to biofouling in the rainy regions and during the heavy rainy season (Young
50 1997). However, a high temperature induces water evaporation by heating the materials.
51 Similarly, the wind can cause a drying phenomenon.

52 The local environmental conditions, such as topography and nature of the ground,
53 presence of moisture and/or industrial activities around the buildings, should be taken into
54 account. As an example, a house roof, located close to seaside, can be totally colonized by
55 nitrophilous lichens, characterized by the yellow or orange, in a few years (Deruelle, 1991).
56 Indeed, sea sprays, by providing nitrogen to the building surface, favor the microorganism
57 growth. In addition, the humid environments set near a lake, a river, trees or shrubs, are
58 favorable to the appearance of stains on the walls.

59 The growth of biological fouling is also depending on the orientation of the facade.
60 Indeed, the north-facing facades which are wetter and less sunny, get colonize faster (Ariño
61 and Saiz-Jimenez, 1996; Young, 1997; Barberousse, 2006). Similarly, a facade exposed to
62 dominant winds seems to be colonized more easily than the other sides of the same building.
63 The wind can transport both the rain and the biological propagules at the facades promoting
64 the biofouling.

65 Parts of building often moistened for long periods, or easily covered of propagules, are
66 highly sensitive to the biological colonization (Wee and Lee, 1980). The biofouling often
67 grows at the foot of walls, the junction of different coatings and the overhanging elements
68 (cornices, moldings, balconies, etc.) (Wee and Lee, 1980; CSTB, 2005; Barberousse, 2006).

69 In addition to external conditions, the biological development is affected by the intrinsic
70 characteristics of the facade, defined as its bioreceptivity (Guillitte, 1995). These

71 characteristics can be divided into physical properties (porosity, roughness and hydrodynamic
72 properties) and chemical ones (chemical composition, pH surface).

73 Since the last thirty years, several researches have been devoted to the study of the
74 building-materials bioreceptivity. In all these studies, the experimental tests were conducted
75 with more or less accelerated methods of the biological growth on materials. The goal of these
76 accelerated methods was to reduce the test duration, which normally required many years in
77 the real-world. The experimental parameters were thus optimized to favor the growth of
78 microorganisms (Dubosc et al., 2001; Barberousse, 2006). Different methodologies,
79 concerning the preparation of materials, the process of inoculation and humidification, the
80 temperature and the photoperiod of the test were developed (Grant and Bravery, 1985;
81 Guillitte and Dreesen, 1995; Ohshima et al., 1999; Dubosc et al., 2001; Miller et al., 2006;
82 Miller et al., 2009; De Muynck et al., 2009; Tran et al., 2012). According to all these studies,
83 the roughness of materials appears as one of the most important parameters. Indeed, the
84 colonization of the building materials by microorganisms is promoted by the roughness of the
85 surface. In fact the roughness provides many asperities and thus increases the physical
86 anchorage of these micro-organisms (Dubosc, 2000; Tran et al., 2012). This effect of
87 roughness on the biofouling was confirmed by observations of buildings in real conditions
88 (Wee and Lee, 1980; Darlington, 1981; Pietrini et al., 1985; Joshi and Mukundan, 1997). The
89 total porosity, the pore size distribution and the porous network, should be taken into account,
90 because the absorption and the water retention of materials are controlled by these
91 characteristics (Warscheid et al., 1993; Ohshima et al., 1999; Crispim et al., 2003; Miller et
92 al., 2006; Miller et al., 2009). Furthermore, the inhibition of the algal growth by high pH of
93 material surface is commonly mentioned (Grant, 1982).

94 Most of previous studies propose only a qualitative evaluation of the biofouling or a
95 comparison of very different materials. In order to accelerate the fouling, mortars are usually

96 aged (by carbonation and/or leaching) to decrease the surface alkalinity. But the effect of the
97 accelerated aging is rarely evaluated. Moreover, the experimental conditions of accelerated
98 tests are significantly far from the real conditions. The investigations in field-scale are rarely
99 conducted because of the long duration of the experiments (Young, 1997; Ohshima et al.,
100 1999; Lengersfeld and Krus, 2004). Thus, the relationship between bench-scale experimental
101 results and field-scale experimental results is scant.

102 This work aims to clarify the effect of porosity, roughness and carbonation, on the
103 building-materials colonization by algae at laboratory and *in-situ* scale. One unique mortar
104 formulation has been investigated. The kinetic of the biological colonization was studied.

105 **2. Materials and methods**

106 *2.1. Preparation and characterization of samples*

107 Portland cement (CEM I 52.5, Holcim), siliceous sand (Sibelco DU 0.1/0.35), calcareous
108 filler (Omya) and cellulose ether (Hydroxylethyl Methyl Cellulose, SE-Tylose) were mixed
109 according to the proportions summarized in Table 1 to prepare the studied samples. The
110 water-to-cement ratio (*w/c*) (wt/wt) was fixed at 1 and 1.2. These two *w/c* were considered in
111 order to modify the porosity without changing the chemical composition of mortars.

112 Table 1

113 Mortar formulation

| Component | Cement | Sand | Calcareous Filler | Admixture ^a |
|-----------------------|--------|------|-------------------|------------------------|
| % mass of dry mixture | 30 | 65 | 5 | 0.27 |

114 ^a in addition to dry mixture (cement, sand and filler)

115

116 The variation of sample surface pH was achieved by a curing process. Mortars were
117 subjected (called carbonated mortars) or not (called uncarbonated mortars) to ageing by
118 accelerated carbonation prior to the start of biofouling test.

119 After mixing, the fresh mortar was casted into 50 cm × 50 cm × 1 cm extruded polystyrene
120 molds. The fresh mortar was stored at 21 ± 1 °C and 95 ± 5 % of relative humidity during 28
121 days for the uncarbonated samples. The mortar plates were then cut into samples of 20 cm × 8
122 cm × 1 cm and of 20 cm × 30 cm × 1 cm for the laboratory tests and the *in-situ* tests,
123 respectively. For the carbonated samples, the fresh mortar was stored in the aforementioned
124 conditions for only 7 days before being cut. This premature curing end aimed to shorten the
125 total duration of specimen production and to obtain the closest total storage time as possible
126 between carbonated and uncarbonated samples.

127 The samples were then exposed to pure CO₂ at 21 ± 1 °C and 65 ± 5 % of relative humidity
128 until the complete carbonation. This carbonation process required 36 days. The pH was
129 verified by spraying a phenolphthalein solution (0.2 % in ethanol) on the entire thickness of
130 specimen. The discoloration of the solution indicated a diminution of pH to about 9 (Thiery,
131 2005).

132 Moreover, the surface pH of mortar was measured by means of a surface electrode (WTW
133 Sentix Sur). For each sample, six measurements were performed at six positions distributed
134 over the sample surface. A constant volume of distilled water (0.1 ml) was deposited on the
135 related position. The electrode was kept in contact with the sample surface during a fixed time
136 of 30 seconds. The pH value was then recorded and the surface pH of sample was the average
137 of these six values. The surface pH of sample was monitored twice a week during the
138 carbonation process.

139 The pore size distribution and the total porosity of materials were determined using
140 mercury intrusion porosimetry (Micromeritics Autopore IV 9400). Three samples were
141 measured for each mortar formulation, i.e. w/c ratio of 1 and 1.2, both uncarbonated after 28
142 days of curing and carbonated after 7 days of curing and 36 days of accelerated carbonation.
143 They were beforehand dried by acetone to evacuate the open pores.

144 To obtain several surface roughnesses, three surface finishing methods were applied during
145 the sample setting. The first one consisted in smoothing the surface of fresh mortars with a
146 ruler and the two others in scratching the surface of the setting mortars with sponges of two
147 different roughnesses.

148 The roughness of the mortar specimens was evaluated by means of an optical profilometer
149 (CHR-150-L). The arithmetic average of the height R_a was determined from the
150 measurements of the surface profile (Gadelmawla et al., 2002). Measurements consisted in
151 analyze 161 profiles of 7 cm length and spaced of 500 μm . The data was recorded each 20 μm
152 along each profile. The value R_a , representative of the sample roughness, corresponded to the
153 average of 563.661 measuring points.

154 Each mortar was labeled by four codes. The first one corresponds to the w/c ratio, the
155 second to the carbonation state, the third to the roughness and the last to the scale of the
156 biofouling test. For example, a sample labeled 1UC-R1-Lab corresponded to the
157 uncarbonated mortar prepared with a w/c ratio of 1 with the smoothest roughness and tested in
158 the laboratory experiment.

159 2.2. Lab-scale and field-scale biofouling tests

160 The laboratory experiments consisted in an accelerated algal growth on cementitious
161 materials. A closed device containing an algal suspension was set up. The microalgae
162 *Klebsormidium flaccidum* was selected in this research basing on its representativeness and

163 ease of liquid culture. The suspension was periodically sprinkled on the samples surface. The
164 run-off period was fixed to 90 min every 12 h. The light was provided by two neon lamps
165 during 12 h per day, and was set to start simultaneously with the run-off cycles. All the
166 experimental configurations (rate of sprinkling, light intensity, temperature, initial
167 concentration of the algal suspension and angle inclination of the samples) were kept
168 constant. The details of the experimental approach were described in previous papers (Tran et
169 al., 2012; Tran et al., 2013).

170 In laboratory experiments, specimens with dimension of 20 cm × 8 cm × 1 cm were tested
171 in triplicate for each material. Carbonated and uncarbonated samples were tested separately.
172 Therefore, 18 samples were introduced in the test-chamber for each experiment.

173 The field-scale experiments consisted in a natural development of microorganisms on
174 cementitious materials. The mortar samples were placed on a stainless steel frame in a private
175 green park close to Grenoble, France. The sample dimensions were larger than the ones used
176 in the laboratory test, i.e. 20 cm × 30 cm × 1 cm. The larger dimensions aimed to minimize all
177 the non-representative effect resulted from the natural incidental events such as a random
178 intense inoculation of microorganisms at an individual sample points, the deposition of bird
179 droppings on the sample surface, etc.

180 The exposure of materials to the outdoor conditions started on 26 June 2009 and lasted one
181 year and a half. The experimental configurations were chosen in order to favor the biological
182 colonization of the sample surface. The samples were placed near trees, facing the north
183 direction and inclined with an angle of 45 °. The specimens were arranged in two rows of
184 which the first was located at 1 m above the ground to avoid splashing during rainy periods.
185 The contamination between specimens by water flow was also avoided (Fig. 1).



Fig. 1. Experimental set-up for the *in-situ* tests.

186

187

188

189 Each material was tested in triplicates. The mortars with a w/c ratio of 1 were studied at
190 carbonated and uncarbonated states, with the three roughnesses. For the samples with a w/c
191 ratio equal to 1.2, only the carbonated mortars with the two highest roughnesses were
192 examined.

193 2.3. Evaluation criteria of biofouling

194 The biological colonization of mortar surface was evaluated by means of image analysis
195 and colorimetric measurements.

196 Concerning the image analysis, the surface of samples was digitized using Epson V300
197 office scanner. The images obtained in the RGB color space were converted into the YIQ
198 color space. The distinction between the areas covered by algae and the surfaces of clean
199 mortars were achieved on the Q channel (Tran et al., 2012). The colonization rate ($X(t)$ (%))
200 was then given by the ratio of the fouled areas to the total surface of the sample. The samples
201 from the lab-scale and field-scale experiments were scanned every weekday and at least every
202 two months, respectively.

203 The colorimetric measurements were performed using colorimeters (Minolta CM-2600d
204 for the laboratory tests and Konica Minolta Chroma CR-410 for the *in-situ* tests). For each

205 specimen, the measurements were conducted at fixed positions spread over the whole sample
 206 surface. Thirty six and eighteen measurements were monitored on a lab sample and an *in-situ*
 207 one, respectively. The sample area of each measurement was of 0.5 cm² and 19.6 cm²,
 208 respectively. The CIE Lab color space data (L^* , a^* , b^* coordinates) were collected. The data
 209 was monitored three times a week on the lab specimens and at least every two months for the
 210 field-scale specimens. In order to evaluate the color changes of the test specimens, for each
 211 color coordinate (L^* , a^* , b^*), the difference between sample at a given time and the same
 212 sample at the initial time (uncolonized sample) was determined (ΔL^* , Δa^* , Δb^*).

213 3. Results

214 3.1. Characterization of materials

215 The porosity, the surface pH and the roughness of each mortar are given in Table 2. For the
 216 samples made with w/c ratio equal to 1 (i.e. uncarbonated and carbonated), the values of the
 217 porosity, the surface pH and the roughness R1 were those of a previous paper (Tran et al.,
 218 2012).

219 Table 2

220 Characteristics of mortars

| | Ratio w/c | Porosity (%) | Surface pH | | Code | R_a (μm) |
|--------------|----------------|-----------------|----------------|-------------|----------|-------------------|
| Uncarbonated | 1 | 37.2 ± 0 | 11 ± 0.4 | Roughness 1 | 1UC-R1 | 29 ± 5 |
| | | | | Roughness 2 | 1UC-R2 | 47 ± 6 |
| | | | | Roughness 3 | 1UC-R3 | 70 ± 8 |
| | 1.2 | 38.9 ± 0.3 | 11.0 ± 0.3 | Roughness 1 | 1.2UC-R1 | 29 ± 5 |
| | | | | Roughness 2 | 1.2UC-R2 | 47 ± 6 |
| | | | | Roughness 3 | 1.2UC-R3 | 123 ± 9 |
| Carbonated | 1 | 32.1 ± 1.9 | 9.0 ± 0.1 | Roughness 1 | 1C-R1 | 29 ± 5 |
| | | | | Roughness 2 | 1C-R2 | 47 ± 6 |
| | | | | Roughness 3 | 1C-R3 | 138 ± 15 |
| | 1.2 | 36.2 ± 0.1 | 8.9 ± 0.1 | Roughness 1 | 1.2C-R1 | 32 ± 4 |
| | | | | Roughness 2 | 1.2C-R2 | 47 ± 6 |
| | | | | Roughness 3 | 1.2C-R3 | 145 ± 18 |

221

222 The total porosity of uncarbonated mortars varied from 37 to 39 %, which is coherent with
223 the literature (Monge 2007). The mortars with the highest w/c ratio were slightly more porous.
224 According to Lafhaj et al. (2006), the porosity increases non-linearly with the w/c ratio. When
225 this w/c ratio is high, its effect on the total porosity is attenuated.

226 The carbonated mortars were less porous than the uncarbonated ones, i.e. 32 % for 1C
227 against 37 % for 1UC and 36 % for 1.2C against 39 % for 1.2UC. It is known that the
228 carbonation leads to a reduction of pore volume. Houst (1992) noted that this reduction of the
229 total porosity is even more enhanced by a low w/c ratio.

230 Depending on the curing process, two levels of surface pH were obtained. The surface pH
231 reached 11 with the standard curing (uncarbonated samples) and 9 by accelerated carbonation.
232 Moreover, no effect of the w/c ratio on the surface pH was detected. Indeed, the same value of
233 surface pH was measured for both w/c ratios.

234 Three levels of roughness were obtained whatever the curing process and the w/c ratio. The
235 smoothest surface (R1), without reliefs, was achieved by the method of finishing to the rule.
236 The rougher surfaces were obtained by scratching the sample surfaces with sponges. Both
237 strong reliefs and asperities appeared on the roughest surface (R3). Moreover, a remarkable
238 discrimination between the highest roughness (R3) and the intermediate one (R2) was
239 observed. In fact, the R_a values were 2.5 to 5 times higher for R3 than for R2. As a
240 comparison, the difference between the R2 and R1 was only about 1.5 times.

241 3.2. *Biological colonization of cementitious materials in laboratory and field test*

242 Fig. 2 illustrates the biological fouling of specimens investigated in the accelerated tests
243 and the *in-situ* experiments over time.

244

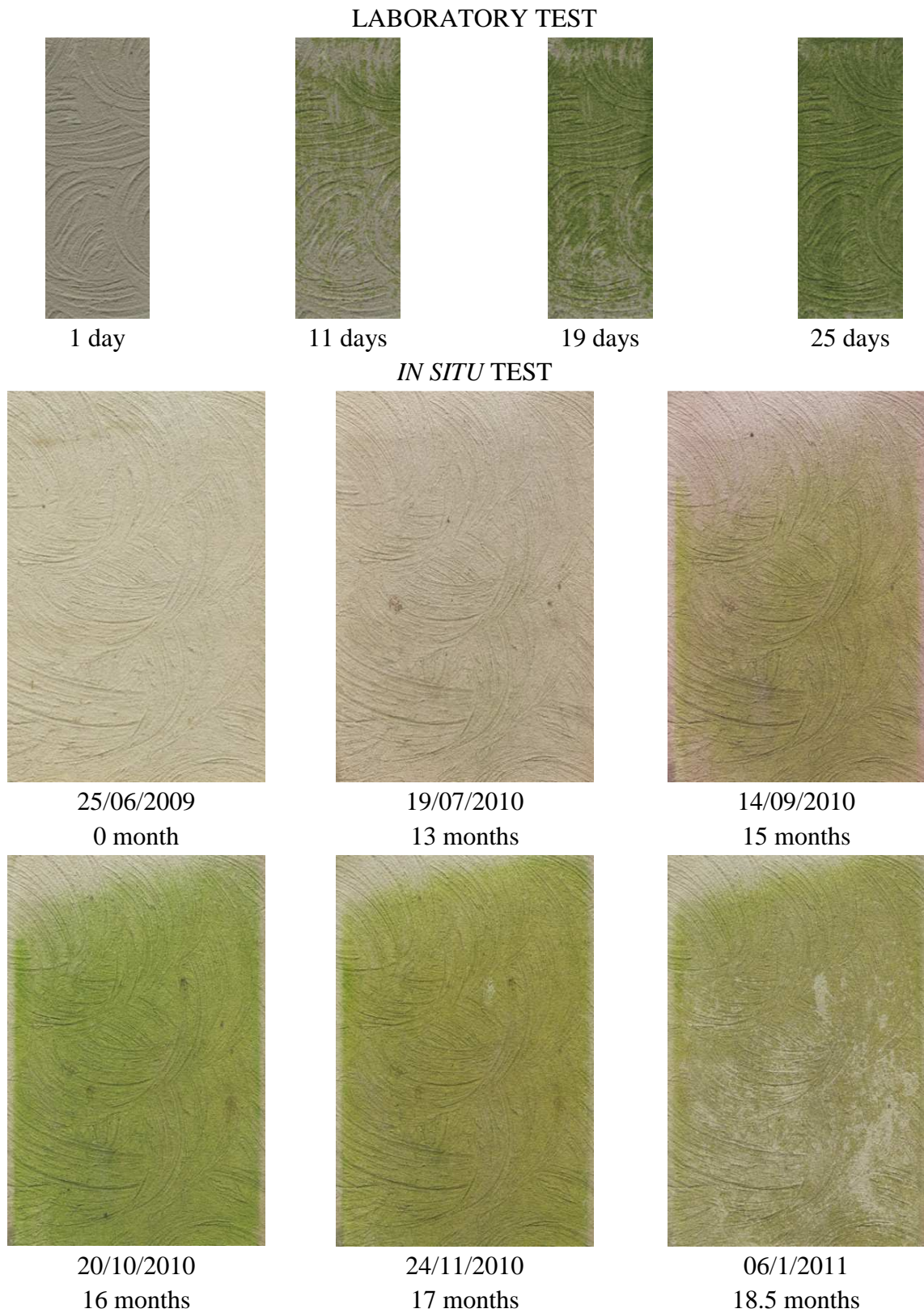
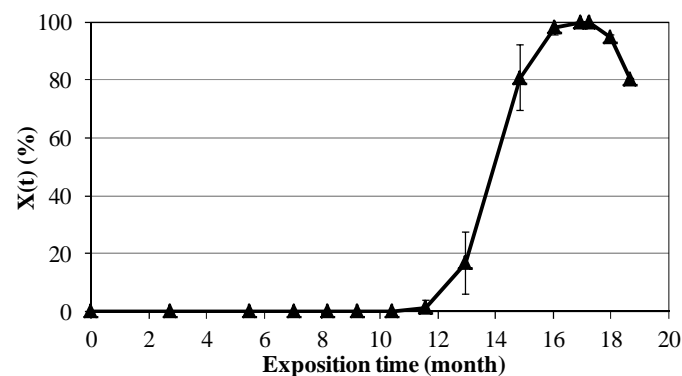


Fig. 2. Surface colonized by algae over time for carbonated mortars, w/c of 1.2 and roughness R3.

246 In the laboratory tests, the colonization of the surface by *Klebsormidium flaccidum* created
247 a dense and velvety mat as previously shown by Tran et al. (2012; 2013). The first algal spots
248 appeared at privileged sites of the surface, such as air bubble holes or asperities formed by the
249 roughness. The extension of fouling resulted from the growth of the existent spots and the
250 adhesion of new ones. On the sample surface of lowest roughnesses (R1 and R2), the algal
251 development formed streaks due to the suspension flow. This fouling form was usually
252 observed on the building facades. On the roughest mortars (R3), the fouling followed the
253 asperities on the surface which conducted the flows of the algal suspension.

254 The sample exposure in the natural conditions began at summer (end of June 2009). The
255 first settlement appeared on the mortars 1.2C-R3, after 11.6 months of exposure. The
256 colonization rate by microorganisms of this mortar surface is shown in the Fig. 3. Following
257 the latency period, the biological colonization developed rapidly. Four and a half months
258 later, the fouling was observed on the whole *in-situ* specimens.

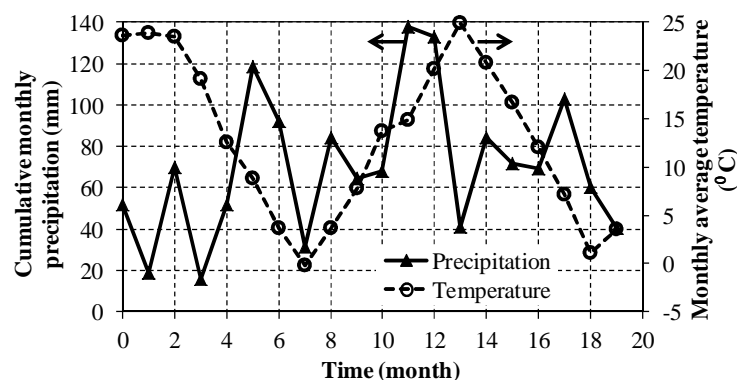


259 **Fig. 3.** Colonization rate for carbonated mortars, *w/c* of 1.2 and roughness R3 in the *in-situ*
260 test.
261
262

263 The same latency time was observed by Dubosc (2000) and Young (1997) for carbonated
264 mortars and sandstone samples, respectively. The biological development accelerated during
265 the autumn period (August to November, i.e. 13 to 17 months of exposure). The same

266 observation was made by Warscheid et al. (1993) on stone samples exposed to outdoor
267 conditions in different locations in Germany.

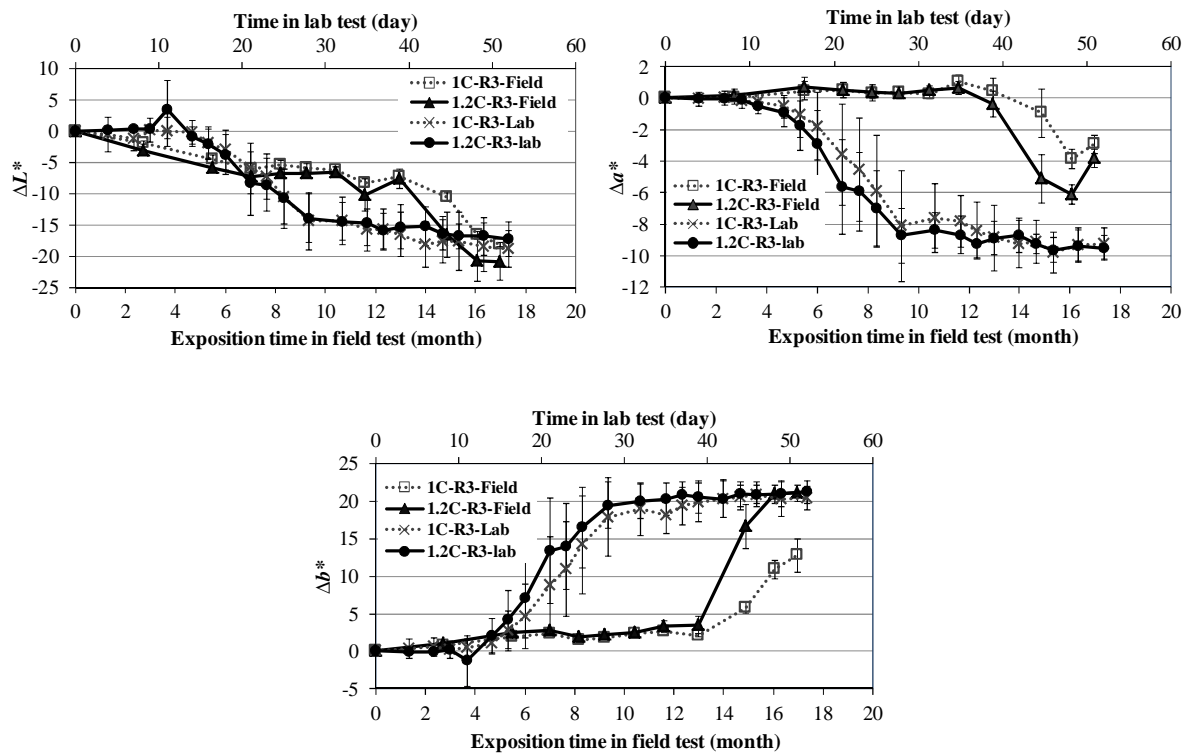
268 The propagules of microorganisms might be already settled on the mortar surface since the
269 spring. Fig. 4 shows the monthly average values of rainfall and air temperature during the
270 experimental time. In spring 2010 (April and May corresponding to 10 and 11 months
271 respectively), the meteorological data highlighted high precipitation and medium temperature
272 (i.e. about 15 °C). The following summer corresponded to the harsher climate, especially in
273 July (12-13 months), with the highest average temperatures and the lowest precipitations of
274 the year. During this period, the water available for the microorganism development was very
275 limited. Then mild temperatures accompanied with a suitable rainfall in autumn (September
276 and October corresponding to 15 and 16 months of exposure respectively) favored remarkably
277 the algal growth. From December (18 months), the detachment of algae from the samples
278 surface was observed. It corresponds to the winter period characterized by very low
279 temperatures, weak density of lighting and snow. The aging of microorganisms in this season
280 might be the reason of their detachment.



281
282 **Fig. 4.** Monthly average air temperature and cumulative precipitation during the *in-situ* test.
283

284 Fig. 5 represents the color changes of carbonated mortars of roughness R3, tested in
285 laboratory and field-scale. All the graphs show the mean values and the standard deviation of

286 three specimens. The evolution of clarity ΔL^* and chromaticities Δa^* , Δb^* showed the same
 287 trends for the two test scales. The ΔL^* , Δa^* decreased while the Δb^* increased with time. The
 288 biological colonization led to more and more dark (ΔL^*), green (Δa^*) and yellow (Δb^*)
 289 sample surface. This color is characteristic of green algae. The main colonizers on the *in-situ*
 290 samples could be attributed to these microorganisms.



291

292

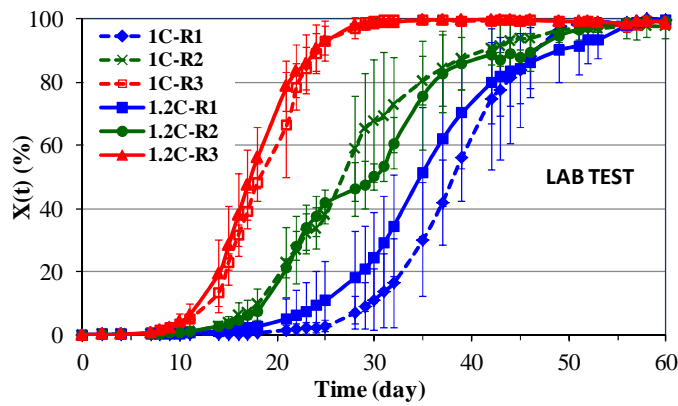
293 **Fig. 5.** Evolution of the clarity ΔL^* and the chromaticities Δa^* , Δb^* of mortars with the
 294 roughness R3 in lab- and field-scale tests.

295

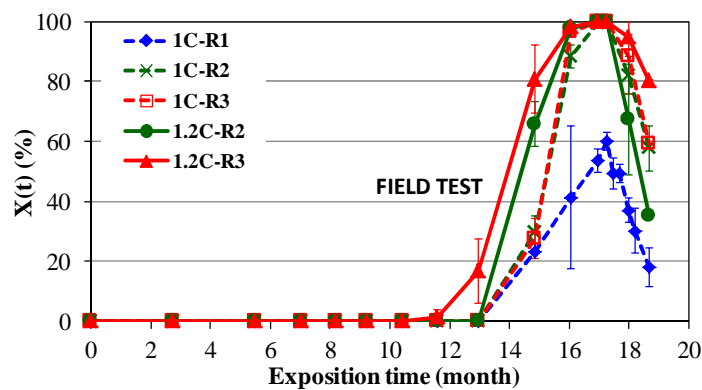
296 3.2.1. Effect of w/c ratio

297 Fig. 6 shows the influence of the w/c ratio (1 and 1.2) on the biofouling rate of carbonated
 298 mortars (three roughnesses).

299



a



b

Fig. 6 Effect of w/c ratio and roughness on the colonization rate for carbonated mortars in laboratory (a) and field test (b).

In the lab test, the algal colonization curve was of sigmoid type. Three steps can be identified: a latency step, an exponential growth step and a step of stagnation. For each roughness (R1, R2 and R3), no effect of the w/c ratio was identified on algal coverage. In fact, for the roughest mortars (R3), the fouling rate evolved quite identically. The algal colonization appeared after 6 testing days and the entire surface was covered after 30 days. For the two other roughnesses, with such experimental standard deviation, the colonization rate of the less porous mortars (1C) could be considered similar to the one of the more porous mortars (1.2C).

Unlike in the accelerated test, in the real conditions, an effect of the w/c ratio was detected. Indeed, the mortars made with a w/c ratio of 1.2 presented a faster microbial development

315 than the ones prepared with a w/c ratio of 1. For the 1.2C-R3 samples, the biofouling
316 appeared between 11 and 13 months of exposure. At the same moment, no algal colonization
317 was observed on the mortar 1C. The following measures, realized two months later, showed a
318 coverage area of 80 % for 1.2C mortars against only 30 % for 1C mortars. For the
319 intermediate roughness (R2), the effect of w/c ratio was less important. 30 % of the mortars
320 1C was colonized while it was 2 times higher on the mortar 1.2C. One month later, only the
321 more porous mortar was totally colonized.

322 The color changes of the roughest mortar confirmed the results obtained from the images
323 analyses (Fig. 5). Indeed, the color intensity of 1.2C-R3-Field mortars evolved faster than the
324 one of 1C-R3-Field mortars. Furthermore, the change in color intensity between these two
325 mortars occurred after 13 months of exposure. Concerning the bench-scale experiments, the
326 evolution of the color and the colonization rate coincided perfectly regardless of the w/c ratio.
327 Indeed, an increase in the color intensity was detected between 4 and 28 days of exposure.
328 Then, no color evolution was observed since the 28th days of testing, corresponding to the
329 necessary time for a complete biofouling.

330 *3.2.2. Effect of the roughness*

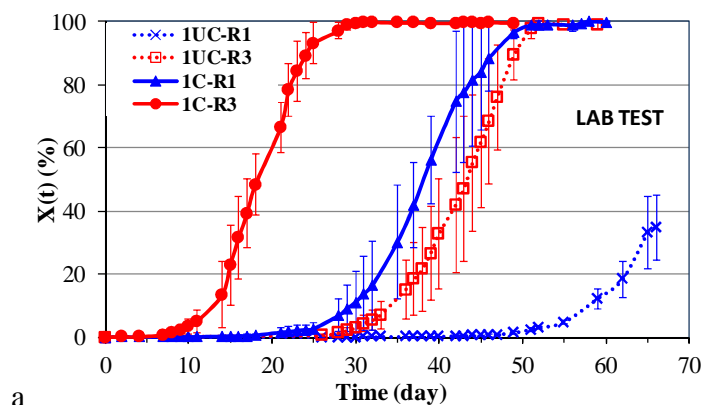
331 For the bench-scale test, the biofouling rate was obviously influenced by the surface
332 roughness of samples (Fig. 6a). The effect of the three levels of roughness tested on the
333 biofouling were quite distinct, whatever the w/c ratio (1 and 1.2). The smoothest roughness
334 (R1) exhibited the lowest colonization rate and the highest latency time. Indeed, the latency
335 time was around 18 days of testing for roughness R1, against 9 and 6 days for roughness R2
336 and R3, respectively. Similarly, the complete colonization was achieved more quickly on the
337 rougher mortars, i.e. in 30 days for R3, against 51 and 56 days for R2 and R1, respectively.
338 Furthermore, the curve slope evolved in the same way as the roughness during the

339 exponential growth phase. The more the roughness was high, the more the colonization was
340 quick.

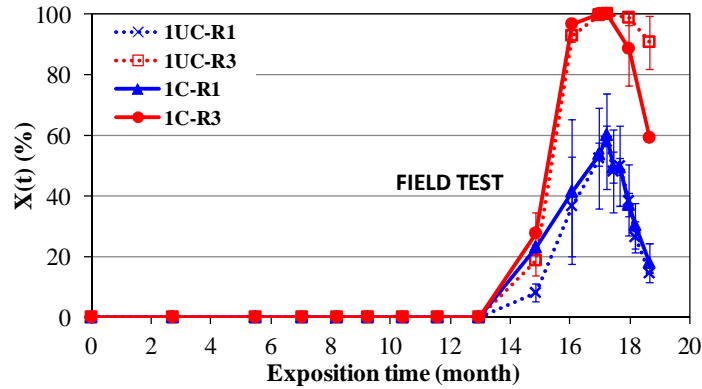
341 The roughness impacted also the biological development of mortars exposed in the real-
342 world (Fig. 6b). However, unlike bench-scale test, only two grades of roughness (i.e. rough
343 and smooth) were observed. The mortars of very different roughness, R2 and R3, showed no
344 difference in bioreceptivity to visible photosynthetic organisms. This result was particularly
345 obvious for $w/c = 1$. Conversely, a transition from a smooth surface (R1) to a rough one (R2)
346 remarkably increased the bioreceptivity of mortars. Indeed, the colonization appeared slower
347 for the mortar R1 than for the two others. The entire surface of samples R2 and R3 was
348 colonized after 17 months (November 2010). For the same duration, only 60 % of mortar
349 surface R1 was colonized. Beyond this time, a decrease in the covered area was noticed for all
350 samples. It has been assigned to detachment of algae. The detachment rate was similar for all
351 mortars of w/c equal to 1. However, for mortar of w/c equal to 1.2, smooth surface showed a
352 faster decrease in the coverage area compared to the rough one. Hence, the effect of
353 roughness on the microbial detachment rate remains difficult to analyze.

354 3.2.3. Effect of the accelerated carbonation

355 Fig. 7 illustrated the evolution of the colonization rate for carbonated and uncarbonated
356 mortars mixed with a w/c ratio of 1, for the lowest and the highest roughness.



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b

Fig. 7. Effect of accelerated carbonation on the colonization rate for mortars mixed with a w/c ratio of 1, with roughness R1 and R3, in laboratory (a) and field test (b).

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The carbonation seemed to be the most decisive parameter on the bioreceptivity of materials for the accelerated fouling test. It significantly shortened the latency time, and accelerated the rate of algal colonization. As showed in the Fig. 7, the latency time of 1C-R1 mortar was of 18 days. This value was 2.6 times less than that of 1UC-R1 mortar (46 days). Similarly, the colonization of 1C-R3 mortar began after 7 days against 26 days for 1UC-R3 mortar. The complete colonization was reached in 30 days for the 1C-R3 mortar against 51 days for the 1UC-R3 one. It is worth noting that, the total colonization of 1UC-R1 was not obtained because of a precocious stop of testing. Nevertheless, basing on the roughest mortars, the slope of the curve was greater for carbonated mortars than for uncarbonated ones.

In the field-scale test, it seems that the accelerated carbonation did not result in different growth profiles. The biofouling evolved quite identically on carbonated and uncarbonated mortars. For both mortars 1UC and 1C, and of roughness R1 or R3, the colonization by microorganisms began at the same moment, i.e. after 13 months. Then, 4 months later (November, corresponding to 17 months of exposure), the entire surface of all rough samples (R3) was colonized. At this same time, the algal colonization reached a common rate of around 60 % for smoothest samples (R1), carbonated or uncarbonated. From 18 months

378 (December 2010), a decrease of fouled area was observed on the entire surface due to the
379 microbial detachment. It seems that the detachment rate was not dependent on the initial
380 carbonation state of mortars.

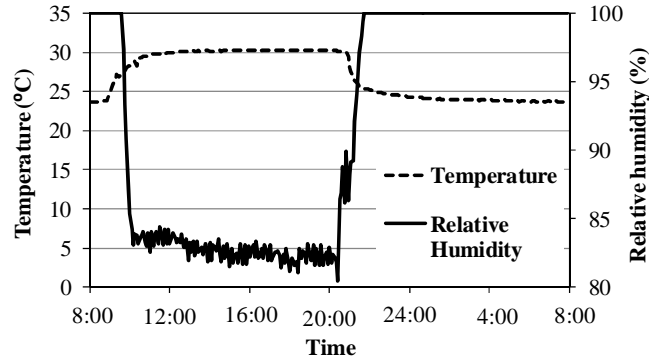
381 **4. Discussion**

382 The results obtained from the color measurements and the image analysis, are coherent and
383 complementary. Indeed, the effect of intrinsic characteristics of mortars on their bioreceptivity
384 was found identical by these two methods. Due to monitoring the entire surface of samples,
385 the color changes are well representative of the fouled area.

386 The color evolution of the surface of the samples, for both laboratory and *in-situ* tests, was
387 quite identical, i.e. more and more dark, green and yellow. The fouling of the *in situ* samples
388 is thus presumably associated to green algae. These microbial species are the primary and
389 main colonizers on building facades as noticed by several authors (Wee and Lee, 1980; Grant,
390 1982; Ortega - Calvo et al., 1995; Tomaselli et al., 2000; Barberousse, 2006).

391 The influence of *w/c* ratio on the accelerated fouling was negligible. The reason is related
392 to experimental conditions rather than to modest difference in porosity between two studied
393 mortars. Indeed, the relative humidity in the laboratory test chamber is permanently very high
394 (i.e. from 80 to 100 %) (Fig. 8). In this condition, the water content into the samples is always
395 abundant and so is sufficient to the algal development.

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Fig. 8. Daily cycle of temperature and relative humidity in the test chamber.

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400 In order to verify this assumption, the filling rate of pore by water (ε (%)) was calculated
 401 (Dubosc, 2000). It corresponds to q/p ratio, where p (%) is the sample porosity, and q (%) is
 402 the amount of water absorbed by a unit of sample volume, calculated as follows:

$$q(\%) = 100 \times \frac{V_{water}}{V_{sample}} = 100 \times \frac{m_{water}}{\rho_{water} \times V_{sample}}$$

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404 V_{water} : volume of water absorbed by the sample (cm^3);

405 V_{sample} : apparent volume of the sample (cm^3), calculated from three dimensions of sample;

406 m_{water} : amount of water absorbed by the sample (g) that is equal to the difference between
 407 the mass of sample in the humid condition and in the dry one.

408 ρ_{water} : density of water (1 g.cm^{-3})

409 The humid sample weight was measured 6 times along the diurnal cycle and 2 times along
 410 the nocturnal one. Before the start of accelerated fouling test, the sample was dried at $40 \text{ }^\circ\text{C}$
 411 until obtaining constant weight. This weight was considered as the mass of dry sample.

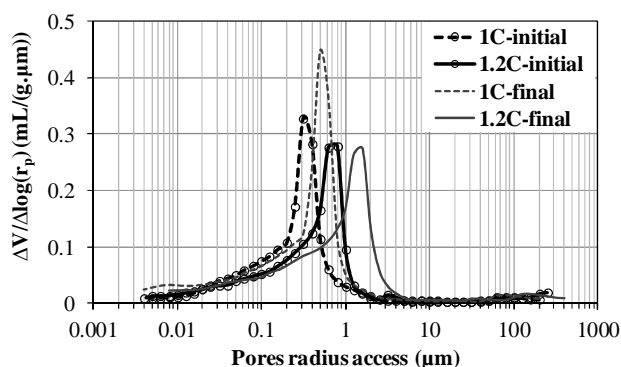
412 The amount of water absorbed by the sample reached a maximum value just after the
 413 sprinkling cycle and remained constant during all the nocturnal period. During the diurnal
 414 period, the water evaporated under the warming effect induced by neon lights. However, the

415 amount of evaporated water was weak. Indeed, at the end of the diurnal period, the remaining
416 water into the mortars filled at least 75 % and 60 % of the porosity of uncarbonated and
417 carbonated samples, respectively.

418 The water, being an indispensable element for the biological metabolism, is thus no longer
419 a limiting factor for the biological development. This observation was confirmed by previous
420 studies regarding two mortars made with *w/c* ratios equal to 0.5 and 1 (Tran et al., 2012;
421 2013). Despite of the considerable difference in porosity, these mortars exhibited completely
422 identical bioreceptivity.

423 In natural condition, the impact of the *w/c* ratio on the colonization rate of samples was
424 exhibited. The microbial fouling seemed to be favored by the highest *w/c* ratio (1.2).

425 To evaluate the porous network of mortars, analyses were performed after 17 months of
426 exposure to the outdoor conditions. PIM's results indicate a modification of the porous
427 network. Indeed, the total porosity of the 1C-R3 and 1.2C-R3 mortars increased from 32 to 36
428 % and from 36 to 38 %, respectively. Moreover, the single peak characterizing the pore
429 entrance diameter shifted to larger diameters (Fig. 9). Indeed, the pore inlet diameter, that was
430 equal to 0.3 μm at the beginning of outdoor exposure, was about 0.5 μm at the end of the
431 experiment for the 1C mortar. For the 1.2C mortar, the pore inlet diameter evolved from 0.75
432 to 1.6 μm . The enlargement of the pore entrance diameter could be due to the leaching effect
433 by rain and to the activities of biological agent such as bacterial communities and/or fungi
434 (Warscheid and Braams 2000). These results, which corresponded to the average values of the
435 entire thickness of samples, might be amplified at the surface.



436
 437 **Fig. 9.** Pore size distribution of carbonated mortars with a w/c ratio of 1 and 1.2 at the
 438 beginning and end of the *in-situ* test.

439
 440 The mortars made with a w/c ratio of 1.2 were more porous than those formulated with a
 441 w/c ratio of 1 and exhibited a greater bioreceptivity. The effect of porosity on the biofouling
 442 rate of samples exposed outdoor are in agreement with the literature (Warscheid et al., 1993;
 443 Ohshima et al., 1999; Crispim et al., 2003; Prieto and Silva, 2005; Miller et al., 2006; Miller
 444 et al., 2009).

445 Previous studies highlighted an important role of roughness on the biological colonization
 446 (Prieto and Silva, 2005; Miller et al., 2006; Miller et al., 2009; Tran et al., 2012; Tran et al.,
 447 2013). By providing asperities, the roughness favors the physical attachment of
 448 microorganisms which is dispersed by the water flow or the wind. Consequently, the rougher
 449 the surface is, earlier the biological fouling begins and faster the colonization is. The recent
 450 accelerated fouling experiments allow confirming the effect of roughness. Indeed, the three
 451 grades of roughness studied induced a discriminating effect on the colonization rate. The
 452 same results were noticed by Tran et al. (2012) on others mortars mixed with a w/c ratio of
 453 0.5.

454 For *in-situ* samples, the visible biofouling due to phototrophic organisms appeared
 455 approximately at the same moment whatever the roughness of mortars. Indeed, for all the
 456 roughnesses, the first spots were detected after about 13 and 15 months of exposure. Based on

457 the results of laboratory tests, the *in-situ* samples of the roughness R3 should have been firstly
458 colonized, successively followed by mortars of the roughness R2 and then R1. However, in
459 the *in-situ* test, the season, the external relative humidity and temperature could impact the
460 colonization and thus hid the roughness effect.

461 In the natural conditions, only two grades of roughness were observed. The two roughest
462 mortars showed the same bioreceptivity to pigmented organisms despite considerably
463 different roughness. This tendency remains coherent with that obtained in laboratory test. In
464 fact, a nonlinear relationship between roughness and algal fouling rate is noticed. A close
465 bioreceptivity was observed between mortars of high roughness (Tran et al., 2012). The non-
466 differentiation in the outdoor test could be induced by a natural inoculation much less intense
467 than in accelerated tests.

468 Concerning the accelerated carbonation, this intrinsic characteristic of the samples
469 appeared as the most important parameter which affected the biological growth in the
470 accelerated fouling test. Unlike the effect of roughness which favors the ability of algae to
471 physically cling to the surface, the pH and/or carbonation disturbs the biological metabolism.
472 Carbonation leads to a decrease in the surface pH of samples and to a lower increase in pH of
473 the algal suspension flowing on test specimens. Consequently, the *K. flaccidum* algal cells are
474 in less alkaline conditions and thus in less stressful environment for their growth with
475 carbonated samples than with uncarbonated ones. Indeed, *K. flaccidum* is known as an
476 acidophilic microorganism. The ability of attaching and growth of algae on a surface are thus
477 promoted by the carbonated of samples (Tran et al., 2012; 2013).

478 The growth of the algal spots on the sample surface is the consequence of the vegetative
479 and cellular multiplication which is favored at low pH (Škaloud, 2006). The attachment of
480 algae on the surface of the substrate is governed by adaptive metabolic interactions between

481 algal cells and the substrate (Fattom and Shilo, 1984; Finlay et al., 2002; Barberousse et al.,
482 2006). The algal extracellular polysaccharides could play the role of glue (Robins et al., 1986;
483 Gantar et al., 1995; Barberousse et al., 2006). These metabolites depend on the algal species
484 considered and the substrate. They are involved in the initial contact between the cell and the
485 surface, and act over time (Barberousse et al., 2006).

486 As a consequence, carbonated mortars are highly receptive to colonization by
487 microorganisms. Algal colonization begins much earlier, and occurs faster compared to the
488 uncarbonated mortars.

489 In the field-scale test, the influence of carbonation was not highlighted. Indeed, the
490 biofouling of mortars, carbonated or uncarbonated, was detected at the same moment.
491 Perhaps, the favorable climatic conditions and sufficient inoculation conditions were not met
492 at the beginning of testing. In fact, the samples were exposed through the summer. Therefore,
493 spring, the most favorable season for the microorganism spreading and their growth was past.
494 Hence, the microbial colonization of carbonated samples was not initiated despite a favorable
495 surface pH. After one year of exposure, the mortars were aged and weathered by leaching and
496 natural carbonation. Consequently, the same surface pH (pH = 8) was measured for all the
497 samples whatever their initial carbonation state. The bioreceptivity of mortars is thus identical
498 when favorable conditions to the biological development have been in place. Therefore, the
499 influence of the initial surface pH on biofouling is completely inhibited.

500 **5. Conclusion**

501 The present research has investigated the effect of the intrinsic parameters (porosity,
502 roughness and carbonation) of mortars on their biofouling by photosynthetic organisms. This
503 study was conducted by both laboratory accelerated tests and *in-situ* ones.

504 The impact of porosity on the biofouling of mortars is different for the two experimental
505 test scales. This parameter has no effect on the biofouling rate in laboratory tests due to the
506 experimental conditions. However, although more experiments of verification still required
507 for the field-scale tests, high porosity seems to favor the biological colonization.

508 For the both test scales, the influence of the roughness is evidenced. A rough surface
509 enhances the biological attachment. The discrimination of roughness grades was better in the
510 accelerated tests than in the field-scale ones. This result could be explained by the fact that the
511 accelerated test is performed in a closed circuit with an intense inoculation while the *in-situ*
512 test involves natural inoculation.

513 Thanks to its experimental configuration, the laboratory tests allow revealing the role of
514 the accelerated carbonation. It is considered as a decisive parameter in the accelerated fouling.
515 Indeed, carbonated mortars lead to a lower pH value of the mortar surface and the algal
516 suspension in the test chamber. The algal development is thus significantly promoted and so
517 the colonization rate is remarkably accelerated in the case of carbonated mortars. While in the
518 natural conditions, due to numerous uncontrolled and random environmental factors, the
519 effect of the carbonation was not observed. The evolution of biofouling is identical on the
520 both carbonated and uncarbonated mortars. Due to the long exposure time and progressive
521 ageing by lixiviation and carbonation, the surface pH, and thus the alkalinity, of carbonated
522 and uncarbonated samples is the same.

523 The divergence of results between the laboratory and field-scale tests, prevent to correlate
524 the two experimental scales. These dissimilarities could be remediated by improving the
525 experimental protocol. In the accelerated test, a drying phase of samples between two
526 sprinkling cycles could allow highlighting the role of porosity. A reasonable choice of the
527 beginning of sample exposure to natural environment could highlight the effect of

528 carbonation. The effect of roughness in accelerated test could be closer than that in natural
529 test by decreasing the inoculation intensity. This decrease in inoculation process could be
530 achieved by reducing, either the initial concentration of the algal suspension or either the run-
531 off.

532

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