

**Spectrophotometric color measurement for early detection and monitoring of greening on granite buildings**

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1 This paper addresses the detection and monitoring of the development of epilithic  
2 phototrophic biofilms on the granite façade of an institutional building in Santiago de  
3 Compostela (NW Spain), and reports a case study of preventive conservation. The results  
4 provide a basis for establishing criteria for the early detection of phototrophic colonization  
5 (greening) and for monitoring its development on granite buildings, by use of color changes  
6 recorded with a portable spectrophotometer and represented in the CIELAB color space. The  
7 results show that parameter  $b^*$  (associated with changes of yellowness - blueness) provides  
8 the earliest indication of colonization and varies most over time, so that it is most important  
9 in determining the total color change. The limit of perception of the greening on a granite  
10 surface was also established in a psychophysical experiment, as  $\Delta b^*$ : + 0.59 CIELAB units  
11 that corresponds, in the present study, to  $6.3 \mu\text{g biomass dry weight cm}^{-2}$  and  $(8.43 \pm 0.24) \times$   
12  $10^{-3} \mu\text{g extracted chlorophyll } a \text{ cm}^{-2}$ .

13 **Keywords:** Preventive conservation; CIELAB color system; biofouling; greening;  
14 monitoring; phototrophic colonization.

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16

## 17 **Introduction**

18 The fouling of stone surfaces by abiotic substances and organisms poses serious problems  
19 for the maintenance of all types of buildings. Fouling by microorganisms, known as  
20 biofouling, is considered to start with phototrophic organisms (algae and cyanobacteria)  
21 conditioning the inert surfaces for subsequent growth of heterotrophic organisms (eg Grant  
22 1982; Albertano 1991; Ortega-Calvo et al. 1993; Saiz-Jimenez and Ariño 1995; Prieto et al.  
23 2005; Miller et al. 2010). However in the absence of a primary colonizing film of  
24 phototrophs, heterotrophic bacteria and fungi can grow on building façades, using organic  
25 compounds from organic pollutants, painted surfaces or conservation treatments (eg  
26 Cappitelli et al. 2005, 2007). Unlike heterotrophic biofilms, phototrophic biofilms have  
27 received little attention until recently (Di Pippo et al. 2009, 2011).

28         Biofilms that grow in areas exposed to light tend to contain photosynthetic organisms  
29 (Ramírez et al. 2010). The presence of these pioneer photosynthetic-based microbial  
30 communities generally result in the appearance of thin green films, which usually adhere to  
31 the stone substrate (ICOMOS-ISCS 2008) in a phenomenon often referred to as greening.  
32 The presence of greening depends on a variety of factors, such as a suitable combination of  
33 dampness, warmth and light on the stone surface (Tiano 2002) as well as the intrinsic  
34 characteristics of stone, such as permeability, porosity and surface roughness, which  
35 influence its bioreceptivity (Guillitte 1995; Prieto and Silva 2005; Miller et al. 2006). Factors  
36 such as the exposure site (Barberousse et al. 2006), the presence or absence of adjacent  
37 vegetation (Smith et al. 2011) and the urban or rural location (Tanaca et al. 2011) also affect  
38 the development of biofouling.

39         The study of greening on building façades deserves considerable attention because  
40 pollution-related soiling has been widely superseded by algal growth. Since the beginning of  
41 the 20<sup>th</sup> century, when the first data on the aspect of some important buildings were recorded,  
42 there has been an apparent decrease in the occurrence of black deposits (Newby et al. 1991;  
43 Davidson et al. 2000; Brimblecombe and Grossi 2009), partly because of the decrease in air  
44 pollution resulting from the declining use of coal and the switch to gas and electrical heating

45 (Brimblecombe 1987; Grossi and Brimblecombe 2008). At the same time, there has been an  
46 increase in biological activity, favored by lower concentrations of sulfur dioxide and greater  
47 deposition of organic compounds and nitrogen (Grossi and Brimblecombe 2008). Higher  
48 humidity caused by climate change also favors the development of greening as it increases  
49 the time that stone structures remain wet and possibly the depth of penetration of moisture.  
50 This has already been observed on buildings in places such as Northern Ireland and London,  
51 as respectively an increased incidence of algal greening (Smith et al. 2011) and decreased  
52 incidence of blackening (Brimblecombe and Grossi 2009).

53         Several authors have demonstrated that the discoloration of stone caused by the  
54 growth of microorganisms can be easily determined by a non-invasive (non-destructive)  
55 technique based on measurement of the reflectance by a spectrophotometer or a tristimulus  
56 colorimeter. The data are expressed in CIE-L\*a\*b\* color system units (Wyszecki and Stiles  
57 1982; Sanmartín et al. 2010). Using this technique, Urzi and Realini (1998) correlated the  
58 orange or grey color of the patina on Noto's calcareous sandstone with the associated  
59 microflora; Prieto et al. (2005) quantified the development of biofilm induced on the open  
60 rock faces of quartz quarries to reduce the visual impact; De Muyneck et al. (2009) evaluated  
61 strategies for preventing algal fouling on two types of concrete (man-made stone), and more  
62 recently Tanaca et al. (2011) evaluated fungal colonization on three fiber cement (man-made  
63 stone) formulations exposed to urban, rural and coastal environments. Nevertheless, to the  
64 authors' knowledge, the color measurement technique has not previously been used to study  
65 the phototrophic colonization (greening) of granite stone buildings naturally exposed to the  
66 outdoor environment.

67         Apart from the chemical and/or physical deterioration of buildings caused by  
68 phototrophic-based microbial communities, greening must be considered as aesthetic damage  
69 that depends not only on the general conditions of the local environment, but also on the  
70 individual perception by the people involved (Smith et al. 2011). In this respect, green is  
71 more easily perceived than other colors, since the differences detected by the human eye are  
72 not of the same magnitude in the different parts of the spectrum; wavelengths close to 400  
73 (blues) and 700 nm (reds) are less important from the point of view of perception than those  
74 around 500 nm. In fact, 560 nm (corresponding to the green area) is the wavelength at which

75 the human eye is most sensitive (Gescheider 1976; McDonald 1997). Green color had been  
76 used for the quantification of biomass of the green alga *Ulva* (syn. *Enteromorpha*) on test  
77 panels coated with a range of experimental formulations (Cassé et al. 2007). Knowledge of  
78 the threshold of perception of greening by the human eye would be very useful for the  
79 effective and sustainable management of stone buildings and monuments. To date, few  
80 studies have addressed the human perception of changes in the appearance of stone due to  
81 general fouling, and most of the existing studies have investigated the changes in appearance  
82 in terms of blackening or darkening (Newby et al. 1991; Andrew 1992; Grossi and  
83 Brimblecombe 2004; Brimblecombe and Grossi 2005). As far as the authors are aware, only  
84 one laboratory-based study has examined the limits of perception of the phototrophic  
85 colonization on building stones (Prieto et al. 2006). In the latter study the qualitative terms,  
86 ranging from inappreciable to very intense, referred to the change in appearance caused by  
87 the live microorganisms on the surface of granite rock were related to the total color change  
88 ( $\Delta E^*_{ab}$ ). However, variations in the three color parameters, lightness-darkness  $\Delta L^*$ , redness-  
89 greenness  $\Delta a^*$  and yellowness-blueness  $\Delta b^*$ , were not analyzed. In the present study, these  
90 variations were taken into account with the aim of selecting the parameter that provides most  
91 information regarding the detection and monitoring of greening on granite façades.

92 Thus, the overall aim of the study was to demonstrate the suitability of a spectrophotometric  
93 measurement technique for detecting phototrophic colonization (greening) and real-time  
94 monitoring of the development of the colonization in a real case of a granite building. The  
95 specific purposes of the study were as follows: (1) to establish the threshold of human  
96 perception of greening on granite rock and its value in terms of partial color differences ( $\Delta L^*$ ,  
97  $\Delta a^*$  and  $\Delta b^*$ ), amount of phototrophic biomass (dry weight) and extracted chlorophyll *a*  
98 content; (2) to analyze the changes in the  $L^*$ ,  $a^*$  and  $b^*$  CIELAB coordinates in a real case  
99 involving the cleaning and recolonization of a granite façade, with the aim of determining  
100 the parameter or parameters that best indicate the start of the greening process and (3) to  
101 describe a methodology that could be used as a tool for making decisions about the required  
102 frequency of cleaning and biocide treatment in a real case.

## 103 **Materials and methods**

104 The perception of color depends on the incident light and variations in illumination, thus  
105 changes in the natural light affect the judgment of the observer about the color observed. For  
106 this reason, a visual sorting task used to estimate the threshold of perception of greening by  
107 the human eye was carried out in a laboratory test, under standard conditions of illumination.  
108 Detection and monitoring of greening were performed outside, since measurements made  
109 with a portable spectrophotometer are not affected by external light parameters.  
110 Psychophysical and monitoring experiments were carried out with granite rocks of similar  
111 color characteristics and mineralogical nature in order to obtain comparable results.

### 112 *Psychophysical experiment*

113 A visual sorting task (psychophysical experiment) was conducted to investigate the threshold  
114 of perception of greening on granite surfaces. Blanco Cristal, a medium-grain,  
115 heterogranular-panalotriomorphic, biotitic adamellitic leucogranite (with dark minerals  
116 absent), and feldspar-K, plagioclases, quartz, biotite, chlorite and moscovite as major  
117 minerals was selected for the experiments. The petrographic characteristics and mineral  
118 composition of this lithotype were described in a previous study (Sanmartín et al. 2011).  
119 Twenty-one blocks (6 x 3 x 1 cm) were cut and sterilized before starting the experiments.  
120 The upper surface of each granite block (18 cm<sup>2</sup>) was inoculated in a laminar flow cabinet  
121 with a mixed culture of three isolates of subaerial stone biofilm-forming cyanobacteria grown  
122 in BG11<sub>0</sub> medium (Rippka et al. 1979), viz. *Nostoc* sp. PCC 9025, *Nostoc* sp. PCC 9104 and  
123 *Scytonema* sp. CCC 9801. The mixed inoculum consisted of 0.21 mg (dry weight) of each  
124 strain per ml of medium giving 0.63 mg (dry weight) per ml of mixed inoculum. Distinct  
125 volumes from 150 µL (the minimum volume required to fully cover the total area of 18 cm<sup>2</sup>  
126 of the surface block) to 540 µL mixed cyanobacteria suspension, were inoculated uniformly  
127 with the point of a pipette onto the surface blocks, in order to adjust phototrophic biomass  
128 between 0 (to exclude the possibility of an abiotic contribution to the green color) and 18.9  
129 µg biomass (dry weight) per cm<sup>-2</sup> surface area. Experiments were performed in triplicate.

130 Each inoculated block was assessed by eight different observers (five females and  
131 three males aged from 25 to 60 years) with normal color vision ie normal trichromat  
132 observers without color blindness (see Fletcher and Voke (1985) for details of color vision  
133 examination). The task of each observer was to decide if the greening due to the presence of  
134 organisms was perceptible. The responses reported by the observers (yes/no) were recorded  
135 and analyzed. The point at which the observers began to note the green color has been coined  
136 as the just noticeable difference (jnd). The conceptualization of the greening threshold has  
137 its roots in the study of thresholds for other sensory-related stimuli (Gescheider 1976).

138 The color of the upper surface of each granite block was measured before and after  
139 inoculation, following the methods proposed by Prieto et al. (2010a, b). The CIELAB  
140 coordinates ( $L^*$ ,  $a^*$  and  $b^*$ ) were measured before and after inoculation, with a portable  
141 spectrophotometer (Konica Minolta CM-700d/600d) equipped with CM-S100w  
142 (SpectraMagic<sup>TM</sup> NX) software; the measuring conditions were illuminant D65, observer  
143 2° and a 8-mm diameter viewing area. A total of 14 readings were taken at different randomly  
144 selected zones on each wet surface block, and the results expressed as the mean values.

145 To relate the phototrophic biomass (dry weight) to color and extracted chlorophyll *a*  
146 content, the concentration of chlorophyll *a* on each block was determined after the visual  
147 evaluation and color measurements, following the protocol for the extraction of chlorophyll  
148 *a* from microorganisms colonizing rocks, recently proposed by Fernández-Silva et al. (2011).  
149 The extracts were measured in a UV–Visible spectrophotometer (UVIKON XS, Bio-Tek),  
150 and the equation proposed by Wellburn (1994) was used to calculate the concentration of  
151 chlorophyll *a*, expressed as  $\mu\text{g}$  chlorophyll *a* per  $\text{cm}^{-2}$  surface area.

152 Finally, the thresholds between imperceptible and perceptible greening, in terms of  
153 partial color differences ( $\Delta L^*$ ,  $\Delta a^*$  and  $\Delta b^*$ ), amount of phototrophic biomass (dry weight)  
154 and extracted chlorophyll *a* content, were estimated (Prieto et al. 2006). For this purpose, a  
155 data table was built in which for each sample (1-21), including its measured parameters, the  
156 eight answers from observers were included (1-8). The whole of cases (1-1, 1-2, (...), 1-8, 2-  
157 1, 2-2, (...), 21-8; ie sample-observer) were sorted into two sets, imperceptible and  
158 perceptible, and cumulative percentages were calculated for each group using the following

159 equations where  $n_p$  represents the number of answers that were “yes, ie perceptible”,  $n_i$   
160 represents the number of answers that were “no, ie imperceptible”, and  $z$  represents answer  
161 1, 2, ...,  $n_p$  or  $n_i$ :

162 Cumulative perceptible,  $z = 100 (z/n_p)$ ;

163 Cumulative imperceptible,  $z = 100 - 100 (z/n_i)$

164 The greening thresholds on granite were derived by plotting the cumulative percentages,  
165 cumulative frequency of answers for each qualitative term (imperceptible and perceptible)  
166 versus ordered partial color differences, phototrophic biomass (dry weight) and extracted  
167 chlorophyll *a* content. The intersection of the two sets of data defines the just noticeable  
168 difference (jnd) in greening (Berns 2000).

### 169 ***Monitoring experiment***

170 The progress of recolonization of the granite façade of the Supercomputing Centre of Galicia  
171 (CESGA, [www.cesga.es](http://www.cesga.es)), a centre par excellence in research and high-performance  
172 computing services built in 1993 in Santiago de Compostela (Galicia, NW Spain), was  
173 monitored fortnightly for a period of ten months (289 days, March/2009-January/2010),  
174 using a spectrophotometer for instrumental color measurements. Three target areas (1, 2, 3)  
175 of the south facing wall, each of which was divided in two subareas, upper (A) and lower  
176 (B), were selected and cleaned mechanically with distilled water and a brush to remove the  
177 existing phototrophic colonization (Figure 1). The six study areas (1A, 1B, 2A, 2B, 3A and  
178 3B), each of 21.5 cm x 31.0 cm, do not receive direct sunlight because of the proximity of  
179 the neighboring buildings. They are also very damp due to water run-off. The weather  
180 conditions in Santiago de Compostela during the experiment (according to data from the  
181 Galician regional meteorological office: MeteoGalicia, [www.meteogalicia.es](http://www.meteogalicia.es)) are shown in  
182 Table 1.

183 Following the methodology proposed by Prieto et al. (2010b), a total of 234 readings  
184 were taken fortnightly at random points in the six study areas. A portable reflection  
185 spectrophotometer (CE-XTH) equipped with OptiviewSilver/i QC Basic software, was used

186 under the following conditions: illuminant D65; observer 2° and a 10-mm diameter viewing  
187 area. Color measurements were analyzed by considering the CIELAB color system (CIE  
188 Publication 15–2, 1986), taking into account the mean values of CIELAB coordinates ( $L^*$ ,  
189  $a^*$  and  $b^*$ ). Variations in the three color parameters (partial color differences), lightness-  
190 darkness  $\Delta L^*$ , redness-greenness  $\Delta a^*$  and yellowness-blueness  $\Delta b^*$ , were calculated as  
191 follows:

$$192 \quad \Delta L^* = L^*_t - L^*_0;$$

$$193 \quad \Delta a^* = a^*_t - a^*_0;$$

$$194 \quad \Delta b^* = b^*_t - b^*_0,$$

195 where  $L^*_0, a^*_0, b^*_0$  = mean value of CIELAB color parameter at the study area after cleaning,  
196 and  $L^*_t, a^*_t, b^*_t$  = mean value of CIELAB color parameter at the study area after a period of  
197 exposure (t).

## 198 **Results and Discussion**

199 The laboratory-induced epilithic cyanobacterial biofilms on granite blocks served to emulate  
200 the presence of initial green fouling. Photographs of the granite blocks inoculated for the  
201 psychophysical experiment, along with the data for the amount of phototrophic biomass (dry  
202 weight) and the concentration of chlorophyll *a* extracted are shown in Table 2.

203 On the basis of the visual evaluation, the threshold of human perception between  
204 imperceptible and perceptible greening in terms of the partial color differences ( $\Delta L^*$ ,  $\Delta a^*$   
205 and  $\Delta b^*$ ), biomass (dry weight) and extracted chlorophyll *a* content, were calculated by  
206 plotting the cumulative frequency of answers given by each observer (Figure 2). The visual  
207 perception of the color difference differed with each observer, and the point at which  
208 individuals began to note the greening on the surface due to phototrophic biomass also  
209 differed among the different observers. From the results summarized in Figure 2, it was  
210 concluded that greening became perceptible when colonization resulted in a decrease of 4.37  
211 CIELAB units in parameter  $L^*$ , or when the values of parameters  $a^*$  and  $b^*$  decreased and  
212 increased, respectively, by only 0.41 and 0.59 CIELAB units. In terms of  $\mu\text{g}$  biomass (dry

213 weight) and extracted chlorophyll *a* content, a lower abundance of phototrophic biomass was  
214 required to reach the greening threshold or just noticeable difference (jnd) in greening in  
215 terms of yellowness-blueness variations,  $\Delta b^*$  ( $6.3 \mu\text{g biomass dry weight cm}^{-2}$  and  $(8.43 \pm$   
216  $0.24) \times 10^{-3} \mu\text{g extracted chlorophyll } a \text{ cm}^{-2}$ ) and a larger quantity was required for the other  
217 two color parameters, lightness-darkness,  $\Delta L^*$ , and redness-greenness,  $\Delta a^*$  ( $12.6 \mu\text{g biomass}$   
218  $\text{dry weight cm}^{-2}$  and  $(20.33 \pm 1.60) \times 10^{-3} \mu\text{g extracted chlorophyll } a \text{ cm}^{-2}$ ). This indicates  
219 that of the CIELAB color parameters, parameter  $b^*$  provides the earliest indication of the  
220 presence of phototrophic microorganisms.

221         The thresholds calculated for the partial color differences were used in the outdoor  
222 experiment, in which the rate of greening on building granite was monitored for ten months  
223 after cleaning, by changes in  $L^*$ ,  $a^*$  and  $b^*$ . The results are shown in Figure 3 as a summary  
224 of the variations in  $L^*$ ,  $a^*$  and  $b^*$  values in the six study areas after the exposure period; the  
225 point at which the observers began to note the greening. This corresponds to  $\Delta L^* < -4.37$ ;  
226  $\Delta a^* < -0.41$  and  $\Delta b^* > +0.59$  CIELAB units, and is indicated by a shaded circle in the  
227 figure. Thus, the first bar of the histograms (BC) represents the color difference before and  
228 after cleaning the colonised surface, and was used as a reference. Subsequent bars reflect the  
229 development of the recolonization process, ie the difference between the value on the first  
230 day after cleaning and the subsequent values. In all cases, the trends were all in the same  
231 direction as the value of the difference between the colonized and the clean rock (BC) (Figure  
232 3), thus indicating that the color change was due to recolonization. Thus, during the  
233 recolonization process a decrease in the  $\Delta L^*$  and  $\Delta a^*$  values and an increase in the  $\Delta b^*$  value  
234 were observed, indicating that the façade became darker and more yellow-greenish color.

235         One important aspect of the results obtained is that analysis of the  $\Delta a^*$  and  $\Delta b^*$   
236 parameters enabled identification of the point at which recolonization began, as in both cases  
237 the difference ( $\Delta$ ) remained constant for a certain period of time and then increased gradually.  
238 The moment at which the difference ( $\Delta$ ) began to increase also coincided with the moment  
239 at which the value of the increase changed direction towards the reference value (BC), thus  
240 indicating the start of colonization. Analysis of the values in Figure 3 shows that  
241 recolonization began at between 80 and 129 days (marked with an arrow in the Figure 3).

242 However, analysis of the  $\Delta L^*$  values was not as useful for identifying the moment at which  
243 recolonization began as the variations were more erratic (Figure 3).

244 Comparison of the day on which recolonization began, determined by analysis of the  
245 trends (direction changes towards reference value) in the  $\Delta a^*$  and  $\Delta b^*$  values, and the  
246 moment at which greening is considered perceptible in terms of the  $\Delta a^*$  and  $\Delta b^*$  values (both  
247 indicated in Figure 3), revealed that in all cases, the earliest detection of recolonization was  
248 by analysis of the trends. Thus monitoring the color of granite façades using a portable  
249 spectrophotometer enables early detection of the appearance of greening, even before it is  
250 perceptible to the human eye. As in most cases, the greening threshold was reached several  
251 days after the value at which colonization was considered to begin, building managers  
252 therefore have a period of time in which to decide when cleaning or treatment should be  
253 applied.

254 Another aspect to take into account is that when biological colonization develops, the  
255  $\Delta b^*$  values, which tend to more positive values, underwent a faster change than the  $\Delta a^*$   
256 values, which tend to more negative values. This is consistent with the previous finding that  
257  $\Delta b^*$  provides the earliest indication of perceptible greening due to the presence of  
258 phototrophic microorganisms. In addition, if the magnitude of the change caused by  
259 colonization is taken into account, it was found that, taking into account the 6 plots, parameter  
260  $L^*$  varied on average by  $6.12 \pm 1.78$  CIELAB units,  $a^*$  by  $3.40 \pm 0.24$  CIELAB units and  $b^*$   
261 by  $9.82 \pm 1.42$  CIELAB units. Therefore parameter  $b^*$  varied most and thus contributed most  
262 to the total variation in color ( $\Delta E^*_{ab} = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$ , Wyszecki and Stiles  
263 1982). This is consistent with previous observations for other stone materials such as quartz  
264 (Prieto et al. 2005) and concrete (De Muynck et al. 2009) and shows that parameter  $b^*$  is the  
265 most informative for detecting phototrophic colonization. This finding suggests that the  
266 technique proposed here can be extrapolated to buildings other than those built from granite.

267 For maintenance of granite buildings affected by greening, decisions about the  
268 required frequency of cleaning and biocide treatment should therefore take into account the  
269 time at which greening becomes perceptible, as well as the time at which colonization is  
270 present but not perceived; both parameters are easily determined by use of a portable

271 spectrophotometer. If the cleaning and/or treatment processes are carried out when  
272 colonization exists but is not yet perceptible, it would be easier to establish a target level of  
273 color of the original surface (Doehne and Price 2010). Moreover, cleaning when greening is  
274 marked can led to the use of chemicals or excessive quantities of water, which may also cause  
275 damage (Maxwell 1992).

276         According to Escadeillas et al. (2007), it generally takes a year for stains to appear on  
277 walls, although under favorable growth conditions, development may be extremely rapid. In  
278 practice, and under favorable growth conditions (porous support such as concrete walls), it  
279 takes one year before the first greening is visible and 2–5 years for intense greening to  
280 develop. Thornbush and Viles (2006) showed that biological colonization of stonework by  
281 fungi was evident after just 2 years. Smith et al. (2004) demonstrated the rapid surface  
282 colonization (less than 2 years) of sandstone by algae in exposure trials in Belfast. In the  
283 present case, recolonization (greening) of the granite façade was apparent within a much  
284 shorter time. Recolonization took no more than 129 days (just over 4 months), and greening  
285 due to the phototrophic microorganisms was perceptible after approximately 178 days  
286 (almost 6 months). This rapid biological colonization was undoubtedly favored by the mild  
287 wet climate of Santiago de Compostela, especially the microclimate affecting the most  
288 shaded walls of the buildings, as also occurs in other buildings in the city (Silva et al. 1997).

## 289 **Conclusions**

290 This study demonstrated that measurement of color changes with a portable  
291 spectrophotometer was a reliable method for the early detection and real-time monitoring of  
292 phototrophic growth (greening) on granite façades, even when it is not perceptible to the  
293 human eye. In this sense changes in parameter  $b^*$  were demonstrated to be the most  
294 informative. Thus,  $\Delta b^*$ : + 0.59 CIELAB units, that correspond in this study to 6.3  $\mu\text{g}$  biomass  
295 dry weight  $\text{cm}^{-2}$  and  $(8.43 \pm 0.24) \times 10^{-3}$   $\mu\text{g}$  extracted chlorophyll  $a$   $\text{cm}^{-2}$ , was established as  
296 the greening threshold.

297         Cleaning and/or treatment should be carried out taking into account both the greening  
298 threshold in terms of  $\Delta b^*$  and the moment that the trend in the value of  $\Delta b^*$  changes and  
299 becomes positive ie indicates the occurrence of phototrophic colonization. In this sense,

300 monitoring changes in  $b^*$  of a cleaned granite façade exposed to the natural environment in  
301 Santiago de Compostela (NW Spain) revealed that recolonization by phototrophic  
302 microorganisms (greening) took no more than 129 days (just over 4 months), and greening  
303 was perceptible after approximately 178 days (almost 6 months).

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429

### 430 **Figure captures**

431 Figure 1. Granite façade of the Supercomputing Centre of Galicia (CESGA), showing  
432 extensive phototrophic colonization (greening) developed as streaks following the path of  
433 water run-off. (a) On site measurement of the color on the façade; (b) overview of the façade  
434 with perceptible colored stains (greening) in humid zones; (c) location of the six study areas  
435 before cleaning; (d) location of the six study areas after cleaning. Six study areas: three target  
436 areas (1, 2, 3) each of which was divided in two subareas, upper (A) and lower (B).

437 Figure 2. Results of the psychophysical experiment. Top: lightness-darkness  $\Delta L^*$ , middle:  
438 redness-greenness  $\Delta a^*$ , and bottom: yellowness-blueness  $\Delta b^*$ . Partial color differences  
439 CIELAB units (on the bottom x-axis) and  $\mu\text{g}$  biomass (dry weight) per  $\text{cm}^{-2}$  surface area and  
440  $\mu\text{g}$  extracted chlorophyll *a* per  $\text{cm}^{-2}$  surface area (in italics) (on the top x-axis) plotted against  
441 the cumulative frequency of answers for perceptible and imperceptible qualitative term (%).  
442 The values of the greening threshold in terms of these parameters are shown in a box in the  
443 figures.

444 Figure 3. Results of the monitoring experiment. Color changes (top:  $\Delta L^*$ , middle:  $\Delta a^*$  and  
445 bottom:  $\Delta b^*$ ) during the period of exposure period (289 days) at the six study areas (1A, 1B,  
446 2A, 2B, 3A and 3B, see Figure 1).  $\Delta L^* = L^*_t - L^*_0$ ;  $\Delta a^* = a^*_t - a^*_0$ ;  $\Delta b^* = b^*_t - b^*_0$ , where  
447  $L^*_0, a^*_0, b^*_0$  = mean value of CIELAB color parameter at the study area after cleaning with  
448 water and a brush, and  $L^*_t, a^*_t, b^*_t$  = mean value of CIELAB color parameter at the study  
449 area after a period of exposure (*t*). For comparison, the color change before and after cleaning  
450 is shown as BC (Before Cleaning). The change in the direction of the partial color difference

451 is indicated by an arrow, and the time when colonization becomes perceptible by a shaded  
452 circle ( $\Delta L^* < -4.37$ ;  $\Delta a^* < -0.41$  and  $\Delta b^* > +0.59$  CIELAB units).

453

454 **Table legends**

455 Table 1. Climatic data: temperature, humidity, sunshine duration and daily global radiation,  
456 during the exposure period (March 2009 to January 2010), recorded at the Santiago\_EOAS  
457 meteorological station (Santiago de Compostela) and captured by Meteogalicia.

458 Table 2. Blanco Cristal granite specimens inoculated with mixed cyanobacterial culture and  
459 used in the psychophysical experiment. Scale = 3.5 cm.

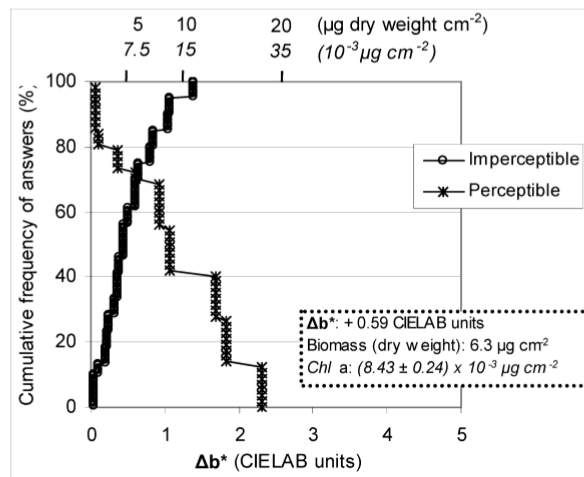
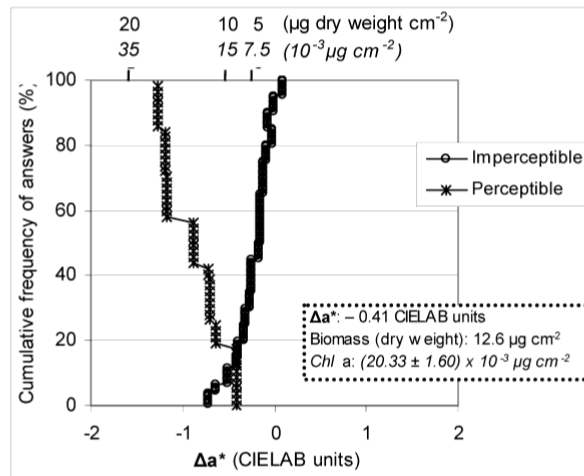
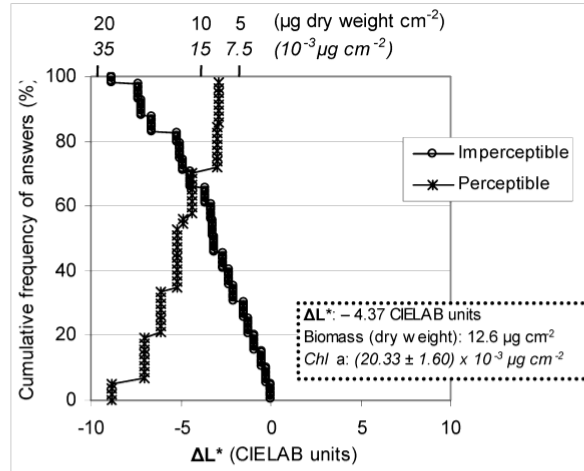
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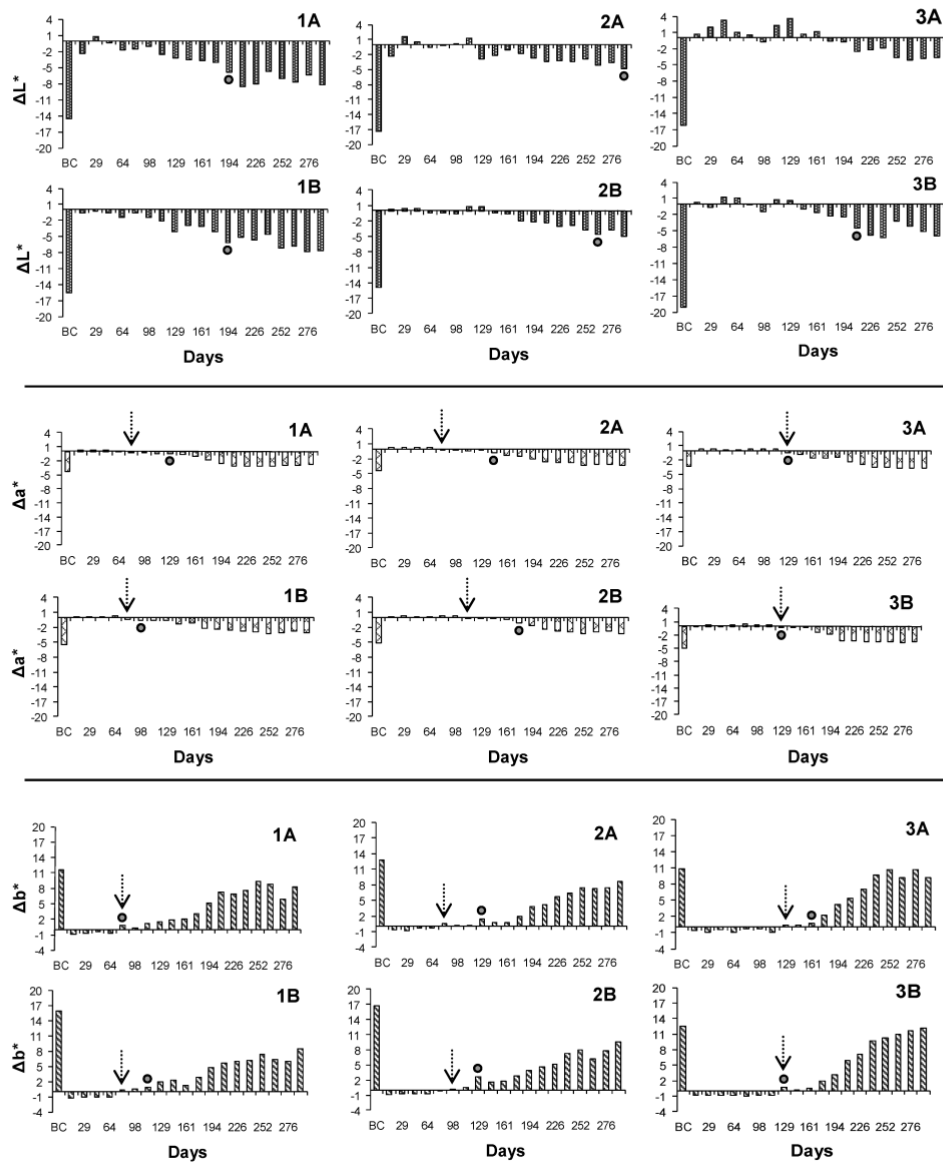
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463 Figure 1



464

465 Figure 2



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467 Figure 3

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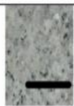









470 **Table 1.**

471

Date	Average air temperature (°C)	Dew point temperature (°C)	Mean relative humidity (%)	Rain (L/m <sup>2</sup> )	Sunshine duration (hours)	Daily global radiation (10KJ/m <sup>2</sup> /day)
March 2009	11.1	4.5	68	30.4	246.6	1617
April 2009	10.3	6.2	79	106.2	177.6	1581
May 2009	14.5	8.9	73	55	241.6	1932
June 2009	17.2	12.9	78	106.6	218.4	1942
July 2009	17.1	13.1	80	105.2	211.4	1917
August 2009	18.8	14.4	78	24.5	236.9	1917
September 2009	18.0	13.5	78	13.5	248.6	1691
October 2009	16.2	13.4	86	189	149.0	935
November 2009	11.7	9.9	92	272.2	60.1	433
December 2009	8.1	5.9	89	424.5	96.6	434
January 2010	7.6	5.6	91	200.2	85.4	489

472

473 **Table 2.**

Photograph of the granite block surface										
Phototrophic biomass ( $\mu\text{g dry weight cm}^{-2}$ )	0.0	0.4	0.7	1.1	1.4	2.1	3.2	6.3	12.6	18.9
Chlorophyll <i>a</i> extracted(*) ( $10^{-3} \mu\text{g cm}^{-2}$ )	0.0	$1.65 \pm 0.12$	$2.49 \pm 0.21$	$4.04 \pm 0.08$	$4.49 \pm 0.20$	$5.74 \pm 0.02$	$4.95 \pm 0.91$	$8.43 \pm 0.24$	$20.33 \pm 1.60$	$31.50 \pm 2.47$

474

(\*) mean value of three replicates  $\pm$  SD.

475