

# Supplementary Materials: Examining the Potential of Enzyme-Based Detergents to Remove Biofouling from Limestone Heritage

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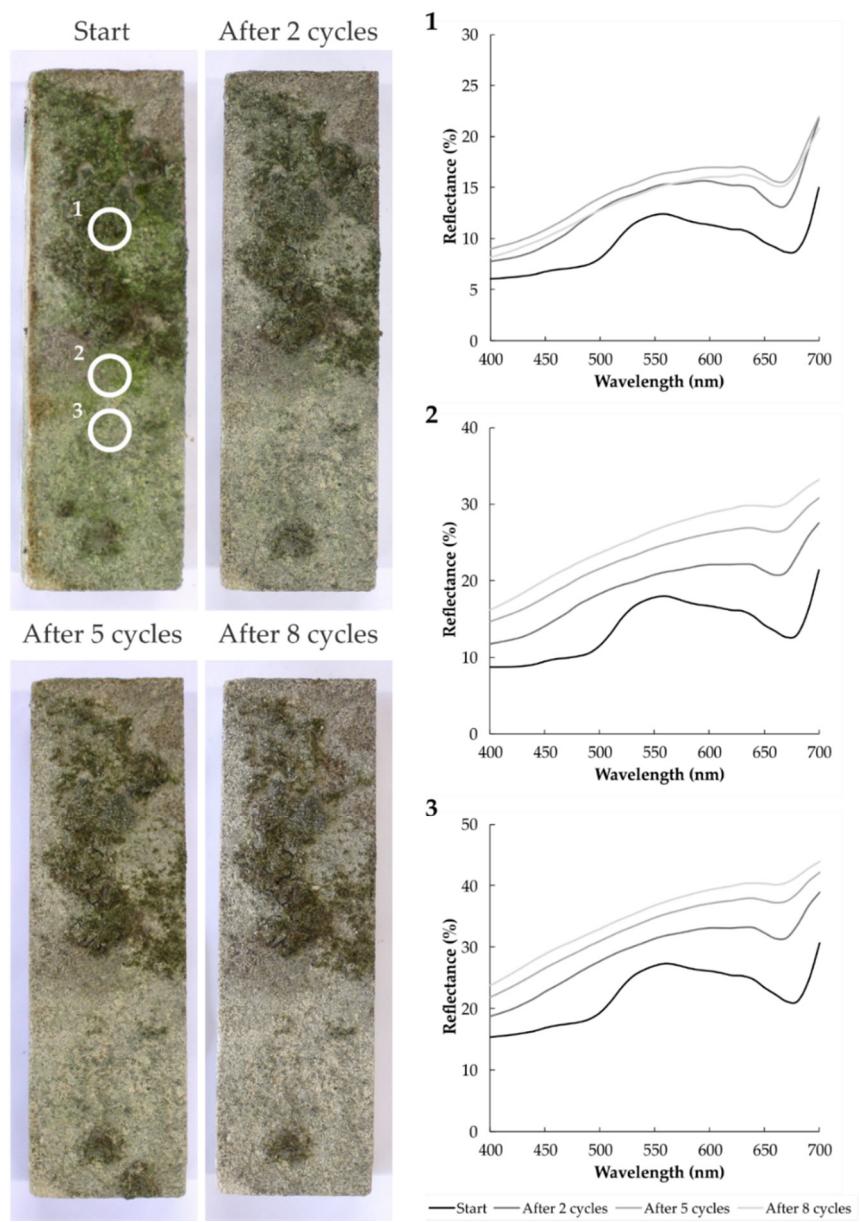
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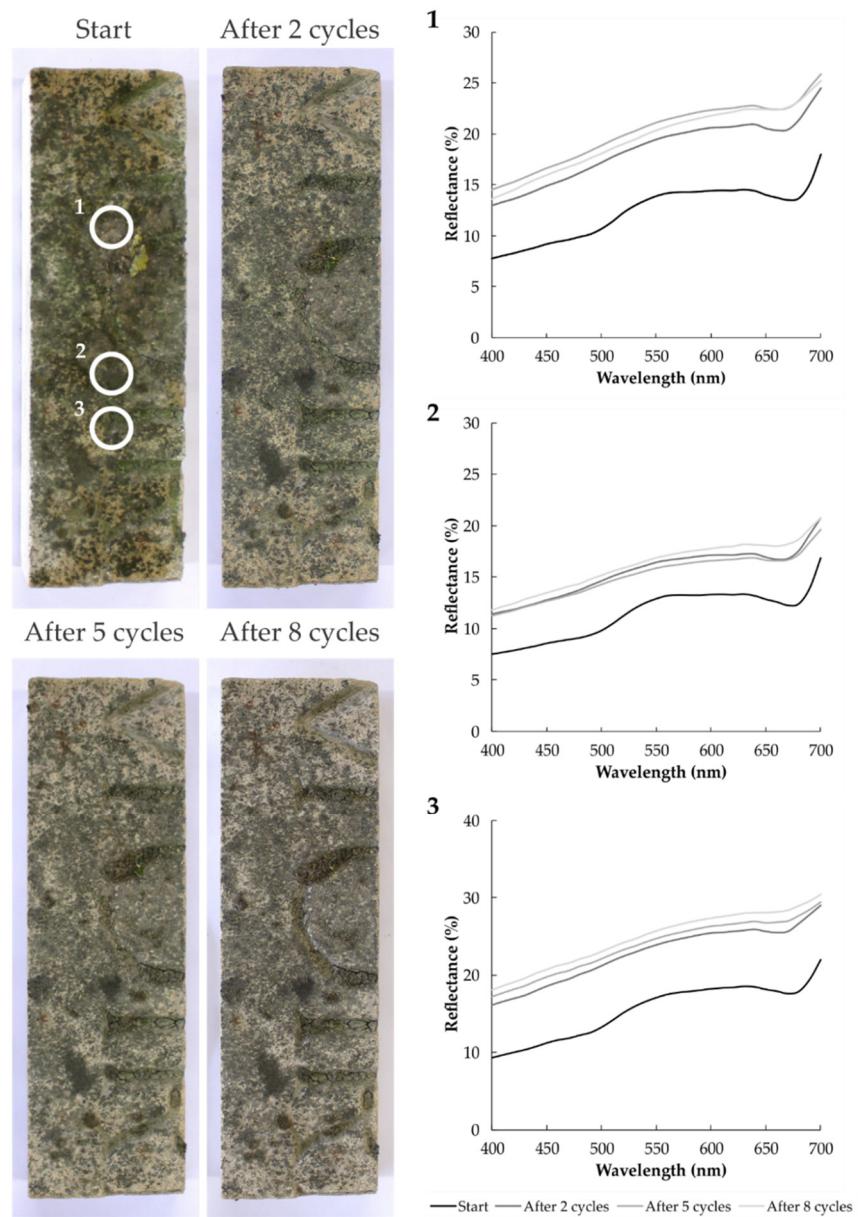
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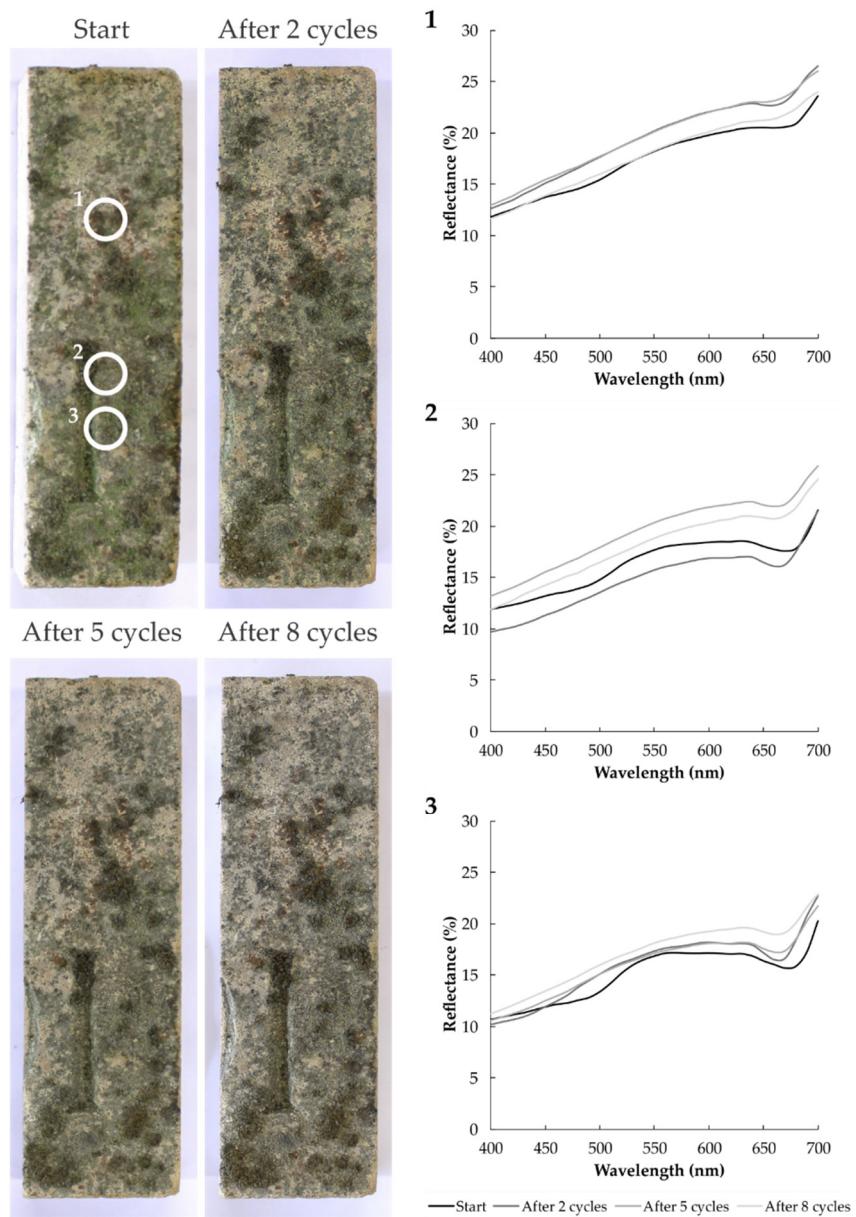
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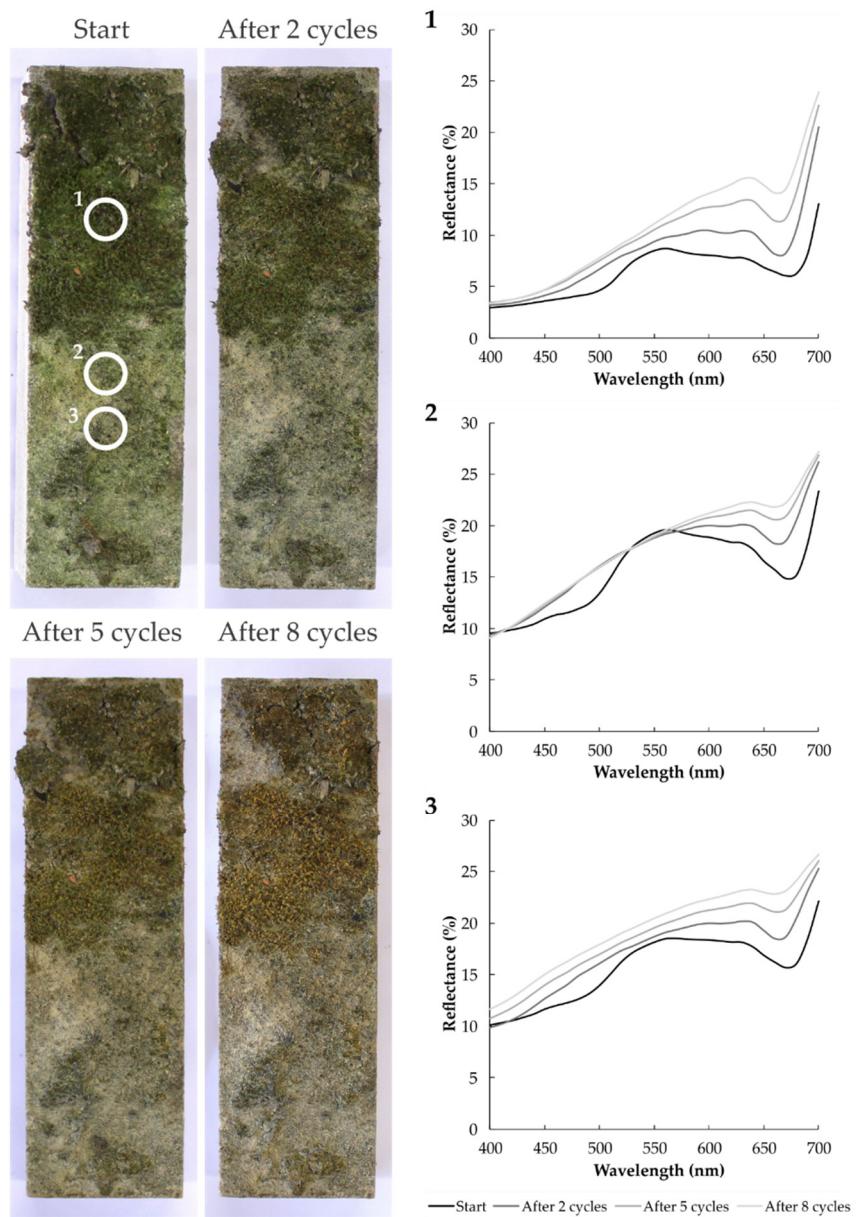
**Figure S1.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of treated Sample 1 from the start and after different cycles of water runoff and drought.



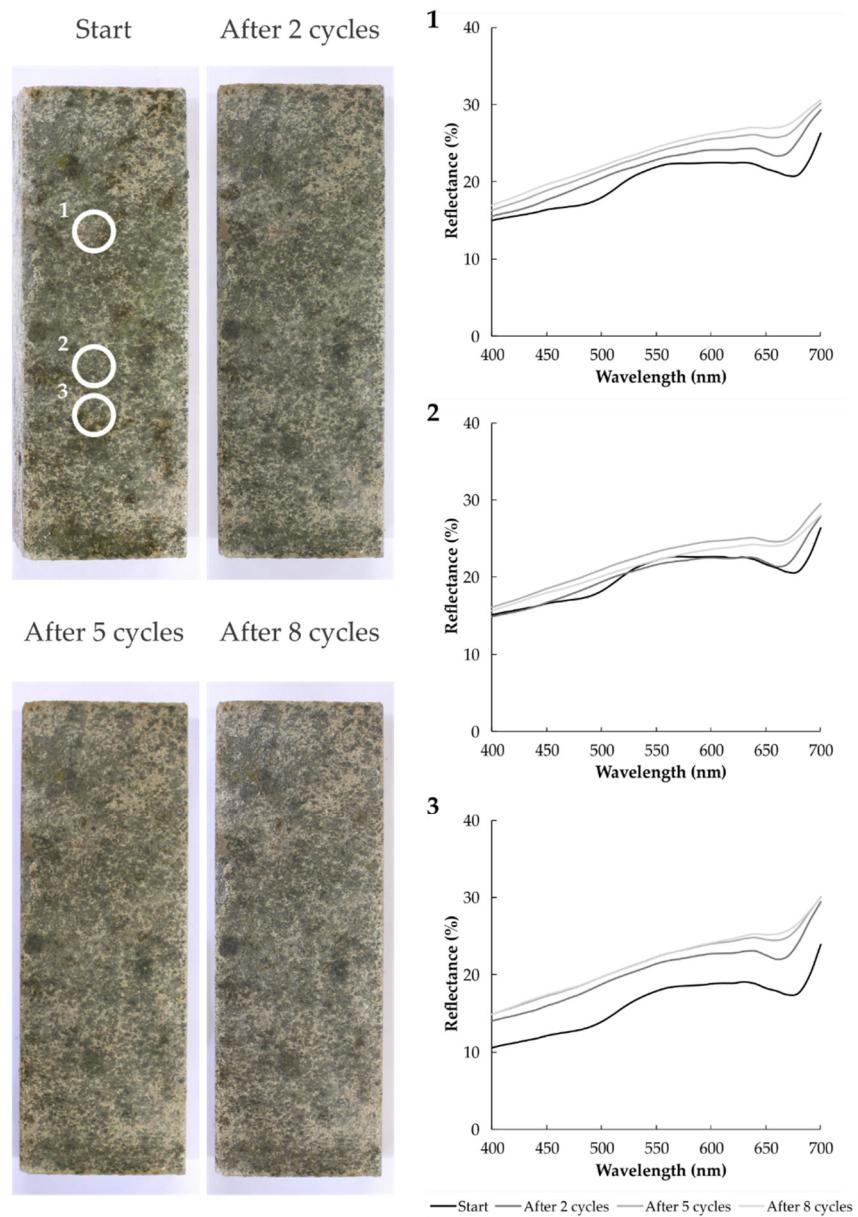
**Figure S2.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of treated Sample 2 from the start and after different cycles of water runoff and drought.



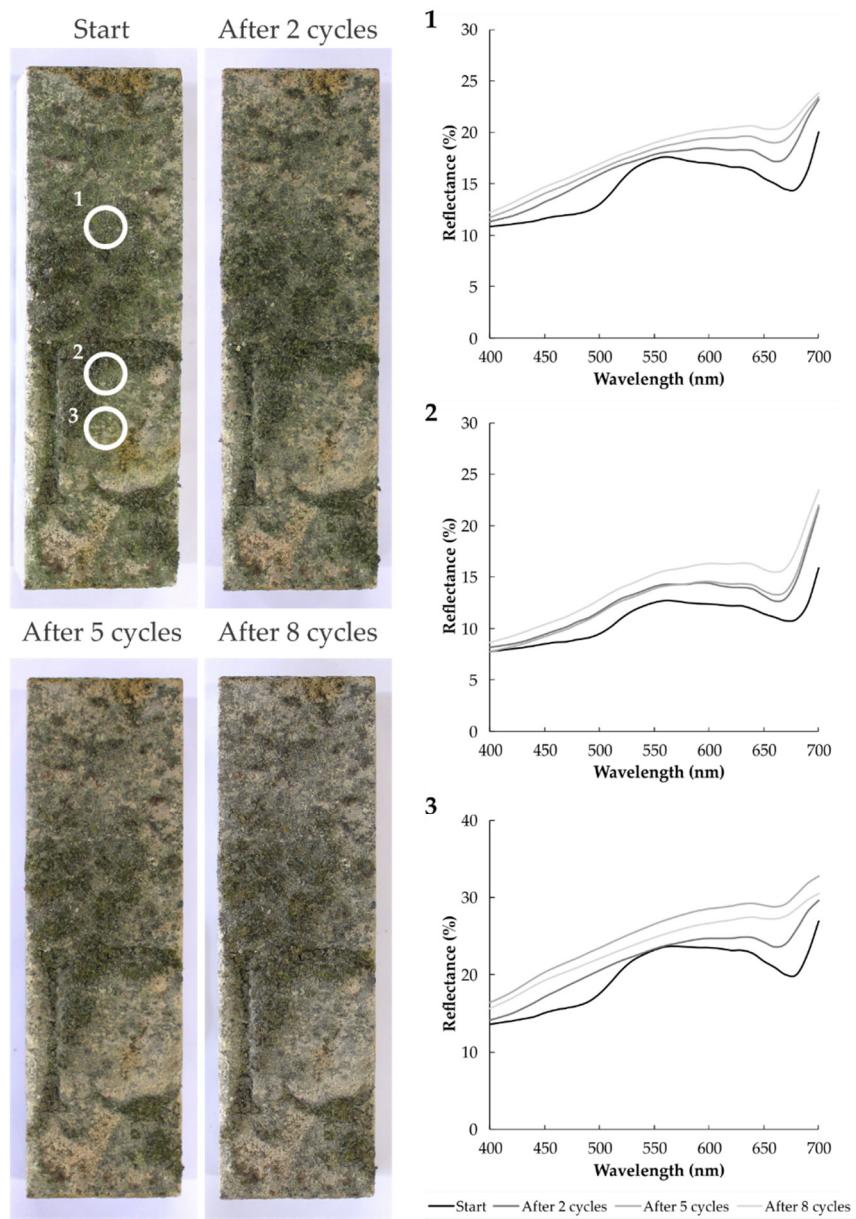
**Figure S3.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of treated Sample 3 from the start and after different cycles of water runoff and drought.



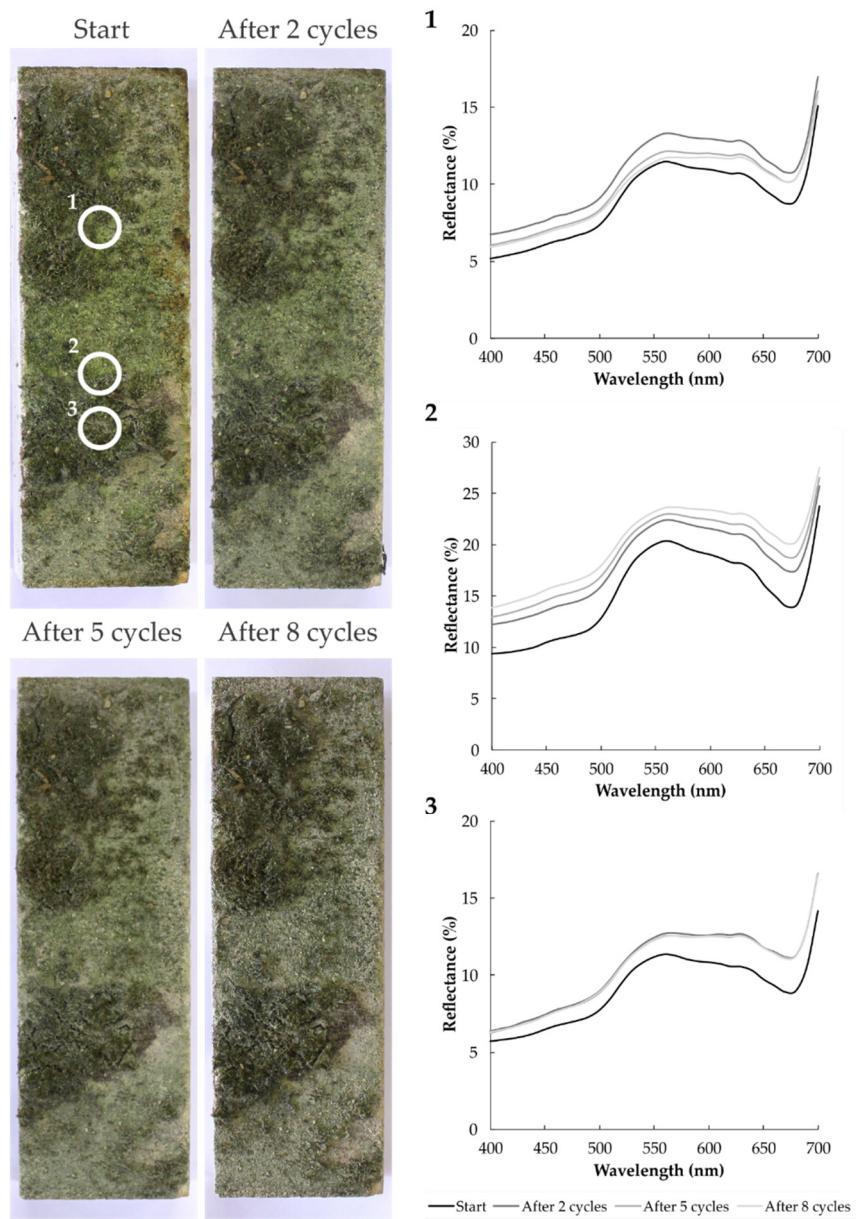
**Figure S4.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of treated Sample 4 from the start and after different cycles of capillary absorption and drought.



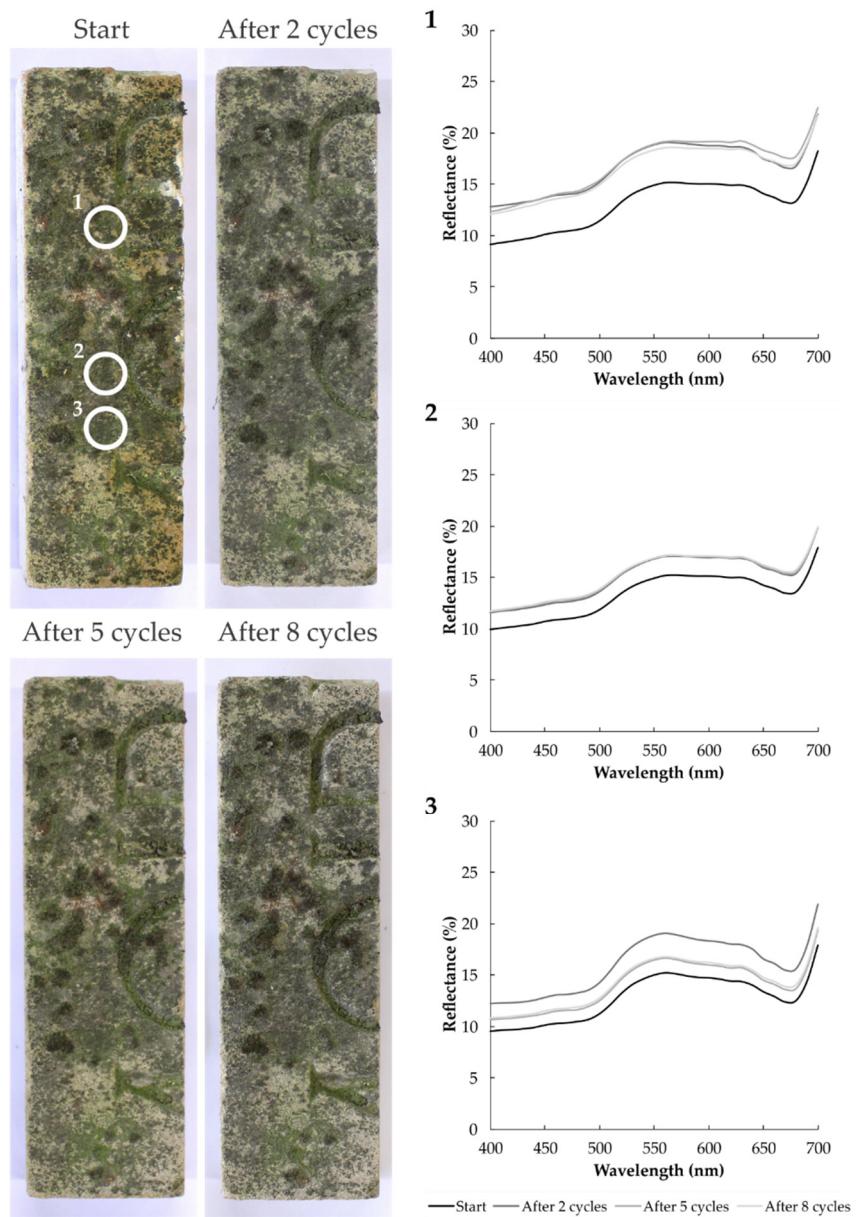
**Figure S5.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of treated Sample 5 from the start and after different cycles of capillary absorption and drought.



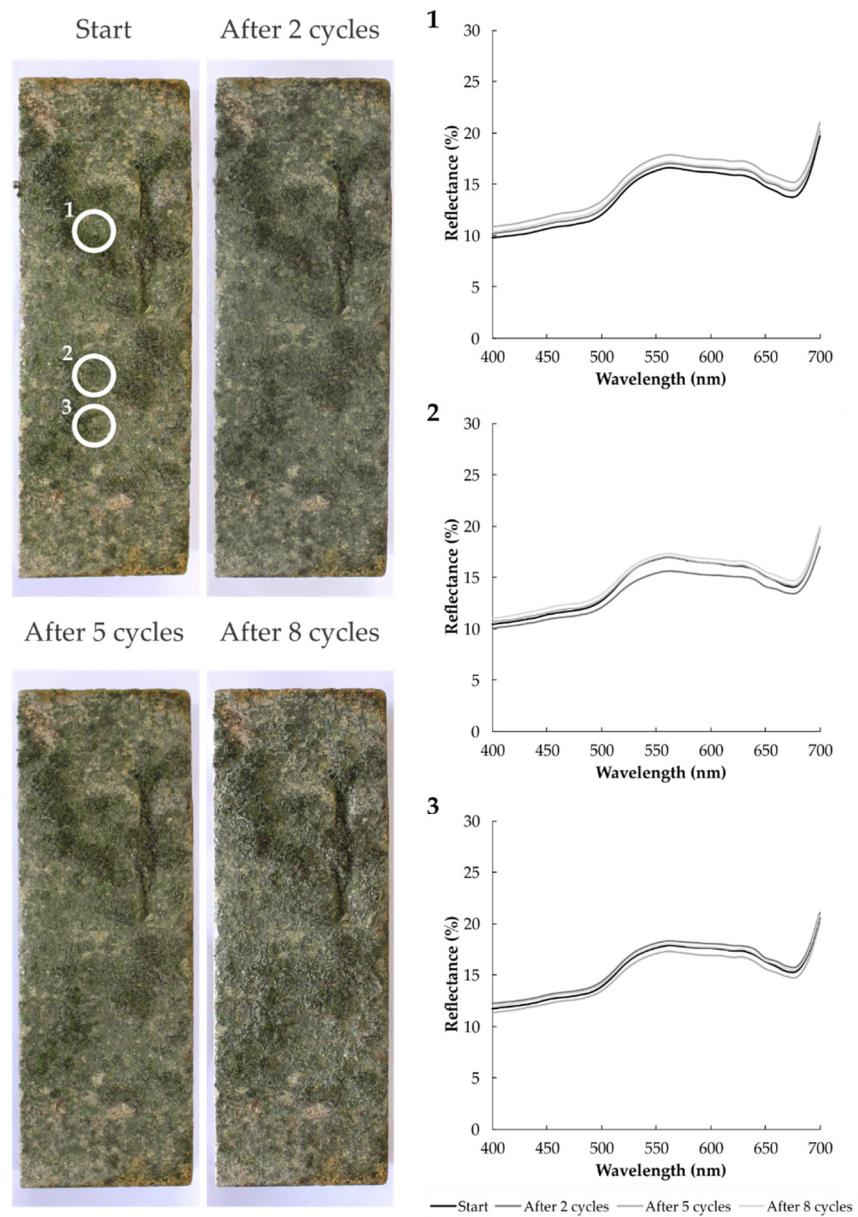
**Figure S6.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of treated Sample 6 from the start and after different cycles of capillary absorption and drought.



**Figure S7.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of untreated Sample 7 from the start and after different cycles of water runoff and drought.



**Figure S8.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of untreated Sample 8 from the start and after different cycles of water runoff and drought.



**Figure S9.** Appearance and spectral reflectance graphs determined at fixed locations 1, 2 and 3 of untreated Sample 9 from the start and after different cycles of capillary absorption and drought.

**Table S1.** Initial CD (%) of 30 measurements at Site A before enzyme-based treatment and  $\Delta$ CD (%) during subsequent field surveys (per date).

Sample	CD					$\Delta$ CD (%)		
	29/04	06/05	13/05	27/05	18/06	05/08	09/09	03/10
1	2.05	-0.49	-6.83	-22.44	-10.24	-5.37	18.05	1.95
2	2.13	-12.21	-11.74	-21.60	-7.04	-22.54	-2.82	-2.35
3	5.61	-34.05	-36.36	-26.56	-40.29	-40.46	-68.63	-19.96
4	5.69	-39.02	-54.48	-12.48	-59.93	-61.51	-72.41	-21.79
5	5.26	-45.63	-55.70	-54.56	-59.32	-61.03	-54.75	-50.57
6	5.87	-51.11	-57.92	-57.41	-65.25	-64.91	-62.69	-58.94
7	2.96	-35.81	-34.12	-40.54	-41.55	-33.11	-25.68	-28.38
8	2.43	-14.40	-25.10	-31.69	-30.04	-28.40	-11.52	-25.10
9	2.80	-18.93	-52.14	-23.21	-33.21	-27.50	-21.43	-13.21
10	5.05	-58.22	-39.41	-41.39	-47.92	-53.47	-65.35	-34.06
11	2.94	-9.18	-25.17	-13.95	-30.27	-28.23	-37.07	6.46
12	2.88	-10.42	-25.00	-20.83	-23.96	-35.76	-42.71	-5.90
13	1.73	2.89	0.58	-19.65	-15.03	-20.81	-4.62	-12.14
14	2.99	-34.78	-44.48	-45.82	-50.17	-47.83	-40.13	-37.46
15	3.91	-37.85	-41.18	-33.76	-51.92	-50.13	-50.90	-37.60
16	4.00	-33.25	-39.00	-31.00	-43.50	-51.75	-53.75	-14.75
17	3.63	-32.23	-39.39	-13.50	-40.22	-34.71	-54.55	-3.03
18	3.58	-34.08	-45.53	-25.14	-48.88	-46.09	-41.06	-45.81
19	2.16	-6.48	-26.85	-25.93	-22.69	-26.39	-15.74	-23.61
20	2.48	-20.16	-20.56	-34.68	-34.68	-25.00	-27.02	-31.85
21	3.70	-34.59	-45.14	-33.78	-40.54	-44.59	-41.35	-23.24
22	3.57	-19.89	-57.14	-30.25	-37.25	-37.54	-48.18	3.08
23	3.28	-7.62	-34.15	-28.05	-24.09	-16.77	-37.50	-10.98
24	4.04	-27.72	-49.01	-29.95	-47.03	-46.53	-43.32	-30.45
25	1.93	10.36	15.54	-9.33	-6.74	0.52	31.09	0.00
26	2.79	-20.07	-51.97	-32.26	-29.03	-20.43	-36.56	-12.90
27	2.31	-30.74	-24.24	-29.44	-10.82	-15.15	-39.83	2.16
28	7.15	-55.94	-57.90	-40.70	-54.41	-60.00	-71.05	-44.06
29	6.71	-46.94	-51.71	-29.21	-52.01	-50.07	-76.60	-30.40
30	4.35	-38.85	-47.82	-46.90	-44.60	-53.10	-48.05	-44.37

**Table S2.** Initial CD (%) of 30 measurements at Site B before enzyme-based treatment and ΔCD (%) during subsequent field surveys (per date).

Sample	CD					ΔCD (%)		
	29/04	06/05	13/05	27/05	18/06	05/08	09/09	03/10
1	1.70	-12.94	-24.12	-28.82	-25.88	-28.82	-20.59	-33.53
2	2.85	-46.32	-56.84	-54.04	-57.54	-57.54	-55.09	-52.98
3	4.97	-67.61	-71.63	-68.61	-64.99	-64.19	-68.81	-68.61
4	5.83	-72.90	-75.81	-71.18	-69.98	-73.41	-75.81	-75.13
5	5.63	-63.94	-67.67	-59.15	-66.61	-68.56	-73.18	-69.98
6	4.17	-58.51	-59.95	-50.84	-58.27	-53.72	-56.83	-57.31
7	5.49	-44.63	-58.47	-58.11	-59.74	-59.56	-60.29	-62.48
8	6.67	-46.18	-61.32	-61.17	-64.92	-63.42	-64.32	-71.81
9	13.91	-72.03	-82.67	-82.96	-83.82	-83.47	-82.24	-83.03
10	13.38	-72.72	-83.71	-83.93	-86.10	-83.26	-84.23	-83.78
11	11.89	-70.23	-79.06	-78.47	-80.40	-83.35	-79.65	-80.15
12	12.38	-73.10	-81.02	-80.94	-81.74	-81.74	-80.78	-80.78
13	2.19	-9.59	-31.51	-30.14	-24.20	-23.74	-24.66	-27.85
14	2.97	-57.91	-65.66	-63.97	-60.94	-61.95	-65.99	-62.29
15	4.27	-48.95	-56.21	-64.87	-61.36	-65.81	-57.14	-64.64
16	10.18	-58.64	-68.17	-70.73	-75.15	-77.31	-75.05	-75.74
17	8.32	-55.65	-67.07	-67.19	-72.60	-72.48	-72.60	-73.68
18	7.94	-62.34	-76.32	-73.43	-70.91	-76.45	-72.54	-73.68
19	2.69	-10.41	-29.00	-15.61	-13.38	-13.01	-13.38	-26.39
20	2.21	-10.41	-38.01	-35.29	-11.76	-10.86	-11.31	-25.34
21	4.43	-55.53	-62.98	-63.43	-63.43	-59.37	-58.01	-59.59
22	5.49	-51.18	-68.67	-64.12	-64.85	-63.93	-63.21	-62.11
23	4.80	-60.21	-72.29	-63.75	-60.83	-63.13	-63.54	-59.17
24	3.29	-34.65	-55.62	-48.02	-44.98	-46.20	-45.29	-42.55
25	7.90	-73.92	-76.20	-78.10	-78.99	-76.08	-81.14	-79.87
26	4.87	-65.09	-74.74	-66.32	-71.46	-68.38	-71.66	-68.17
27	6.04	-57.45	-65.73	-70.36	-69.54	-64.40	-67.55	-68.71
28	3.03	-15.84	-44.22	-45.87	-37.29	-31.35	-31.35	-37.29
29	3.26	-29.75	-50.31	-50.31	-46.01	-43.87	-52.15	-48.47
30	5.15	-57.48	-66.02	-70.10	-64.08	-63.11	-66.02	-64.85

**Table S3.** Initial CD (%) of 30 measurements at Site C before enzyme-based treatment and ΔCD (%) during subsequent field surveys (per date).

Sample	CD					ΔCD (%)		
	29/04	06/05	13/05	27/05	18/06	05/08	09/09	03/10
1	5.89	-65.53	-75.89	-66.72	-66.38	-68.42	-69.78	-60.44
2	4.52	-54.87	-67.04	-61.50	-63.94	-60.18	-63.27	-48.89
3	5.60	-50.00	-66.61	-58.39	-64.64	-63.04	-65.00	-39.11
4	2.58	-31.01	-37.21	-32.17	-40.70	-38.37	-32.56	-8.91
5	4.32	-45.83	-55.32	-48.38	-52.78	-54.40	-53.70	-25.23
6	3.66	-58.47	-59.02	-52.46	-56.56	-54.37	-49.18	-44.26
7	5.84	-67.12	-66.61	-70.21	-68.49	-69.86	-70.89	-60.45
8	5.35	-57.94	-66.92	-64.30	-72.71	-71.03	-68.41	-62.06
9	4.07	-43.24	-63.14	-55.04	-63.64	-54.79	-64.62	-33.91
10	4.23	-46.10	-70.45	-56.97	-56.74	-50.35	-52.96	-19.62
11	3.44	-45.64	-63.66	-52.03	-42.15	-38.95	-54.65	-35.47
12	2.95	-36.61	-48.47	-40.68	-42.37	-38.64	-43.39	-38.31
13	3.85	-56.62	-61.30	-60.00	-60.78	-58.44	-58.70	-45.97
14	10.96	-82.76	-87.68	-85.04	-85.68	-86.50	-85.95	-81.57
15	2.10	-15.71	-66.19	-31.90	-37.14	-37.14	-48.10	-27.14
16	2.42	-14.05	-51.65	-34.71	-43.80	-31.40	-42.98	-12.40
17	2.28	-14.04	-61.40	-35.96	-28.95	-30.70	-44.30	-35.09
18	12.86	-87.09	-91.06	-85.93	-88.10	-89.42	-86.86	-84.84
19	5.14	-49.22	-63.62	-56.42	-3.31	-53.11	-51.36	-47.67
20	4.88	-47.75	-58.20	-49.18	-9.43	-53.07	-51.23	-48.36
21	4.95	-61.21	-70.30	-64.04	-24.44	-58.59	-63.03	-48.69
22	4.17	-59.95	-64.75	-53.96	-15.11	-53.96	-55.64	-55.64
23	3.14	-32.17	-52.87	-53.18	-46.82	-44.59	-43.63	-39.17
24	2.87	-39.37	-47.74	-40.77	-41.11	-36.93	-42.51	-23.34
25	8.98	-72.05	-79.40	-77.39	-77.95	-75.95	-75.28	-68.93
26	6.82	-64.37	-72.87	-72.87	-70.53	-67.74	-65.40	-58.65
27	6.18	-61.97	-73.14	-65.86	-67.15	-63.27	-66.02	-64.08
28	3.11	-41.80	-66.88	-47.91	-54.34	-48.87	-51.45	-50.80
29	4.88	-57.99	-71.93	-67.21	-63.73	-58.61	-62.70	-53.89
30	3.32	-49.70	-59.04	-57.23	-64.76	-63.55	-67.47	-58.73

**Table S4.** Initial CD (%) of 30 measurements at Site D before enzyme-based treatment and  $\Delta$ CD (%) during subsequent field surveys (per date).

Sample	CD					$\Delta$ CD (%)		
	29/04	06/05	13/05	27/05	18/06	05/08	09/09	03/10
1	4.19	-51.79	-60.14	-55.13	-54.18	-54.89	-58.71	-12.17
2	4.58	-51.09	-54.15	-51.75	-53.06	-60.26	-64.63	-53.71
3	11.47	-56.41	-72.97	-75.41	-75.33	-73.58	-66.00	-31.91
4	10.26	-53.51	-72.90	-73.29	-76.71	-73.59	-71.35	-48.54
5	9.67	-38.78	-70.32	-72.70	-73.73	-69.80	-71.46	-52.74
6	10.86	-51.75	-74.68	-75.14	-75.41	-72.65	-71.27	-55.06
7	3.61	-37.40	-37.12	-43.77	-48.48	-45.98	-39.34	-10.25
8	3.57	-34.73	-36.41	-71.15	-42.30	-38.38	-33.61	-11.48
9	5.36	-47.76	-54.66	-56.72	-54.48	-53.36	-53.17	-36.57
10	5.59	-54.38	-58.14	-60.29	-58.14	-56.53	-56.71	-43.83
11	5.49	-51.91	-51.73	-54.10	-52.64	-50.82	-41.53	-23.13
12	6.86	-38.48	-54.52	-52.04	-50.00	-59.48	-42.71	-2.33
13	3.76	-54.26	-50.00	-47.07	-50.00	-48.40	-37.77	13.56
14	3.65	-46.30	-44.38	-40.27	-42.74	-45.48	-45.48	-41.37
15	5.54	-59.21	-57.94	-60.83	-57.22	-58.84	-41.70	9.57
16	5.61	-55.97	-57.58	-56.86	-58.11	-55.44	-39.22	-13.37
17	4.58	-48.25	-47.38	-52.18	-47.16	-41.92	-26.42	0.00
18	6.58	-58.51	-60.79	-63.83	-62.61	-61.09	-54.56	-2.43
19	5.24	-50.76	-46.76	-49.05	-56.30	-58.40	-52.29	-23.66
20	6.53	-51.61	-51.15	-58.35	-57.27	-56.81	-60.64	-48.24
21	7.32	-49.73	-61.34	-60.79	-61.61	-65.16	-62.02	-40.16
22	8.03	-51.56	-64.13	-62.52	-62.89	-66.13	-64.38	-50.93
23	7.41	-52.36	-58.57	-61.27	-60.73	-59.38	-52.90	-39.14
24	7.90	-49.62	-61.90	-65.19	-62.03	-64.30	-57.34	-43.04
25	5.21	-51.44	-69.87	-62.00	-54.89	-59.69	-50.10	-62.57
26	4.05	-46.67	-54.07	-46.67	-42.96	-46.42	-40.25	-37.78
27	2.72	-23.16	-38.97	-23.90	-30.88	-23.16	-24.63	1.10
28	5.93	-56.49	-59.87	-55.14	-58.35	-54.47	-54.64	-43.68
29	5.03	-41.75	-51.29	-42.94	-47.32	-44.93	-33.80	16.70
30	4.39	-51.03	-56.95	-50.11	-48.97	-41.69	-41.46	-27.56