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# Partial replacement of cement for waste aggregates in concrete coastal and marine infrastructure: A foundation for ecological enhancement?

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1 **Partial replacement of cement for waste aggregates in concrete coastal and**  
2 **marine infrastructure: a foundation for ecological enhancement?**

3

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13 **Abstract**

14 The effects of climate change and an expanding human population are driving the need for  
15 the expansion of coastal and marine infrastructure (CMI), the development of which is  
16 introducing hard substrate into the marine environment on a previously unseen scale. Whilst  
17 the majority of previous research has focussed on how physical features affect intertidal  
18 macrobiotic communities, this study considered the effects of differences in the chemical  
19 composition of concrete on subtidal biofilm and macrobiotic communities. Two commonly  
20 used cement replacements, pulverised fly ash (PFA) and ground granulated blast-furnace  
21 slag (GGBS), were used in a combination of proportions to assess how concrete tiles with  
22 differing surface chemistries affect development of early successional stages of marine  
23 biofouling communities. Controlled leaching experiments showed that although total metal  
24 leaching varied considerably between tile type, tiles containing GGBS resulted in statistically  
25 lower amounts of metal released compared with tiles containing PFA. Concrete treatment  
26 had no effect on the percentage cover or richness of diatoms, but there were significant  
27 increases in both over the duration of the experiment. Concrete treatments containing GGBS  
28 had a lower richness of native macro-fouling species compared to the control, but there was  
29 no significant difference in non-native species richness among treatments. Results suggest

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30 that different components can be used to alter the surface chemistry of concrete to further  
31 enhance the ecological value of CMI more than physical features can achieve alone.

32

33 **Key words: Marine, concrete, ecology, biodiversity, water quality, elements**

## 34 **1. Introduction**

### 35 *1.1 Expansion of coastal and marine infrastructure (CMI)*

36 With the pressures of climate change and many of the world's population living on or near  
37 the coast (Small and Nicholls, 2003), there is growing demand for development of the  
38 marine and coastal environment (Nicholls and Kebede, 2012). Furthermore, with the  
39 introduction of artificial hard substrata into the marine environment on a large scale (*sensu*  
40 "ocean sprawl"), there is a pressing need to determine the effects that this is having on the  
41 biofouling communities that colonise them, their ecology and impacts on the wider marine  
42 environment. The recent surge of interest in the ecology of the built environment has yielded  
43 a wealth of research describing the impacts of these structures on the receiving environment  
44 (Dafforn et al. 2015, Bishop et al. 2017), the fundamental differences in both the structure  
45 and functioning of artificial habitats compared to their natural analogues (see Firth et al.  
46 2016a for review) and changing attitudes of humans to hard artificial structures (Evans et al.  
47 2017; Morris et al. 2015; Scyphers et al. 2015). There is a growing consensus that artificial  
48 structures are characterised by lower diversity and abundance of native species (Aguilera et  
49 al. 2014; Burt et al. 2009; Chapman, 2006; Firth et al. 2013) and are known to support high  
50 diversity and abundance of non-native and opportunistic species (e.g. Bulleri and Airoidi,  
51 2005; Firth et al. 2011, 2015; Mineur et al. 2012).

52  
53 The nature of the material used in CMI can influence the ecological attributes (e.g.  
54 biodiversity, community composition) of organisms that settle on it. This is largely due to  
55 variations in habitat heterogeneity at a range of spatial scales (Anderson and Underwood,  
56 1994; Chapman and Underwood, 2011; Connell, 2000; Coombes et al. 2015; Firth et al.  
57 2013; Harlin and Lindbergh, 1977; Moschella et al. 2005), but is also linked to chemical cues  
58 (Anderson, 1996; Neo et al. 2009) and even colour (Pomerat and Weiss, 1946; Satheesh  
59 and Wesley, 2010). Because artificial environments generally have lower native species  
60 richness and are more likely to be dominated by opportunistic or non-native species, there  
61 are opportunities to enhance CMI for conservation purposes and ecosystem services  
62 (Chapman and Underwood, 2011; Dafforn et al. 2015; Firth et al. 2016a; Seaman, 2007). To  
63 date, the majority of research has focussed on the physical properties of artificial structures,  
64 but knowledge gaps exist of other factors that could be driving the differences between the

65 ecology of natural and artificial marine environments; for instance, substrate surface  
66 chemistry (but see Nandakumar et al. 2003; Perkol-Finkel and Sella, 2015; Sella and Perkol-  
67 Finkel, 2015).

68

## 69 1.2 *Replacing cement with waste aggregates in coastal and marine infrastructure*

70 When hard substrata, artificial or otherwise, are introduced into the marine environment,  
71 biofilms (formed of benthic microalgae, bacteria and other micro-organisms embedded in a  
72 matrix of extracellular polymeric substances (EPS)) are the first colonisers. Previous  
73 research has shown that biofilm succession and species composition can be affected by  
74 fine-scale substratum roughness (Sweat and Johnson, 2013), environmental pollution (Sanz-  
75 Lazaro et al. 2015) and surface chemistry (Nandakumar et al. 2003). Marine biofilms are  
76 known to interact directly with macro-fouling organisms (Salta et al. 2013) and differences in  
77 biofilm community structure may influence their attachment (Ank et al. 2009).

78

79 Portland cement is the major construction material used globally in artificial structures, often  
80 making up over half of coastal and marine developments (Kampa and Laaser, 2009).

81 Although biotic communities can and do colonise concrete substrates (e.g. Firth et al. 2016b;  
82 Griffin et al. 2010), concrete is considered a poor substrate material for biotic recruitment  
83 due to its high surface alkalinity (pH ~13) and the fact that it can contain toxic metals, which  
84 can interfere with larval settlement (Nandakumar et al. 2003), and affect the emergent  
85 community structure and its functioning (Perkol-Finkel and Sella, 2015; Sella and Perkol-  
86 Finkel, 2015). Typically, concrete has four main ingredients: coarse aggregate (e.g. gravel),  
87 fine aggregate (usually sand), cement and water; although its properties can be amended to  
88 improve strength or resistance to sulphate and chloride attack (Snelson and Kinuthia, 2010).  
89 This can be achieved by changing the types and proportions of ingredients, as well as by  
90 using additional ingredients such as silica fume, pulverised fly ash (PFA), ground granulated  
91 blast-furnace slag (GGBS), carbon fibres (Graham et al. 2013) and hemp fibres (Dennis et  
92 al. in press). Not only do these materials change the physical properties of concrete, but they  
93 can also inherently change its chemical composition. Cement production is extremely  
94 energetically expensive, accounting for 8% of global CO<sub>2</sub> emissions (Achterbosch et al.  
95 2011). There is therefore an incentive to use cement replacements, not only as it reduces  
96 the carbon footprint of the end product (concrete), but also uses waste products that may  
97 otherwise go to landfill (Bignozzi, 2011).

98

99 PFA is a waste product from burning coal, removed from flue gases by electrostatic  
100 precipitators. It is well reported that the addition of fly ash into cement mixes can greatly  
101 improve durability, resistance to sulphate attack and chloride penetration, and reduce the  
102 likelihood of leaching effects (e.g. Chalee et al. 2010; Thomas and Matthews, 2004). GGBS  
103 is a by-product of the steel industry made by grinding iron slag into a fine powder, which  
104 when used as a (partial) replacement for cement, can provide resistance to sulphate attack  
105 without any loss of durability or compressive strength (Pavia and Condren, 2008). The use of  
106 PFA and GGBS as replacements for cement in concrete is well established. GGBS can be  
107 used as a direct replacement for Portland cement, on a one-to-one basis by weight, with  
108 replacement levels of between 30% and 85% reported. Typically, 40 to 50% replacement is  
109 most common in order to maintain structural integrity. PFA replaces a certain percentage of  
110 Portland cement, usually between 6 to 35% according to British standard EN 197-1: 2011  
111 (British Standards Institute, 2011). Both replacements are primarily composed of calcium  
112 oxide, and silicate and aluminate glass spheres. As such, they are highly alkaline (pH > 9)  
113 and contain relatively high concentrations of trace metals (Table 2, Mullauer et al. 2015)  
114 such that while the use of waste products in concrete is positive from a sustainability point of  
115 view, these concretes may not be as environmentally sound when it comes to enhancing  
116 ecological value in terms of marine conservation, biodiversity and ecosystem services.  
117

### 118 1.3 *Chemical leaching from coastal and marine infrastructure*

119 Previous studies have shown that metal leaching can occur at the outer layer of the concrete  
120 during the first 30 days of immersion (Shi and Kan, 2009), but some metals can leach in  
121 greater concentrations from PFA and GGBS over longer time periods (Jang et al. 2015). The  
122 general consensus in the literature appears to be that metals within cement and fly ash can  
123 leach out of concrete, albeit at relatively low concentrations owing to cement hydrates  
124 immobilising the majority of the metals, particularly in the core, with only the outer surface  
125 leading to the dissolution of metals into the aqueous phase. Despite this evidence, it is  
126 apparent that leaching of metals still occurs, and in some cases, exceeds the standards set  
127 for protecting marine species and the environment.

128

129 The majority of previous research on cement replacements has focused on the chemical  
130 properties of cement replacements, but few studies have investigated the ecological effects

131 (Izquierdo et al. 2009; Jang et al. 2015; Müllauer et al. 2015). Here, the leaching of metals  
132 and the effects of the chemical nature of concrete on colonising communities were  
133 investigated using two cement replacements (PFA and GGBS). This research seeks to open  
134 up new opportunities for research into alternative cement replacements and their use in  
135 enhancing the ecological value of biofouling communities. Using a combination of laboratory  
136 and field experiments, we aimed to determine: (1) the metal composition of the concretes  
137 and its variability; (2) the significance of the leaching of metals from cement replacements  
138 into seawater; (3) the impacts of cement replacements on colonising biofilms in terms of  
139 species richness and cover; (4) the effects of cement replacements on colonising macro-  
140 biota, with particular reference to differential responses of native and non-native species.

141

## 142 2. *Materials and Methods*

### 143 2.1 *Study site*

144 Fieldwork was carried out at the Plymouth University Marine Station at Queen Anne's  
145 Battery marina in Plymouth Sound at the mouth of the Tamar Estuary (50.3648° N, 4.1298°  
146 W). Queen Anne's Battery is a sheltered marina located in a busy shipping area for  
147 recreational boating, commercial fishing as well as international commercial transport of fuel  
148 and bulk aggregates. The colonising communities on the marina pontoons (floating docks)  
149 were characterised by ascidians, bryozoans and macro-algae, including a high abundance of  
150 non-native species (Arenas et al. 2006; Bishop et al. 2013). Due to its importance as an  
151 international shipping port with a long history of marine biological research, Plymouth Sound  
152 is well known for being the site of many first records of non-native species (Knights et al.  
153 2016). Salinities over the course of the sampling period (summer) vary between 28 and 30  
154 ppt, pH between 8.0 and 8.4, and temperature between 16 and 18°C (Langston et al. 2003).  
155 These conditions are typical of temperate coastal waters of equivalent northern or southern  
156 latitudes, which suggests wide applicability of the data to other sites worldwide.

### 157 2.2 *Experimental design and concrete production*

158 Our goal was to devise an experiment that provided the opportunity to observe potential  
159 differences in chemical leaching and species recruitment between concrete mixtures, whilst  
160 being realistic in terms of the mixtures used commercially in construction, and therefore the  
161 physical integrity required for their engineering application. As we wished to achieve

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162 relatively high replication for the leaching trials, the number of concrete mixtures used was  
163 limited to four (Table 1), but with recognition that many more cement replacement  
164 combinations could be tested.

165  
166 In this experiment a replacement of 24% cement for GGBS/PFA (direct replacement-by-  
167 weight) was chosen. While a relatively low level of replacement for GGBS, this is toward the  
168 middle to higher-end level of replacement for PFA. Given that one of the mixtures contains  
169 both PFA and GGBS, testing of any chemical interactions led to a replacement of 48%  
170 cement by PFA and GGBS, but still within the range of percent replacement by either GGBS  
171 or PFA used in industry (see above). Using the replacement percentages meant that the  
172 metal composition of the base components would be sufficiently differentiated to allow the  
173 analytical methodologies to determine any differences in leaching behaviours between  
174 concrete types.

175  
176 Concrete tiles were constructed using four different mixtures. We refer to the different  
177 mixtures as 'types'. Standard 'control' concrete was made using a mixture of standard high  
178 strength cement (CEM-1), sand, crushed granite (4-10 mm) and water. The three other  
179 concrete types had cement partially replaced by pulverised fly ash (PFA), ground granulated  
180 blast slag (GGBS), or a mixture of both PFA and GGBS (Mixed; see Table 1 for details).

181  
182 Tiles of two different sizes (25 x 25 x 5 cm; and 2 x 2 x 1.5 cm) were cast in silicone trays to  
183 aid mould release and to negate the use of a release agent, then covered in a polyethylene  
184 sheet and allowed to cure for 2-wk (Zemajtis, 2016). The larger-sized tiles were used for the  
185 chemical analyses whilst the smaller tiles were used for biological analyses. The smaller size  
186 was specifically chosen to allow tiles to be placed in an electron microscope for biofilm  
187 analysis.

188

189 Table 1. Description of concrete treatment compositions. Percentage of ingredients used to  
190 create the different concrete types expressed as a percentage by-weight. Note that 4%  
191 PFA/GGBS corresponds to by-weight ~24% (4/17) replacement of cement.

<b>Concrete Type</b>	<b>Cement</b>	<b>Sand</b>	<b>Crushed Granite</b>	<b>Water</b>	<b>PFA</b>	<b>GGBS</b>
Control	17%	27%	47%	9%	0%	0%

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PFA	13%	27%	47%	9%	4%	0%
GGBS	13%	27%	47%	9%	0%	4%
Mixed	9%	27%	47%	9%	4%	4%

192

### 193 2.3 Chemical testing

194 To minimise the possibility of contamination of the materials and samples with trace metals,  
195 all equipment used for collecting samples was washed in 10% nitric acid and rinsed 3-5  
196 times with deionised water (Milli-Q; >18 MΩ.cm, Merck Millipore, USA) prior to use.

197

#### 198 2.3.1 Media Aqua Regia digestion of the concrete components

199 In order to determine the metal content of the base components of the concrete mixes used,  
200 samples (5g) of the individual concrete components (GGBS, PFA, granite, sand, cement)  
201 were placed into 250ml glass beakers. Owing to its size (4-10mm), the aggregate was  
202 ground down into a powder using a mortar and pestle. 70ml of nitric acid (Sigma-Aldrich  
203 S.G.) and 30ml of hydrochloric acid (Sigma-Aldrich S.G 1.18) was added and left to stand for  
204 60 min. Samples that had not fully dissolved had 5ml Aqua Regia solution added and were  
205 placed on a hot plate at 90°C and allowed to simmer for 60 min until fully dissolved. The  
206 solution was then filtered through a Whatman cellulose acetate membrane into volumetric  
207 flasks and made up to a known volume using 2% nitric acid.

208

#### 209 2.3.2 ICP-MS determination

210 All acid digested samples were determined for elemental composition using a Thermo  
211 Scientific X-Series 2 Inductively Coupled Plasma – Mass Spectrometer (ICP-MS). Standards  
212 for seawater analysis were made using CPI International multi-element quality control  
213 standard (P/N 4400-013) and Romil Mercury element reference solution, diluted to working  
214 ranges using Milli-Q water. Control samples and calibrations were run at the beginning and  
215 end of each batch of analysis and filter and acid blanks determined to check for sample  
216 contamination. Recovery was compared against independent control samples and were 107  
217 ±10% for Cr, 106 ±8% for Zn, 105 ±11% for Mo and 105 ±10% for Ba. Limits of detection  
218 based on using 3 times the standard deviation of the blank were 0.01, 0.09, 0.25, 0.11, 0.16,  
219 0.16 and 0.01 µg/l, 0.16 for Cr, Cu, Zn, Se, Mo, Ba and Pb respectively.

220

#### 221 2.3.3 Chemical leaching tests

222 It was assumed that there was a certain degree of heterogeneity on the surface of each type  
223 of tile exposed to seawater for any given concrete mix. Consequently, all tests were  
224 undertaken using five replicates for each treatment. Individual large concrete tiles were fully  
225 submerged in tanks of seawater and 125ml water samples were collected at fixed time  
226 points from 5 cm depth at the centre of the well-mixed tank. Water samples were stored in  
227 HDPE bottles (Nalgene). After 18 h immersion, the seawater in each tank was replaced with  
228 fresh seawater to ensure leaching rates were not affected by increased chemical  
229 concentrations within a tank that might result in reduced leaching due to a change in  
230 concentration gradient. In total, tiles were immersed for 1296 h (54 days). The cumulative  
231 chemical concentration leached into the water was calculated over time to: (i) determine total  
232 chemical concentration leached from each tile, and (ii) to identify the point of asymptote, i.e.  
233 when no further chemicals were leached from the tiles. At each time interval, measurements  
234 of pH, salinity and temperature were also taken using a YSI 63-25 multi-parameter probe.

235  
236 All water samples were filtered through a 0.45µm polycarbonate membrane previously  
237 cleaned with 30% hydrochloric acid for 24 hours and rinsed 3-5 times with Milli-Q water  
238 (Merck Millipore, USA). After filtration, 1ml of seawater was decanted into a 15ml  
239 polyethylene centrifuge tube and 9 ml of 2% nitric acid added as a matrix diluent and  
240 preservative. Chemical concentrations were then determined using ICP-MS determination  
241 techniques as outlined above.

242

#### 243 2.4 *Study of colonising biota*

244 To study the growth of biofouling assemblages on the submerged tiles, small concrete tiles  
245 were attached to wooden beams using cable ties and hung from pontoons at a depth of  
246 0.5m for 49 days (28/6/16 to 16/8/16). Tiles were arranged randomly and orientated so that  
247 the front faces of the tiles were vertical.

##### 248 2.4.1. *Assessment of colonising biofilms*

249 Colonising biofilms were assessed by collecting fifteen replicate samples from each of the  
250 four treatments (i.e. concrete types) at weekly intervals over 4 weeks (05/07/2016 to  
251 26/07/2016). Each sample was fixed and preserved in 5% glutaraldehyde solution, using  
252 seawater as a buffer, and kept refrigerated. Samples were washed and then air-dried for 7  
253 days before imaging using low-vacuum scanning electron microscopy (LV-SEM; JEOL

254 6110). LV-SEM negates the need for sample preparation by spatter-coating; a process  
255 required for high-vacuum SEM. From these images, all diatoms present were identified to  
256 genus and percentage cover estimated using a 10 x 10 virtual grid quadrat (500 x 500 µm).  
257 Here, given the expertise available, we focused on diatoms but acknowledge that future  
258 studies should consider all microorganisms in the biofilm.

259

#### 260 *2.4.2. Assessment of colonising macro-fouling communities*

261 Colonising macro-fouling communities were determined using five replicates collected from  
262 each of the four concrete mixture types after seven weeks and transported to the lab where  
263 they were kept in aerated seawater aquaria. In the laboratory, all macro-organisms on the  
264 front face of the tiles were identified to species and presence/absence recorded using  
265 classical taxonomic methods.

266

#### 267 *2.5 Statistical analyses*

##### 268 *2.5.1 Chemical analysis*

269 Change in the combination of chemicals leached from tiles of different construction materials  
270 (Table 1) were compared using two analyses. Firstly, the 'community' of chemicals leached  
271 by tiles over time were compared using typical ecological tools (i.e. PERMANOVA,  
272 Multidimensional scaling (nMDS) and diversity indices), where the 'chemicals' are  
273 considered the 'species' and the chemical concentration the 'species abundance'. Chemical  
274 identity and concentration matrices were compared using PERMANOVA based on 9999  
275 unrestricted permutations (Anderson, 2001). Chemical community dissimilarity (see  
276 Oksanen et al. 2013) was calculated using Euclidean distance (see Clarke and Warwick,  
277 1994) and plotted using nMDS. Environmental fitting using the 'envfit' function (vegan  
278 package, R Core Team, 2016) was also used to overlay (1) a vector of the effect of time (a  
279 continuous variable) on dissimilarity, and (2) to determine the centroid (average) location of  
280 communities in relation to tile type (a categorical variable).

281

282 A univariate comparison of chemical leaching was also undertaken by reducing the  
283 community of chemicals into a univariate diversity index (i.e. Shannon-Wiener Index [H']) to  
284 test for differences in the evenness (proportional contribution to the total abundance) of  
285 chemicals leached from each tile type. One-way ANOVA was used to test for differences in  
286 H' values between each tile type, and post-hoc SNK tests used to show pairwise differences

287 between groups. Data were normal distributed but variances heterogeneous and were  
288 therefore log-transformed in order to remove heterogeneity.

289

290 The leaching of individual chemicals over time was assessed using LOESS curve fitting  
291 (local regression in the R package 'ggplot2') due to the temporal autocorrelation inherent in  
292 the data over time. Individual concentrations were log-transformed and assessed using  
293 goodness of fit ( $R^2$ ).

294

### 295 2.5.2 *Biological communities*

296 Two-way analysis of variance (ANOVA) was used to determine whether there was a  
297 significant effect of treatment (concrete type) on biofilm percentage cover and richness using  
298 the fixed factors: 'type' (four levels, fixed) and 'time' (4 levels (weeks), random, orthogonal).

299

300 One-way ANOVA was used to analyse the effect of concrete type on macro-fouling species  
301 richness after seven weeks using the fixed factor concrete 'type' (four levels, fixed). Data  
302 were analysed separately for native and non-native species. One-way ANOVA was also  
303 used to test the effect of concrete type on the relative proportion of non-native species. Post-  
304 hoc SNK tests were used to show pairwise differences between groups. GMAV version 5 for  
305 windows (Underwood & Chapman, 1998) was used for all analyses.

306

307 PERMANOVA was used to test for differences in multivariate native and non-native species  
308 compositions among concrete types, based on 9999 unrestricted permutations of raw  
309 presence/absence data (Anderson, 2001). Percentage contributions of individual taxa and  
310 functional groups to dissimilarity between communities were calculated using SIMPER  
311 (Clarke, 1993). SIMPER analysis in the PRIMER package was used to assess which  
312 species were most influential in causing similarity among plots within treatments and  
313 dissimilarity among different treatments (Clarke and Warwick, 1994).

314

### 315 3. Results

#### 316 3.1 Metal composition of the tiles

317 Metal concentrations in the individual components of the concrete are provided in Table 2a.  
 318 The sand and granite components contain relatively low acid-extractable metal  
 319 concentrations, reflecting their geological origins (high quartz content) and relatively small  
 320 surface area available to leach and/or desorb metal. The cement, PFA and GGBS, however,  
 321 have elevated concentrations for certain elements owing to their finer grain size providing a  
 322 larger surface area to leach/desorb metals and also their more varied and metalliferous  
 323 source material. Cement itself exhibited the highest levels of chromium and copper, with  
 324 PFA having greatest quantities of nickel, arsenic, molybdenum, cadmium, mercury, thallium  
 325 and lead; GGBS exhibited the highest concentrations of zinc and barium. These measured  
 326 concentrations are in line with those reported previously (Brigden and Santillo, 2002;  
 327 Ilyushechkin et al. 2012; Moreno et al. 2005; Yu et al. 2005). Estimated combined  
 328 concentrations of metal within each type of tile was calculated (Table 2b) based on the  
 329 composition of the tile type taken from Table 1 and multiplied by the metal content in the  
 330 individual constituents. Although there were significant variations in the concentrations of  
 331 elements present in the individual components owing to the replacement being only up to  
 332 8% by weight, the combined estimated concentrations in the final concrete mix do not vary  
 333 dramatically. Even then, only for Ba, Mo and Ni does the variation between tile types reach a  
 334 factor of two (Table 2b).

335 Table 2. Comparison of the mean metal concentrations in (a) the individual concrete  
 336 components and (b) in the four treatment types. (c) EQS = Environmental Quality Standard  
 337 each metal. Values in bold represent those that are above EQS. \* = Highest value for the  
 338 comparison among either the individual components or among treatments. SD = Standard  
 339 deviation.

		Concentration (mg/kg)										
(a)		Cr	Ni	Cu	Zn	As	Mo	Cd	Ba	Hg	Tl	Pb
Cement	Mean	<b>41.6*</b>	<b>17.9</b>	<b>65.5*</b>	<b>45.9</b>	9.92	1.19	<b>0.31</b>	129	0.04	<0.01	<b>27.8</b>
	SD	0.6	1.94	1.62	0.62	10.54	0.11	0.05	0.009	1.52	n/a	0.0003
Granite	Mean	0.17	0.12	2.34	1.25	0.62	0.01	0.01	3.3	0.02	0.06	1.2
	SD	0.0007	0.0019	0.0782	0.0573	0.0198	0.0117	0.0017	0.0002	0.099	0.0006	0.0024
Sand	Mean	0.14	0.03	1.13	1.13	0.67	<0.01 <sup>5</sup>	0.01	7.1	0.02	0.06	0.76

	SD	0.0064	0.0008	0.0538	0.0633	0.0412	n/a	0.0006	0.0011	0.2219	0.0005	0.0033
PFA	Mean	<b>32.1</b>	<b>47.7*</b>	<b>39</b>	<b>50.6</b>	23.1*	3.92*	<b>0.33*</b>	180	<b>0.08*</b>	0.19*	<b>50.2*</b>
	SD	0.3	0.3	0.38	1.53	0.65	0.13	0.11	0.008	5.51	0.0012	0.0049
GGBS	Mean	<b>26.5</b>	2.04	2.04	<b>147.5*</b>	10.2	<0.01 <sup>5</sup>	0.07	603*	0.01	0.01	<b>8.2</b>
	SD	0.27	1.56	0.5	2.99	5.63	n/a	0.009	0.009	9.61	0.003	0.001
<b>(b)</b>												
Control	Mean	<b>7.2*</b>	3.1	<b>12.5*</b>	<b>8.7</b>	2.2	0.21	0.06*	25.4	0.022	0.053	<b>5.5</b>
PFA	Mean	<b>6.8</b>	4.3*	<b>11.5</b>	<b>8.9</b>	2.7*	0.32*	0.06*	27.4	0.023*	0.059*	<b>6.4*</b>
GGBS	Mean	<b>6.6</b>	2.5	<b>10</b>	<b>12.8</b>	2.2	0.16	0.05	44.4	0.02	0.051	<b>4.7</b>
Mixed	Mean	<b>6.2</b>	3.7	<b>8.9</b>	<b>12.9*</b>	2.7	0.27	0.05	46.4*	0.022	0.057	<b>5.6</b>
<b>(c)</b>												
EQS <sup>1</sup>		0.6 <sup>2</sup> (32)	8.6 <sup>4</sup> (34)	3.76 <sup>2,3</sup>	7.92	252	n/a	0.24	n/a	(0.07) <sup>4</sup>	n/a	1.3 <sup>4</sup> (14)

340

341 <sup>1</sup> Environmental Quality Standard; <sup>2</sup>WFD, (2015); <sup>3</sup> Assumes DOC < 1 mg/l; <sup>4</sup>EU, (2013) ; <sup>5</sup>  
 342 For the purpose of calculations < values converted to half the LOD. All EQS as annual  
 343 averages unless in brackets denoting maximum admissible concentrations.

344

### 345 3.2 Metal leaching from the tiles

346 There were significant differences in the profiles of the elements and their concentrations  
 347 leached from the different treatments over time (Table 3a, Fig. 1a). While differences in  
 348 chemical composition were apparent, treatment only accounted for 6% of the variability,  
 349 whereas time accounted for 38% of the variability (Table 3a, R<sup>2</sup> values). Comparison of  
 350 chemical composition using a diversity index (Shannon-Wiener) better revealed differences  
 351 in chemical leaching between treatments (Fig. 1b). The PFA treatment (Type 2) exhibited  
 352 significantly greater log H' chemical compositions than GGBS and Mixed treatments  
 353 respectively, indicating increased leaching (Fig. 1b). The GGBS treatment had the lowest H'  
 354 values, indicating reduced leaching, although this reduction in H' was counteracted in the  
 355 Mixed treatment; although this experimental design cannot be used disentangle the effect of  
 356 introduced PFA from a reduced amount of cement in the mixture. Control concrete H' values  
 357 were statistically the same as tiles including both GGBS and PFA (Mixed treatment; p >  
 358 0.05), but higher than H' values in treatments with just GGBS (Type 3).

359 There were differences in the concentration of chemicals leached from the tiles (Fig. 2) over  
 360 time. In all instances, asymptote was reached (no further leaching) after a total of 250 h of

Disclaimer: This is the pre-proof version of the paper. Please go to <http://www.sciencedirect.com/science/article/pii/S0925857417303907> for the final version of the paper: McManus RS, Archibald N, Comber S, Knights AM, Thompson RC, Firth LB (2017) Partial replacement of cement for waste aggregates in concrete coastal and marine infrastructure: a foundation for ecological enhancement. Ecological Engineering, <https://doi.org/10.1016/j.ecoleng.2017.06.062>

361 immersion for all chemicals. Zinc (Zn) was the most abundant chemical and lead (Pb) the  
362 least abundant chemical leached from the tiles (Fig. 2).

363

364 Table 2. (a) PERMANOVA comparing the chemical types and concentrations leached  
365 Control, PFA, GGBS and Mixed tiles. Significant P-values are highlighted in bold. (b)  
366 ANOVA comparing Shannon diversity of chemicals leached from tiles of different type (as  
367 described in (a) above).

368

369 a).

Source	Df	MS	F	R2	P
Type	3	518.0	13.58	0.06	<b>&lt;0.001</b>
Time	1	9727.1	255.04	0.38	<b>&lt;0.001</b>
Type x Time	3	67.3	1.77	0.008	0.151
Residuals	371	38.1		0.55	

370

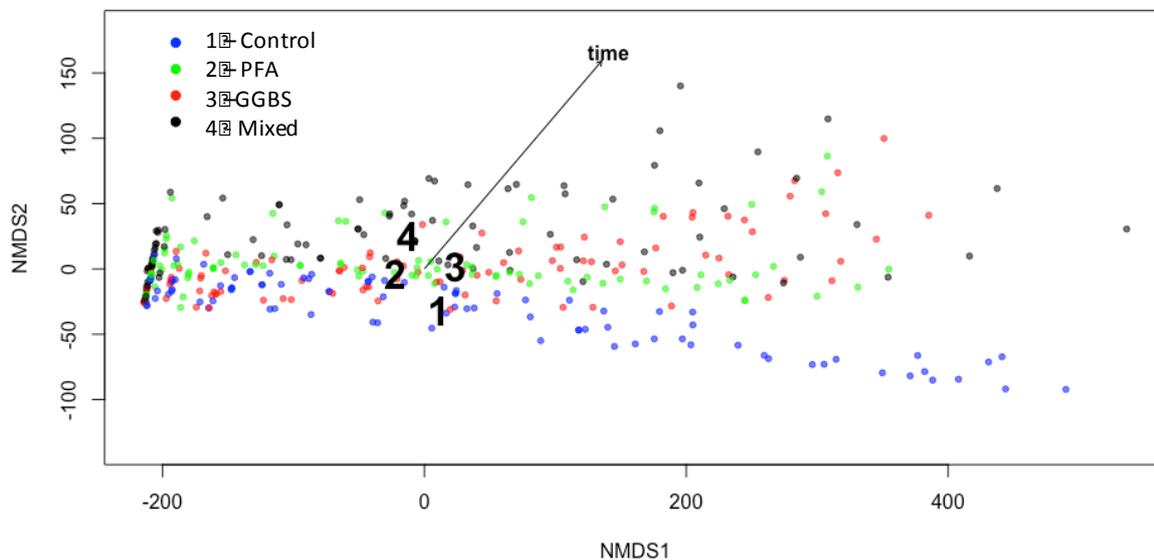
371 b).

Source	Df	MS	F	P
Type	3	0.546	12.25	<b>&lt;0.001</b>
Residuals	375	0.045		

372

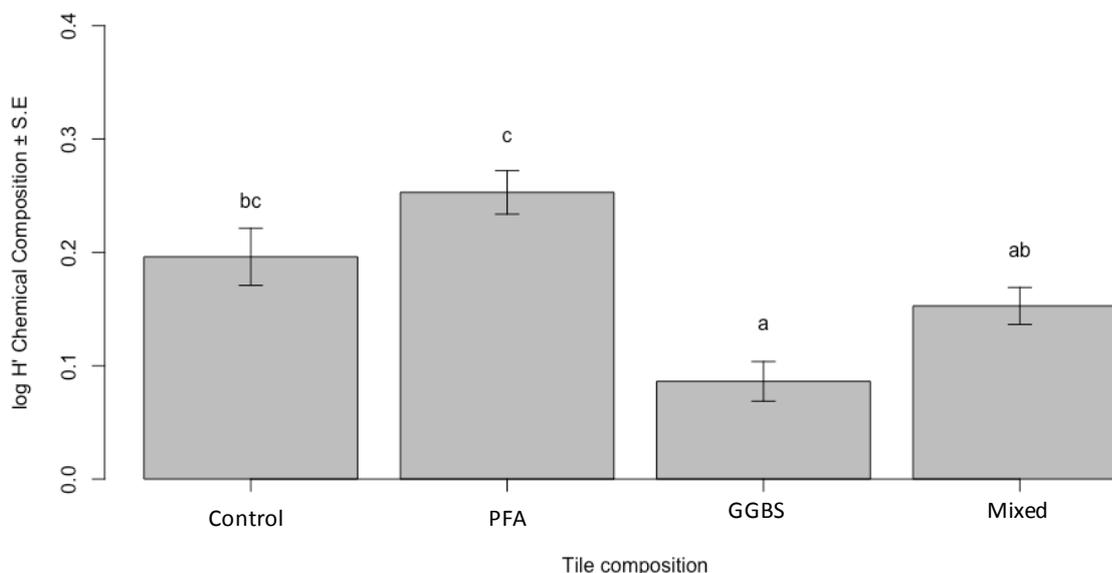
373

374 a)



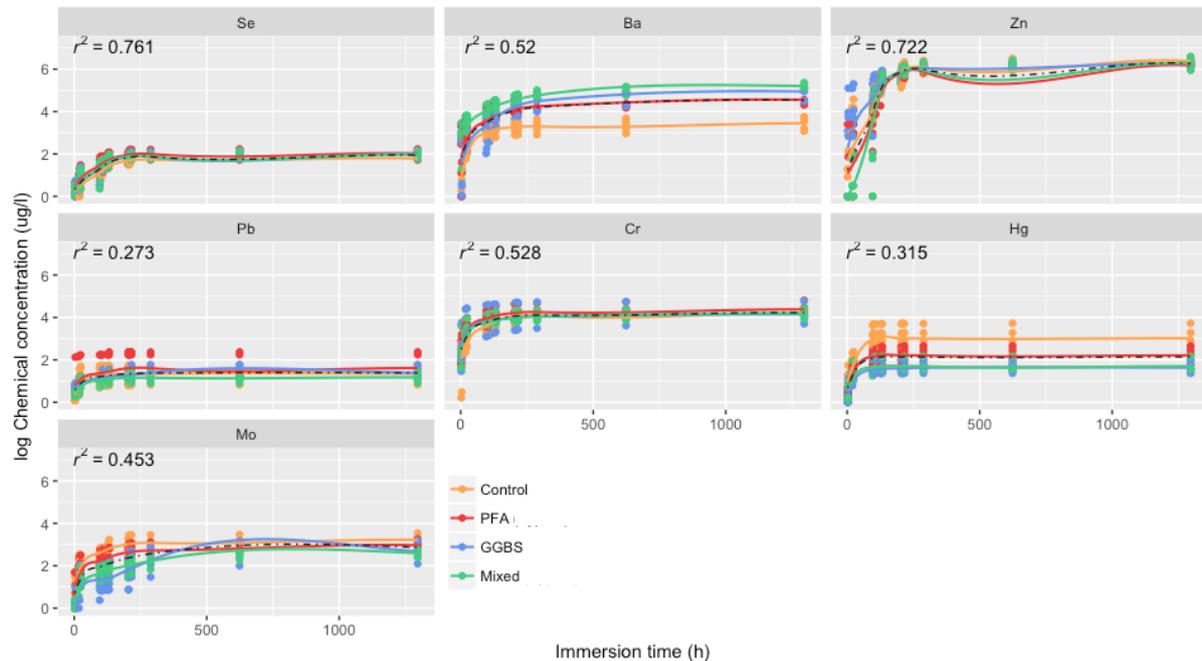
375

376 b)



377

378 Figure 1. (a) nMDS plot of dissimilarity of chemical composition (identity and concentration)  
379 leached from different tile types over time. Significant vector (time,  $p < 0.001$ ,  $R^2 = 0.56$ ) and  
380 the centroid (average location) of each tile type (Type 1 – 4) shown in bold lettering ( $p <$   
381  $0.05$ ,  $R^2 = 0.03$ ) are shown following environmental fitting. Stress = 0.01. (b) log mean  
382 Shannon diversity ( $H' \pm S.E.$ ) of chemicals leached from tiles of different composition. Letters  
383 denote statistical groupings; groups identified by post-hoc SNK tests; groups that do not  
384 share a letter are significantly different from one another ( $p < 0.05$ ).



385

386 Figure 2. Cumulative concentration of seven chemicals (ug/l) leached from tiles over the  
387 significant main effect of time (hours of immersion). Data are pooled across all tile types.  
388 Local regression (LOESS) lines are fitted and confidence limits (grey shading) are shown.  
389 Goodness-of-fit ( $R^2$ ) values for each LOESS line are shown.

390

391

### 392 3.3 Effects on colonising biofilms

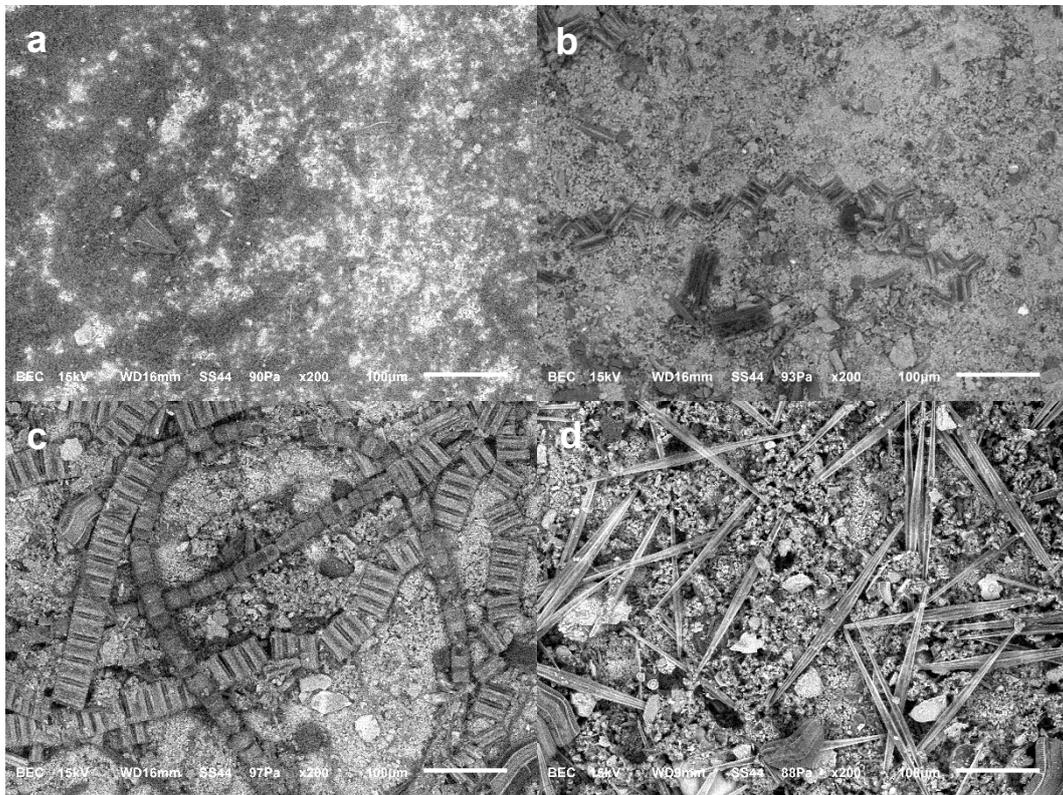
393 A total of seven genera of diatom were found to have colonised the tiles (*Thalassiothrix*,  
394 *Fragilaria*, *Asterionella*, *Amphiprora*, *Grammatophora*, *Surirella* and *Tabellaria*). Plate 1  
395 shows examples of images obtained from SEM. The ANOVA showed that there was a  
396 significant difference between treatments (Table 3), however post-hoc tests failed to identify  
397 where these differences were (Fig. 3a). There was no significant difference in biofilm  
398 diversity among treatments (Fig. 3b, Table 3), but both biofilm % cover and diversity  
399 increased significantly through time as expected (Fig. 3c,d, Table 3).

400

401

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404

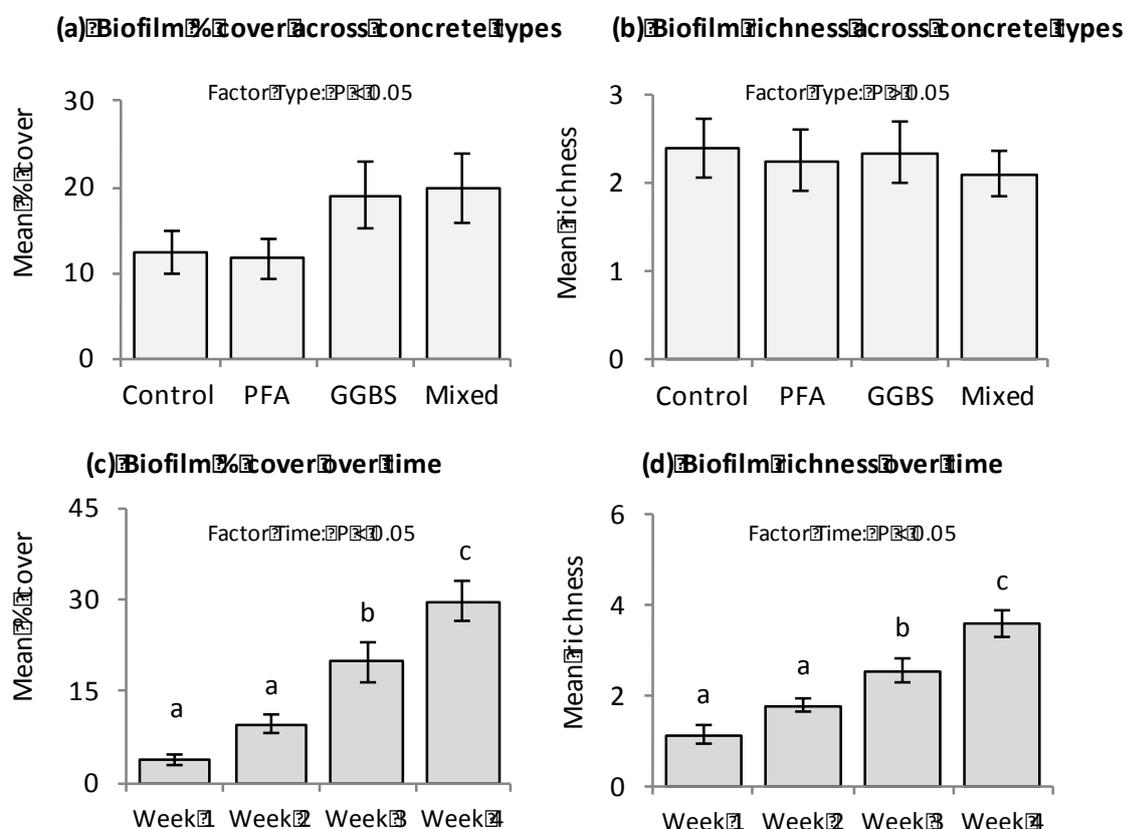
405 Plate 1. Examples of SEM images (x 200 zoom) obtained from control concrete blocks after  
406 (a) 1-wk, (b) 2-wk, (c) 3-wk, and (d) 4-wk.

407

408 Table 3. ANOVA comparing % cover and richness of biofilm among the four different  
409 concrete types across four weeks. Significant P-values are in bold.

Source	df	% cover			Richness		
		MS	F	P	MS	F	P
Type	3	369.35	3.21	<b>0.029</b>	0.35	0.26	0.857
Week	3	2628.15	22.86	<b>0.000</b>	22.15	16.18	<b>0.000</b>
Type x Week	9	125.36	1.09	0.383	0.31	0.23	0.989
Res	64	114.99					

410



411

412 Figure 3. Comparison of mean % cover and richness of biofilms across the significant main  
 413 effects of treatments (a,b, pooled over time) and time (c,d, pooled over treatment) over a 4-  
 414 week period (n = 20, ± SE). Letters denote statistical groupings; groups identified by post-  
 415 hoc SNK tests; groups that do not share a letter are significantly different from one another.  
 416 Despite ANOVA detecting a significant effect of treatment for biofilm % cover (a), post-hoc  
 417 SNK tests failed to detect any significant differences among treatments.

### 418 3.4 Effects on colonising subtidal macro-biota

419 A total of 27 species colonised concrete tiles after 7 weeks (Table S2). These comprised  
 420 ascidians (10 species), bryozoans (5 species), algae (5 species), amphipods (3 species),  
 421 sponges, hydroids, annelids and barnacles (1 species each). Five of the 27 species  
 422 identified were non-native to Britain but common in Plymouth Sound: the bryozoans  
 423 *Tricellaria inopinata* and *Watersipora subtorquata*, the hydroid *Bugula neritina*, the ascidian  
 424 *Botrylloides violaceus* and the barnacle *Austrominius modestus*.

425

426 Treatment had a significant effect on native species richness but no significant effect on  
 427 either non-native species richness or the proportion of non-native species per treatment  
 428 (Figure 4, Table 4). Control tiles had the highest mean native species richness (9.2) whilst  
 429 mixed tiles had the lowest (7.4). Control tiles had significantly greater native species  
 430 richness than both GGBS and mixed, but were not significantly different to PFA.

431  
 432 There was a significant difference in native species composition among treatments but no  
 433 significant difference was detected for non-native species (Fig. 7, Table 5). The native  
 434 macro-fouling assemblages associated with the controls exhibited the highest average  
 435 similarity (68.5%); Mixed exhibited the lowest average similarity (57.9%) and PFA and  
 436 GGBS were in between (61.3% and 61.4% respectively). This is reflected in the clustering of  
 437 the shapes for the controls and dispersed shapes for Mixed in Figure 5. SIMPER analysis  
 438 revealed that there were greater numbers of taxa associated with controls (the amphipod,  
 439 *Jassa marmorata*, the bryozoan, *Cradoscrupocellaria reptans*, the alga, *Ceramium rubrum*,  
 440 the ascidian *Ciona intestinalis*) than the other treatments. The ascidians *Ascidiella aspersa*  
 441 and *Botryllus schlosseri* were more positively associated with PFA and GGBS respectively  
 442 than the other treatments. Furthermore, two native species were unique to the Control (*Ulva*  
 443 *linza* and *Corella parallelogramma*). Non-native species were found on all concrete types,  
 444 but the invasive barnacle *Austrominius modestus* was unique to GGBS and Mixed  
 445 treatments.

446  
 447

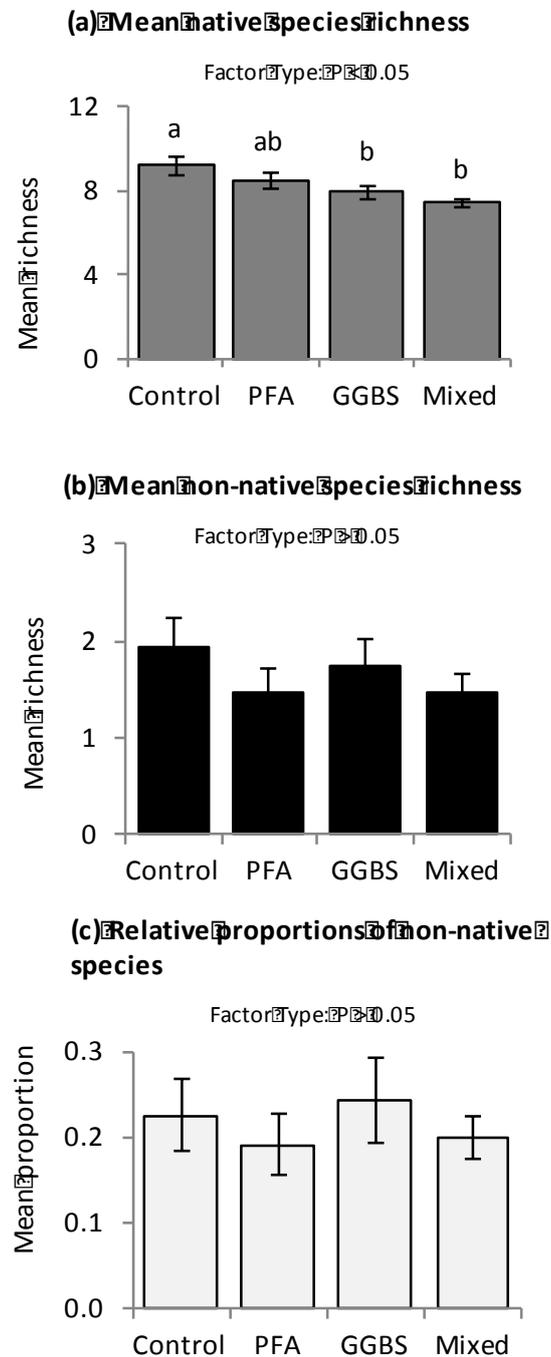
448 Table 4. ANOVA comparing mean richness of native and non-native species among the four  
 449 different concrete types. Significant P-values are in bold.

Source	df	(a) Native richness			(b) Non-native richness			(c) Proportion of non-native species		
		MS	F	P	MS	F	P	MS	F	P
Type	3	8.86	4.83	<b>0.005</b>	0.77	0.75	0.52	0.0086	0.38	0.7692
Res	56	1.83			1.02			0.0229		

450  
 451  
 452  
 453  
 454

455

456



457

458 Figure 4. Comparison (a) mean species richness of native species, (b) mean species  
 459 richness of non-native species (c) mean proportion of non-native species across the four  
 460 concrete types after 7 weeks (n = 15, ± SE). Letters denote statistical groupings; groups

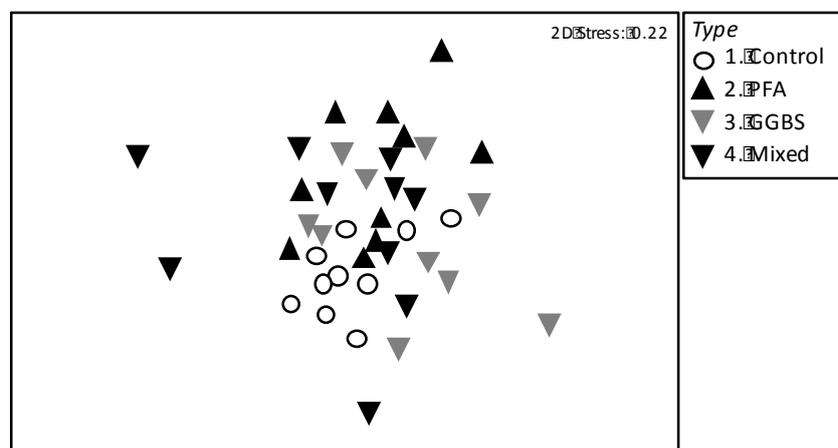
461 identified by post-hoc SNK tests; groups that do not share a letter are significantly different  
 462 from one another. Grey bars = native species, black bars = non-native species.

463

464 Table 5. PERMANOVA comparing native and non-native species compositions among the  
 465 four different concrete types. Significant P-values are in bold.

Source	df	Native composition			Non-native composition		
		MS	F	P	MS	F	P
Type	3	1776.5	2.16	<b>0.006</b>	0.53	0.52	0.929
Res	56	822.72			1.02		

466



467

468

469 Figure 5. MDS plot of native species assemblage dissimilarity among different concrete  
 470 types (see legend).

## 471 4. Discussion

### 472 4.1 Leaching chemistry of the concretes

473 Although cement replacement material had metal concentrations significantly higher than  
 474 Portland cement, leaching patterns were very similar reflecting the immobility of the metals  
 475 present. For example, Se in PFA was almost two orders of magnitude higher but was not  
 476 detectable in the seawater after leaching periods of up to 672 hours. This is likely a result of  
 477 the overall composition of the concrete containing relatively low percentages of PFA (24%  
 478 replacement of cement, but only 4% of the entire concrete mix by weight) and the fact that  
 479 the metals present are likely to be present as highly insoluble oxide and hydroxide minerals

480 as a product of the high temperature combustion processes from which they were derived.  
481 Leaching chemistry will only be occurring on the surfaces exposed to seawater (van-  
482 Jaarsveld et al. 1999). Thus the leaching rate of metals is a function of abundance and  
483 distribution of individual metals on the outer surface of the concrete (Shi and Kan, 2009).  
484 Despite efforts to ensure homogenous mixing, the potentially uneven distribution of metals  
485 within the concrete matrices, especially towards the exterior of the concrete is likely to be a  
486 factor in the leaching concentrations of metals observed in this study. In all cases, however,  
487 cumulative concentrations plotted against time show an equilibrium being reached at around  
488 250 hours.

489  
490 Successive immersions of the tiles in fresh seawater were undertaken to ensure that an  
491 'unnatural' equilibrium was not achieved in the immersion tanks, and successive reductions  
492 in the concentration of metal leached reflected a combination of surface chemistry,  
493 particularly chloride interactions (Perkol-Finkel and Sella, 2015) and natural physico-  
494 chemical partitioning processes. The final equilibration concentration tended to reflect the  
495 concentration of metal present in the concretes with Zn and Ba the highest and Hg the  
496 lowest, with elements such as As, Tl, Cd and Ni being present at below the analytical limits  
497 of detection. Based on this study, whether the leached metals are a cause for concern  
498 regarding toxicity to colonising organisms is difficult to determine. The tiles exposed to the  
499 natural seawater to allow biofilm growth benefit from almost infinite dilution into the  
500 surrounding seawater which cannot be replicated within the laboratory environment. There  
501 are likely to be elevated metal concentrations at the surface of the tile in contact with the  
502 biofilms, but again this is not practically measurable. It was for this reason that a number of  
503 water replacements were carried out to establish if fresh seawater interactions results in  
504 more or less leaching. The use of tiles submerged in tanks better mimics situations where an  
505 enclosed body of seawater is retained between tidal flushing events (e.g. harbours and  
506 marinas, where concrete is particularly prevalent). Under these conditions, it is then possible  
507 to compare observed leached concentrations with available EQS (Table 2). Concentrations  
508 of Zn in the bulk solution did exceed new UK EQS of 7.9 µg/l during individual immersions  
509 for all of the treatments reflecting the relatively high concentration in the base Portland  
510 cement material as well as the PFA and GGBS replacements. Hg also exceeded the EQS  
511 initially but thereafter was mostly less than the limit of detection (0.01 µg/l). The EQS for Hg  
512 (0.07 µg/l) is so low, it would require specialist analytical methods to accurately measure  
513 trends in the leaching rate. Lead would, in some cases, exceed the annual average EQS but

514 given the leaching appears to be largely ephemeral in comparison with the short-term,  
515 maximum admissible concentration (MAC) would be more appropriate and the critical EQS  
516 value of 14 µg/l was never exceeded at any point. The same could be said about Cr, with the  
517 annual average of 0.6 µg/l being exceeded for all treatments, but not the MAC (32 µg/l).  
518 There are no UK EQS for Mo, Se, or Ba. Although comparison with these EQS is in some  
519 ways indicative of potential impacts, the actual exposure scenarios for organisms in the  
520 vicinity to concretes in the environment may differ significantly.

521  
522 Based on likely exposure scenarios, it may be expected that the metal leached would at  
523 worst only cause impacts for a limited time as leaching occurs, and only at the very surface  
524 of the substrate or in an enclosed environment such as a disused dock with no tidal  
525 exchange. In reality, actual impacts will also depend on the speciation of the metal, which  
526 controls its bioaccessibility (Van veen et al. 2001). The fact that concentrations of metals  
527 leached from the PFA and GGBS amended concretes were of no higher magnitude than that  
528 of the Portland cement-based matrix suggests the benefits of using these replacement  
529 materials outweigh the risks. However, the findings of this study need to be tempered by the  
530 fact that the metal content of PFA and GGBS can vary significantly between sources of  
531 waste material. As an example, Hg can occur in PFA at over 20 times higher concentrations,  
532 Cd 5 times higher, Zn and Cr 3 times higher concentrations and Cu, Ni and As typically 2  
533 times higher. For GGBS, Cd can be up to 40 times higher in other similar materials, Cu 30  
534 times, Pb 20 times, Cr 5 times and Zn 2 times. These variations suggest that careful analysis  
535 of the replacement materials might be required prior to use within sensitive environments.  
536

#### 537 4.2 *Effects on colonising biofilm assemblages*

538 There was a significant main effect of concrete type on biofilm % cover, but post hoc SNK  
539 tests failed to detect differences between paired comparisons. SNK tests are not as sensitive  
540 to differences as F tests (ANOVA); in this instance, effect sizes were insufficiently large to  
541 assign any effect of concrete type to any differences in biofilm % cover. Additionally, there  
542 was no significant effect of concrete type on biofilm richness, suggesting that the cement  
543 replacements used have little or no effect on the development of colonising biofilms, at least  
544 not in the proportions used. Whilst some studies have found that surface chemistry affects  
545 colonising biofilm assemblages (Ista et al. 2004), others have suggested that it may be more  
546 important for bacteria than for microalgae (Cooksey and Wigglesworth-Cooksey, 1995).

547 Given the expertise available, we focussed only on diatoms. We acknowledge that this is a  
548 limitation of the present work and suggest that future studies should focus on all biofilm  
549 assemblages, particularly if there are differential responses to surface chemistry. Cloning  
550 and sequencing of 16S rRNA genes would also identify any effects on bacterial communities  
551 present in the biofilm (Lee et al. 2008). This could prove an important factor later in  
552 succession, as biofilm dynamics are thought to play a major role in accelerating the  
553 settlement of macro-algae (Park et al. 2011) and succession by invertebrates on artificial  
554 surfaces (Siboni et al. 2007), with implications for antifouling technologies and aquaculture  
555 (Qian et al. 2007). Research has suggested that differences in the dynamics of microbial  
556 biofilms can alter (enhance or inhibit) precipitation of calcium carbonate onto artificial  
557 structures, having implications for engineering applications such as bio-grouting and self-  
558 healing concrete (Darquennes et al. 2016; Decho, 2010). As this study has focused on the  
559 early successional phases of biofilms over a 7-wk period, we are therefore unable to shed  
560 further light on the role of biofilms in promoting later successional phases and long-term  
561 development of macrofouling communities, although this may well be an important process  
562 in the colonisation of artificial structures post-construction.

563  
564 It is important to note that in this study, the largest total cement replacement was 48%,  
565 whereas cement replacements of up to 80% GGBS and 50% PFA are routinely used in the  
566 marine environment (Chalee et al. 2010; Thomas and Matthews, 2004). They therefore have  
567 the potential to have a much greater effect on surface chemistry and therefore biofouling  
568 assemblages.

#### 569 570 4.3 *Effects on colonising macro-fouling assemblages*

571 The control treatment exhibited highest native species richness which generally decreased  
572 significantly with the addition of GGBS. The addition of PFA alone had no significant effect  
573 on native species richness compared to the Portland cement control. Conversely, the  
574 replacement of cement with GGBS (in both GGBS only and Mixed treatments) significantly  
575 lowered native species richness compared to the control, suggesting that GGBS may have a  
576 greater negative effect on macro-fouling species than PFA. Leaching tests, however,  
577 showed slight, but statistically different leaching characteristics, with the concrete containing  
578 GGBS leaching less metal overall (Fig. 1), but individual metal concentrations did vary.  
579 GGBS contained higher concentrations of Zn and Ba, but lower in all of the other metals

580 detected compared with PFA or Portland cement. The higher Zn content of GGBS could, for  
581 example, have an impact owing to its biocidal properties, exploited in anti-fouling paints  
582 using active ingredients such as zinc pyrithione (Cima & Ballarin, 2015). Interestingly, both  
583 PFA and GGBS contained lower concentrations of Cu than the cement, which too, has  
584 biocidal properties and is widely used in anti-fouling paints. Whether it is these specific  
585 elements that are having an impact on the species richness or possibly more likely,  
586 colonisation is being controlled by surface interactions affecting the bioavailability and  
587 speciation of the leaching metals rather than impacts within the bulk solution would need  
588 further elucidation.

589

590 As concrete types with cement replaced by GGBS exhibited lower native species richness, it  
591 can be inferred that these communities have a lower stability and a lessened ability to  
592 recover from disturbance events (Oliver et al. 2015). Given the fact that the concrete types  
593 tested in this study had relatively low percentage replacements of cement, ecological effects  
594 may be even greater in marine concretes of higher cement replacement. Alternatively,  
595 differences could be down to effects other than this such as micro-scale differences in  
596 surface roughness caused by the cement replacements, although the lack of effect on  
597 biofilm cover does not lend itself to this conclusion.

598

599 Concrete type had a significant effect on native species composition. Macro-fouling  
600 assemblages associated with the control treatment exhibited the highest similarity compared  
601 to the Mixed treatment exhibiting the lowest similarity (Figure 5), suggesting that the  
602 assemblages that colonise Portland cement CMI are fairly homogenous and comprise a  
603 predictable suite of species. Conversely, assemblages that colonise CMI comprising a  
604 mixture of cement replacements are less predictable and are comprised of a greater range  
605 of species with varying tolerances to the chemicals associated with the cement  
606 replacements.

607

608 There were no significant differences in mean richness, mean proportion or composition of  
609 non-native species among concrete types. Many studies have stated that artificial structures  
610 facilitate the spread of marine invasive species (Airoldi et al. 2015; Mineur et al. 2012;  
611 Simkanin et al. 2012). Although this is usually attributed to low habitat heterogeneity, results  
612 here suggest that differences in relative abundance of non-native species may be due to the  
613 harsher chemical environment, where invasive and opportunistic species can thrive (Como

614 and Magni, 2009). The results suggest that although concrete CMI may facilitate threats to  
615 native biodiversity, there may be ways of limiting this by altering the materials used in marine  
616 concretes. Biological invasions by non-native species are acknowledged as one of the most  
617 important factors affecting the structure and functioning of marine ecosystems (Ojaveer and  
618 Kotta, 2015). They generally threaten native species and lower conservation value (Kernan,  
619 2015). Whilst it is often thought that native species outperform non-natives in an ecosystem  
620 function role (Strayer, 2012), recent studies are finding that non-natives are in fact  
621 performing at similar (Zwersche et al. 2016) and sometimes higher levels than their native  
622 equivalents (Borsje et al. 2011). Despite this, it has been suggested that non-native species  
623 do not generally impair ecosystem function, and may actually expand it by adding new  
624 ecological traits, expanding existing ones and increasing redundancy of functional groups  
625 (Reise et al. 2006). Some research has even highlighted that as biological invasions are  
626 ultimately inevitable because of climate change, non-native species may become integral to  
627 future conservation plans, and may even become valued for the ecosystem services they  
628 provide, particularly as they tend to be more resilient and persistent than their indigenous  
629 counterparts (Hobbs et al. 2006; Schlaepfer et al. 2011).

630

#### 631 4.4 *Future research directions*

632 This study represents one of the first studies to empirically combine chemical leaching data  
633 with biological data of micro- and macro-fouling species. It must be acknowledged that the  
634 results presented here reflect a very short-term study carried out over just 7-wk at a single  
635 location. Despite the limitations of the conclusions that can be drawn from a small-scale  
636 study such as this, we argue that the study system warrants further investigation to better  
637 understand the potential broader implications of cement replacement by waste aggregates.  
638 Future studies should consider all micro-organisms present in the biofilm and not just  
639 diatoms. Here, macro-fouling species were only assessed after 7 wk and a longer-term study  
640 would be beneficial to better understand colonisation patterns and the role of biofilms in  
641 facilitating macro-fouling on artificial structures. Furthermore, % cover of both macro- and  
642 micro-fouling assemblages (including bacteria) should be considered as much insight can be  
643 gained from assessing live cover in addition to species richness. Given that CMI can span  
644 the entire vertical gradient from subtidal to intertidal, it is very possible that the chemical  
645 leaching from the concrete may interact with air, and indeed there may be interactions with  
646 local weather conditions (temperature, precipitation); therefore, we advocate that future work

647 should conduct experiments in both the intertidal and subtidal environment and across the  
648 salinity gradient.

## 649 **5. Conclusions**

650 Native species richness and composition was affected by concrete type, but non-native  
651 species richness and composition was unaffected. This implies that the use of different  
652 concrete types does not influence the ability of non-native species to colonise the concrete  
653 surface, but does appear to impact native species. Sella and Perkol-Finkel (2015)  
654 highlighted the lower abundance of invasive species and higher abundance, richness and  
655 diversity on native species on EConcrete® compared to regular concrete. This branded  
656 version of environmentally friendly concrete has a lower pH (9–10.5) compared to a standard  
657 Portland cement (12.5–13.5). This research supports the conclusion that differences in  
658 species richness on different concrete types are potentially caused by differences in surface  
659 chemistry.

660

### 661 *5.1 Potential applications of the results*

662 The information presented in this study could be used to inform further research to enhance  
663 the ecological value of concrete marine infrastructure by enhancing ecosystem services, as  
664 well as by adding nature conservation value. Studies have shown that biodiversity  
665 associated with sea-defences has beneficial effects, such as attenuating waves, trapping  
666 sediments and even strengthening the structures (*sensu* 'bioprotection'; Gowell et al. 2015;  
667 Risinger, 2012; Coombes et al. 2017). This dynamic interaction between ecology, chemistry  
668 and engineering can be implemented to enhance the ecological potential of coastal  
669 defences. This research suggests that chemical composition should also be considered  
670 when designing artificial structures.

671

672 Concrete is the major construction material used in the creation structures like harbour walls,  
673 marinas and other semi-enclosed marine environments. In such places, a high level of  
674 human activity (Knights et al. 2011; Pearson et al. 2016) often means that water quality is  
675 compromised by high concentrations of nutrients leading to algal blooms (de Jonge et al.  
676 2002). It may be possible to enhance ecosystem functions like filter-feeding by tailoring  
677 attachment substrate for desired species, and improve water quality through biofiltration

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678 (Wilkinson et al. 1996). Other marine structures, such as artificial reefs, could benefit from  
679 manipulating the biodiversity present, whilst providing a more suitable material than for  
680 example, old tyres which have been used previously (Morely et al. 2008) to construct better  
681 artificial reefs, with a more natural benthic community, in turn encouraging a more natural  
682 structure of associated fish populations (Perkol-Finkel and Benayahu, 2007).

683

684 Extensive research has shown that physical features of artificial marine structures such as  
685 topographic complexity and water retaining features have a major role in enhancing their  
686 ecological attributes. However, results presented here highlight the fact that differences in  
687 concrete composition can have significant effects on the biodiversity of subtidal fouling  
688 organisms that colonise artificial surfaces. This information could be used in future to help  
689 design features that enhance biodiversity and the ecosystem services this provides at little or  
690 no extra cost.

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696

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