

Leaching of flame-retardants from polystyrene debris: Bioaccumulation and potential effects on coral

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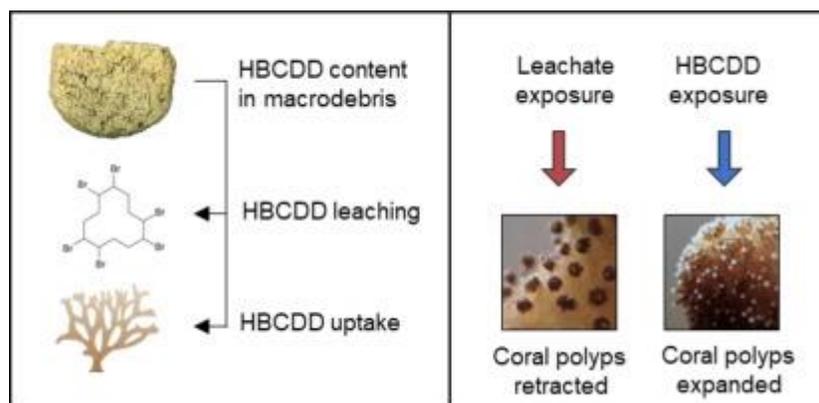
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Abstract :

Marine plastic debris can act as a reservoir of chemical additives that can pose a potential threat to sensitive ecosystems such as coral reefs. A survey of foam macrodebris collected on beaches indeed revealed high concentrations of hexabromocyclododecanes (Σ HBCDD) in polystyrene (PS) samples (up to 1940 $\mu\text{g g}^{-1}$). Results also showed that PS fragments can still leach over 150 $\text{ng g}^{-1} \text{d}^{-1}$ of Σ HBCDD (primarily as the α -isomer) for relatively long durations, and that these additives are readily bioaccumulated and well-retained by corals. Despite significant HBCDD bioaccumulation in coral tissue, short-term exposure to HBCDD or PS leachate had no considerable effect on coral photosynthetic activity, symbiont concentration and chlorophyll content. Exposure to the PS leachate did however cause consistent polyp retraction in nubbins over the 5-day exposure. This response was not observed in animals exposed to HBCDD alone, suggesting that another constituent of the leachate stressed corals.

Graphical abstract



Highlights

► Most beached foam macrodebris had elevated concentrations of HBCDD. ► HBCDD containing polystyrene can release additives over relatively long durations. ► Corals can bioaccumulate these brominated flame retardants. ► Polyp retraction was the only short-term observed physiological effect of leachate.

Keywords : Marine plastic, Polystyrene foam, Leachates, Hexabromocyclododecanes, Corals, Bioconcentration

54 **1 Introduction**

55 Plastic pollution is found in all ecosystems on the planet, especially in oceans where their
56 persistence and potential adverse effects to marine life and human health has become a major
57 concern (Chiba et al., 2018; Lebreton et al., 2017; Rochman, 2018). Emerging questions, such
58 as the role of plastic debris as a source of chemical contaminants in marine ecosystems, have
59 consequently been of pressing interest. Polystyrene (PS) is among the most used plastic in the
60 world, and as a result one of the most common type of plastic debris found in ocean, coastlines
61 and beaches (Balbi et al., 2017; Browne et al., 2008). In addition to its persistence and long-
62 range transportability, PS can contain large amounts of chemical additives, such as flame
63 retardants and plasticizers, which can leach from the plastic and pose an added threat to marine
64 biota. Hexabromocyclododecane (HBCDD) is now a restricted brominated flame retardant
65 (BFR) that has been primarily used in extruded and expanded polystyrene (PS) for building
66 materials, upholstery textiles and electrical devices (Law et al., 2005). With over 10,000 tonnes
67 annual market demand, HBCDD was the second most-used BFR in Europe in 2010 (de Wit et
68 al., 2010), after earlier limitations on the use of polybrominated diphenyl ethers (PBDEs).
69 HBCDD persistence and ability for long-range transport resulted in its ubiquity in abiotic and
70 biotic compartments (Covaci et al., 2006; Munschy et al., 2013). HBCDD was found to
71 biomagnify along the food chain (Ruan et al., 2018), which, together with its toxicity
72 (reproductive, developmental and behavioural effects; Marvin et al., 2011), prompted its
73 inclusion in the Stockholm Convention list on Persistent Organic Pollutants in 2013.

74 However, even after its production ban, HBCDD-containing plastics in the ocean act as a
75 reservoir of the compound, promoting its long-range transport and its stability. For example,
76 earlier studies found elevated concentrations of HBCDD in several common marine debris
77 items, including aquaculture buoys, and found that mussels growing on these PS buoys
78 accumulated high amounts of HBCDD (Jang et al., 2017, 2016). Despite these findings, the

79 impacts of plastic pollution and associated chemicals on many species, including corals, is still
80 largely unknown. Understanding the impacts of leaching chemicals from plastics in coral reef
81 environments has recently been highlighted as an important research priority (UNEP, 2019).
82 Indeed, coral reefs are one of the most important biodiversity hotspot on earth, hosting more
83 than 25% of all marine species, in addition to provide food and services to more than 850
84 million people (Pratchett et al., 2014). Their preservation has therefore become a priority for
85 many countries. The studied coral species *Stylophora pistillata* has a wide distribution
86 throughout all tropical reefs and is one of the main reef builders of the Red Sea. As a thermally
87 tolerant species, it is likely to be one of the main reef builders and ecosystem sustainers of future
88 reefs in a context of global changes. It is a well-studied coral and one of the most common
89 species used for coral physiology and ecotoxicology investigations.

90 The objectives of this study were to: (1) quantify the HBCDD content and isomeric profile of
91 foam-like plastic debris collected along the French Riviera coastline, (2) determine the leaching
92 kinetics of plastic-contained HBCDD to the dissolved phase, (3) assess the bioaccumulation of
93 HBCDD in the scleractinian coral *Stylophora pistillata*, and (4) investigate potential effects of
94 PS foam leachate and HBCDD in this species.

95 **2 Materials and methods**

96 **2.1 Sampling and characterisation of plastic debris**

97 Beached foam-like plastic macrodebris (n = 17) were collected from three sites along 50 km of
98 the French Riviera coastline in 2018 (Figure 1a; Cannes, Beaulieu-sur-Mer and Menton). Only
99 debris larger than 2 cm were targeted to provide sufficient material for further leaching
100 experiments. The outer surface of each macro debris (2–3 mm) was removed using a pre-
101 cleaned stainless-steel blade and only pristine material within the interior was analysed for
102 chemical composition and additives. A small fragment (< 1 mg) of each sample was

103 characterised by FTIR spectroscopy, in transmission mode (ALPHA II, Bruker UK Limited,
104 Coventry, UK), to identify the type of plastic.

105 **2.2 HBCDD leaching experiment**

106 Cubic fragments (0.5 to 1 mm) were evenly sliced from the interior of sample 1, found to
107 contain high concentrations of HBCDD isomers and confirmed as PS. Fragments (309 mg)
108 were placed in a sealed stainless steel meshed pouch and immersed in a 280 mL glass chamber,
109 filled with 1 µm-filtered clean seawater (FSW, IAEA seawater system). A peristaltic pump
110 continuously provided 1.8 mL min⁻¹ of clean FSW down the bottom of the chamber. The
111 overflow was collected for about 2 h at regular intervals to measure the leached HBCDD. The
112 first sample was taken 2 h after immersion of the plastic, after a volume of 280 mL has passed
113 through. PS fragments were kept under constant agitation in a room maintained at 20 – 24 °C
114 for 32 d (83 L of seawater). Leachate water samples (250 mL) were collected on day 0, 1, 2, 3,
115 7, 9, 10 and 14 and the PS fragments were analysed at the end of the 32 d leaching period.

116 **2.3 Coral exposure to HBCDD and PS leachate**

117 **2.3.1 Husbandry**

118 Five colonies of the scleractinian coral *Stylophora pistillata* (Esper 1797) from the Red Sea
119 (Gulf of Aqaba, Jordan) were used to generate a total of 54 nubbins (11/colony) of about the
120 same size (4 cm long; 2 cm wide). During the 3 weeks of healing, nubbins were maintained in
121 4 aquaria (30 L) continuously supplied with oligotrophic seawater at a flow rate of 20 L h⁻¹.
122 Constant seawater temperature of 25 ± 1 °C was maintained using temperature controllers
123 (Toshniwal N6100, ToshconH, West Instruments, Brighton, UK; ± 0.1 °C) and submersible
124 resistance heaters (Visi-ThermH Deluxe, Aquarium Systems, France). Three metal halide
125 lamps (Philips, HPIT 400W, Distrilamp, Bossee, France) provided a constant irradiance of 200
126 µmol photons m⁻² s⁻¹.

127 **2.3.2 Experimental design**

128 After healing, nubbins were randomly distributed in sixteen 500-mL glass beakers (3 nubbins
129 per beaker), and beakers were divided into the four following treatments (4 beakers per
130 treatment): (1) PS leachate (“leachate”), (2) HBCDD spiked in seawater (“HBCDD only”), (3)
131 a methanol control (“MeOH”) and (4) clean seawater (“control”). The PS leachate used for the
132 coral exposure experiment (1) was prepared by stirring 6 g of millimetric PS fragments from
133 debris #1 in 10 L of 1 μm -FSW (IAEA seawater system), in a glass container. Although PS
134 fragments tend to float, magnetic stirring ensured a thorough water exchange around the plastic
135 fragments. After 21 d, the leachate was filtered through GFF membranes (0.7 μm pore size)
136 and chemical analysis revealed a composition of 208 ± 36 , 16 ± 6 and 8 ± 6 ng L^{-1} of α -, β -, and
137 γ -HBCDD, respectively. The HBCDD exposure condition (2) was prepared by spiking the 3
138 isomers at concentrations comparable to those determined in the leachate (average measured
139 concentrations [$n = 7$] of 220 ± 128 , 25 ± 25 and 2 ± 3 ng L^{-1} of α -, β -, and γ -HBCDD,
140 respectively) with 20 μL of methanol as carrier solvent. Same amounts of the carrier solvent
141 methanol (20 μL) were added to seawater for the methanol control condition (3). The control
142 condition (4) consisted of clean FSW only. Corals were exposed to these treatments for 5 d,
143 with daily medium renewal. Water samples (10 mL) were taken every day after medium
144 renewal. Water temperature was kept constant at 25 $^{\circ}\text{C}$ using a water bath and gentle magnetic
145 stirring ensured homogeneity in the beakers. After the 5 d-exposure period, one nubbin per
146 beaker was sampled for physiological measurements, and a second nubbin was sampled for
147 HBCDD analysis. The remaining nubbins were transferred to the 30 L maintenance aquarium,
148 which was continuously supplied with clean seawater, and allowed to decontaminate over 9 d.

149 A preliminary experiment was also conducted to determine the bioaccumulation behaviour of
150 HBCDD in *Stylophora pistillata*. Nine 500-mL glass beakers containing 400 mL of FSW were
151 spiked with α -, β -, and γ -HBCDD as described for the main experiment. Coral nubbins in

152 duplicate were immersed for 24 h in one of three treatments (3 beakers per treatment): (1)
153 seawater spiked with α -, β -, and γ -HBCDD, (2) FSW (control), and (3) FSW preconditioned in
154 HBCDD for 24 h to determine the partitioning of HBCDD within the test system. Water
155 samples (10 mL) were collected from all 9 beakers at regular time intervals: T0, 0.5 h, 1 h, 2 h,
156 4 h, 6 h, 8 h, 10 h, 24 h, and the corals were weighted and frozen at -20 °C for chemical additive
157 analyses (see 2.4) at the end of the experiment. The beaker walls were also extracted using
158 MeOH to provide insights on the potentially sorbed HBCDD.

159 **2.3.3 Coral physiology**

160 Net photosynthesis (P_n) and respiration (R) rates were assessed on 4 nubbins per condition using
161 oxygen flux measurements. For this purpose, nubbins were incubated under constant stirring in
162 a thermally regulated 50 mL Plexiglas beaker completely filled with 0.45 μm -FSW. Oxygen
163 fluxes were monitored over 60 min (in 1 min intervals) in the dark (R rates) and at 200 μmol
164 quanta $\text{m}^2 \text{s}^{-1}$ (P_n rates), respectively, using an Unisense optode connected to a computer with
165 Oxy-4 software (Chanel fiber-optic oxygen meter, PreSens, Regensburg, Germany). Optodes
166 were calibrated before each experiment against nitrogen-saturated and air-saturated seawater
167 for the 0% and 100% oxygen, respectively. Light was provided by a metal halide lamp (Philips,
168 HPIT 400W, Distrilamp, Bossee, France). P_n and R rates were then estimated by regressing
169 oxygen data against time and gross photosynthesis (P_g) was calculated according to $P_g = P_n -$
170 R . At the end of the incubations, nubbins were frozen for the subsequent determination of
171 symbiont density and total chlorophyll concentration. Coral tissue was removed from the
172 skeleton of each colony using an air pick and homogenized with a Potter tissue grinder.
173 Symbiont density was quantified on a Coulter Counter (Beckman Coulter, France) and
174 chlorophyll concentrations were determined following a spectrophotometric method (Jeffrey
175 and Humphrey, 1975). Both symbiont density and chlorophyll content, together with the

176 respirometry data, were normalised to the skeletal surface area measured using the single wax-
177 dipping technique (Veal et al., 2010).

178 **2.4 Analysis of chemical additives**

179 Concentrations of the brominated flame retardants: HBCDD (α -, β -, and γ -isomers),
180 tetrabromobisphenol A (TBBPA), 2,4,6-tribromophenol (TBP) and the plasticizer bisphenol A
181 (BPA) were evaluated in all plastic samples and HBCDD isomers in leachate and coral samples.
182 $^{13}\text{C}_{12}$ TBBPA, β -HBCDD d18, γ -HBCDD d18 were used as surrogate standards. All native and
183 isotope-labelled standards were of the highest purity commercially available and purchased
184 from Wellington-Labs (Ontario, Canada), Accustandard (New Haven, USA), Sigma-Aldrich or
185 LGC Standards. Solvents were of HPLC or LCMS grade and provided by Fisher Scientific
186 (France). Ultra-pure water was produced on-site using a Millipore Milli-Q system (specific
187 resistivity of 18.2 M Ω cm - 25 °C).

188 For plastic fragments, 4–5 mg of each piece was spiked with 50 ng of labelled surrogate
189 standards and dissolved in 0.5 mL of dichloromethane (DCM). The polymer was precipitated
190 upon addition of 0.5 mL of methanol (MeOH), and the solvent mixture was concentrated by
191 evaporation to a volume of 0.3 mL. The clear supernatant in MeOH (DCM evaporated) was
192 collected after centrifugation and spiked with 10 ng of the internal standard (α -HBCDD d18)
193 before analysis.

194 Water samples (250 mL), spiked with 20 ng of labelled surrogate standards, were acidified to
195 pH 2 and DCM extracted with three times 5 mL of solvent. It was found that acidification
196 improved the recoveries of TBP through protonation. The combined extracts were dried with
197 sodium sulphate, concentrated to 0.3 mL under a gentle stream of nitrogen and spiked with 10
198 ng of the internal standard before analysis.

199 For the analyses of HBCDD in the coral tissues, nubbins were first freeze-dried, crushed, and
200 then spiked with 20 ng of labelled surrogate standards and extracted in 3 times 10 mL of DCM
201 in a sonication bath. The combined extracts were concentrated to 1 mL and purified on a 2 g
202 silica cartridge with 10 mL of DCM (lipid removal $91\pm 1\%$). The final extract was concentrated
203 to 0.3 mL and spiked with the internal standard before analysis. HBCDD concentrations in coral
204 tissue were normalized to the total dry weight (dw) of the coral nubbin.

205 Analyses were performed by ultra-performance liquid chromatography coupled with tandem
206 mass spectrometry, interfaced with an electrospray ionisation source, operated in negative mode
207 (UPLC-ESI-MSMS, Waters H-Class, Xevo TQ-D). Chromatographic separation was achieved
208 on a Waters BEH C18 column (100×2.1 mm, particle size $1.7 \mu\text{m}$) within 12 min using as
209 mobile phases ultra-pure water and methanol both containing 0.2 mM of ammonium acetate, at
210 a flow rate of 0.3 mL min^{-1} and a column temperature of $60 \text{ }^\circ\text{C}$. The chromatographic gradient
211 went from 50 to 100% methanol in 8 min, for a total run time of 12 min. Sample injection
212 volume was set at $10 \mu\text{L}$. Ionization and mass spectrometer parameters are reported in Table
213 S1.

214 The accuracies, given in Figure S1, were found to be in an acceptable range of 80–120%
215 regardless of the matrix, with variability below 15% in water and tissues and between 6 and
216 36% in the polymers.

217 Recoveries of the surrogate standards were satisfactory: 73 ± 35 , 64 ± 16 and $48 \pm 19\%$ for
218 $^{13}\text{C}_{12}$ TBBPA, β -, and γ -HBCDD-d18 in plastic pieces ($n = 35$), 70 ± 16 and $70 \pm 15\%$ for β -,
219 and γ -HBCDD-d18 in water ($n = 129$) and 81 ± 14 and $77 \pm 13\%$ for β -, and γ -HBCDD-d18 in
220 corals ($n = 30$). Although no standard guidelines are available for the analysis of flame
221 retardants in plastic matrices, these values were acceptable according to the requirements of the
222 European Commission guidance document SANTE/11813/2017 for pesticide residues analysis

223 in food, with recoveries in the acceptable range of 70-120% or RSD below 20% (European
224 Commission, 2017).

225 **2.5. Statistical analysis**

226 Concentrations of each HBCDD isomer measured in the plastic debris pre- and post-leaching
227 were analysed using a two-way ANOVA. Concentrations of Σ HBCDD in the coral nubbins
228 after the exposure and depuration phases were analysed using a two-way ANOVA with time
229 (exposure and depuration) and treatment as factors followed by Sidak's multiple comparison
230 tests. Coral physiological data were analysed using one-way ANOVAs followed by Tukey's
231 multiple comparison tests. All data met assumptions of normality and homogeneity of variance
232 and were analysed using GraphPad Prism 8.0. Differences are considered significant when $p <$
233 0.05.

234

235 **3 Results and discussion**

236 **3.1 Characterisation of the plastic debris**

237 **3.1.1 Polymer type**

238 The FTIR spectra of 16 of the 17 plastic pieces had a typical profile of PS, with aromatic and
239 aliphatic carbon-hydrogen bond stretches around $3,000\text{ cm}^{-1}$ and an aromatic carbon-carbon
240 bond stretch at $1,600\text{ cm}^{-1}$ (spectrum provided in Figure S2). Sample #13 had a distinct
241 spectrum, attributed to polyurethane by library match. This agreed with its different appearance
242 and the fact it did not dissolve in DCM, unlike the other debris.

243 **3.1.2 Additive concentrations**

244 Of the 16 PS debris screened for plastic additives, HBCDD was found in 8 fragments at
245 concentrations above $100\text{ }\mu\text{g g}^{-1}$ (sum of the isomers) and up to $1,940\text{ }\mu\text{g g}^{-1}$ (Figure 2a). The

246 highest Σ HBCDD concentration was observed in a sample of black colour (#7), attributed to
247 graphite beads typically used in insulation boards. The phenolic compounds were found in only
248 3 samples (PS #15, 16, 17), with BPA at concentrations up to $4,565 \mu\text{g g}^{-1}$ and TBP and TBBPA
249 at approximately $1,000 \mu\text{g g}^{-1}$ (Figure 2.b). The consistency in the concentrations of all
250 compounds between these 3 PS pieces suggests that they originated from a similar source,
251 possibly from the breakdown of a larger piece.

252 HBCDD concentrations in brand new expanded PS material were found to be comparable to
253 concentrations in a wide range of consumer products (Rani et al., 2014), with generally higher
254 concentrations in construction material. For this application, concentrations up to 1 order of
255 magnitude higher than those measured in our study (0.2 to 2.4% by weight) have been reported
256 (Jeanner at et al., 2016). In marine expanded PS debris, Jang et al. found large variations in
257 their HBCDD concentrations, ranging between 86 and $4,680 \mu\text{g g}^{-1}$, which encompass
258 concentrations reported in this study (Jang et al., 2017).

259 Regarding the phenolic compounds, their presence in extruded PS boards has not been
260 described elsewhere. TBBPA is used as an additive flame retardant in polyolefin resins, and
261 high-impact polystyrene resins (Li et al., 2019), whereas TBP is mainly used as a reactive
262 intermediate in the preparation of flame retardants (Pivnenko et al., 2017), and found also mixed
263 with the other BFRs in brominated high impact polystyrene plastics (Grabda et al., 2018). To
264 our knowledge, there is no data available on concentrations and possible leaching of unreacted
265 brominated phenols from plastic containing fire retardants derived from TBP (Howe et al.,
266 2005). As TBP and BPA are also degradation products of TBBPA, the presence of these
267 phenolic compounds in some of the PS debris (#15, 16, 17) could also be derived from the
268 degradation of TBBPA during migration from the plastic debris or photodegradation that might
269 occur in the PS (Khaled et al., 2018).

270 **3.1.3 Diastereoisomer compositions**

271 The HBCDD isomeric profiles differed between most samples (Figure 2a). β -HBCDD was the
272 lowest in all 8 samples (7–30% of Σ HBCDD), α -HBCDD prevailed in 5 samples (PS #1, 14,
273 15, 16, 17) while γ -HBCDD was dominant in 3 (PS #7, 9, 10). HBCDD is incorporated in the
274 polymer as a technical HBCDD mixture composed of 11.8% α -HBCDD, 5.8% β -HBCDD, and
275 81.6% γ -HBCDD (Heeb et al., 2005), which differs from its composition at equilibrium,
276 consisting of approximately 80% α -HBCDD, 11% β -HBCDD, and 8% γ -HBCDD (Li and
277 Wania, 2018). During thermal processing of the polymeric material, HBCDD isomeric profile
278 will partially shift towards this equilibrium values depending on the processing time and
279 temperature. This thermal isomerization of γ -HBCDD to α -HBCDD results in a distribution of
280 α - to γ -rich profiles in the end product. Although our samples might have been processed long
281 before collection, the differing isomeric ratios were attributed to this initial processing as
282 isomerisation during remaining polymer lifecycle stages is assumed to be insignificant (Li and
283 Wania, 2018).

284 **3.2 Leaching ability**

285 HBCDD was released from the PS debris with concentrations in the leachate between 20 and
286 132 ng L⁻¹ (sum of isomers) within 14 days, at room temperatures (20–24 °C). The first sample
287 (taken 2 h after immersion of the plastics) showed the highest concentration (Σ HBCDD 132 ng
288 L⁻¹), whilst samples from day 2 to day 14 had consistent lower concentrations (Σ HBCDD 20–
289 61 ng L⁻¹), with relatively minor fluctuation over time (43% RSD; data not shown). This
290 behaviour could relate to the rapid desorption of HBCDD on the surface of the particles,
291 followed by a slower release of internal HBCDD, with kinetics limited by its diffusion within
292 the polymer. Note that the desorption kinetics were determined on a pristine PS material, which
293 would likely be affected by biofouling in the marine environment. After 32 d of leaching,
294 HBCDD concentrations in the plastic debris were not significantly different to the initial ones
295 for all 3 isomers (Two-way ANOVA; $F(1, 6) = 1.78$, $p = 0.23$; data not shown). Between 70

296 and 230 ng of Σ HBCDD was leached into the water each day (fluxes over 150 ng g⁻¹ d⁻¹),
297 yielding to a loss of concentration comprised between 7 and 24 μ g g⁻¹ of Σ HBCDD in the debris,
298 which is smaller than our standard deviation on HBCDD concentration in plastic (30 μ g g⁻¹ on
299 4 replicates). The variability of the measurement of high concentrations of HBCDD in PS can
300 be a limitation to the accurate evaluation of the amounts leached, which are better characterised
301 through the water analysis. In a previous 6-month field study, slow or no losses of HBCDD was
302 found from expanded PS spherules, after an initial rapid (< 7d) release (Rani et al., 2017).
303 Physical properties of the microplastics such as their density, size, or surface-area-to-volume
304 ratio as well as other factors such as temperature, fouling or roughness of the sea will likely
305 influence the leaching kinetics and comparisons between studies are not straightforward
306 (Teuten et al., 2009).

307 The α isomer was found to be the prevailing diastereomer of HBCDD in the leachate with 78 \pm
308 3% of the Σ HBCDD throughout the whole 14 days, despite contributing to only 50 \pm 4% of the
309 total HBCDD in the PS plastic debris. This leaching behaviour is consistent with the higher
310 solubility of α -HBCDD (48.8 μ g L⁻¹) compared to that of β -HBCDD (14.7 μ g L⁻¹) and γ -
311 HBCDD (2.1 μ g L⁻¹) at 20 °C (Covaci et al., 2006) and with other landfilled waste studies
312 (Stubbings et al., 2016), where α -HBCDD was preferentially leached at 20 °C compared to γ -
313 HBCDD. HBCDD diastereomer leaching was found to be temperature dependant, such that
314 increasing temperatures from 20 to 80 °C caused a ~30-fold increase in γ -HBCDD in the
315 leachate, while the α -HBCDD concentrations only increased by a factor of 4. Although the
316 amounts and kinetics are specific to experimental designs, our results provide further evidence
317 that PS debris can release HBCDD during their long-distance transport in the ocean, potentially
318 contaminating remote areas receiving plastic litter (Lebreton et al., 2018).

319

320 **3.3 Bioaccumulation by *Stylophora pistillata* corals**

321 Results of the preliminary experiment for HBCDD bioaccumulation kinetics in corals are
322 provided in the supplementary data (Figure S3 and Figure S4). In brief, water concentrations of
323 the 3 isomers of HBCDD remained constant during 24 h in the absence of coral nubbins (Figure
324 S3), indicating no significant HBCDD adsorption on the wall of the glass beakers or on the stir
325 bar. On the contrary, the treatment with coral nubbins showed rapid bioaccumulation of
326 HBCDD in coral tissue, with water concentrations rapidly decreasing to under 15% of its initial
327 value in 6 h. Corals exposed accumulated up to $150 \text{ ng g}^{-1} \text{ dw}$ ($109 \pm 44 \text{ ng g}^{-1} \text{ dw}$) Σ HBCDD,
328 the majority of which was in the form of α -HBCDD ($74 \pm 1.5\%$) (Figure S4).

329 In the main coral exposure experiment, HBCDD concentrations (sum of the isomers) in coral
330 tissue were not significantly different between the PS leachate and HBCDD alone, with $30 \pm$
331 $12 \text{ ng g}^{-1} \text{ dw}$ and $53 \pm 28 \text{ ng g}^{-1} \text{ dw}$, respectively (Figure 3; Two-way ANOVA; $F(1, 12) =$
332 1.00 , $p = 0.34$). However, the observed variability (40–53% RSD) within treatments limited
333 further comparisons. No HBCDD in coral tissue was found in the control conditions.

334 The depuration experiment showed that coral exposed to the leachate eliminated HBCDD much
335 slower than coral exposed to the HBCDD alone (Figure 3; Two-way ANOVA; $F(1, 12) =$
336 10.10 , $p = 0.008$). Specifically, after 9 days of depuration in clean seawater (flow-through
337 condition), the concentration of HBCDD in the corals exposed to HBCDD alone decreased 4-
338 fold to $13 \pm 3 \text{ ng g}^{-1} \text{ dw}$ ($p = 0.007$). This is contrary to corals exposed to the leachate, which
339 had no significant loss of HBCDD after the depuration phase (final concentration of $20 \pm 6 \text{ ng}$
340 $\text{g}^{-1} \text{ dw}$; $p = 0.628$). This suggests an underlining effect from co-contaminants leached from the
341 PS.

342 Diastereoisomer analysis found α -HBCDD to be the dominant isomer in coral tissue (Figure 3),
343 with 98% of the Σ HBCDD after the 5 d exposure and 99.5% after the 9 d depuration period (a
344 value of half the limit of detection [LD/2] was used for the non-detected β - and γ - isomers).
345 This shift in isomeric profiles from 40% in the PS debris, to 90% in the leachate and 98% to

346 99.5% in the corals indicates isomer-specific transfer processes. If the higher aqueous solubility
347 of α -HBCDD (Covaci et al., 2006) can account for the PS-to-water transfer, preferential
348 accumulation of α -HBCDD in the coral tissues most likely relates to isomer-specific biological
349 processes. Selective accumulation of the most stable α isomer has been observed in fish species
350 (Du et al., 2012; Janák et al., 2005), where bioisomerization of γ -HBCDD to α -HBCDD seems
351 to be one of the reasons explaining the dominance of α -HBCDD in the majority of biota samples
352 (Zhang et al., 2014). Other studies involving *in vitro* assays with higher trophic level organisms
353 showed that the cytochrome P450 system was involved in the relatively rapid
354 metabolism/degradation of γ -HBCDD, whereas the α -isomer was not significantly
355 biotransformed. These different reactivities of the stereoisomers with the enzymatic system is
356 a likely process to explain the exclusive accumulation of α -HBCDD in the organisms (Zegers
357 et al., 2005). In this respect, the enrichment of the α -HBCDD in the coral tissues could also be
358 explained by a xenobiotic response of the enzymes P450s present in *Stylophora pistillata*. Other
359 environmentally induced changes in cytochrome P450s activity were observed in the coral
360 *Stylophora pistillata* after exposure to hyposaline conditions (Downs et al., 2009).

361

362 **3.4 Physiological effects of PS leachate-derived HBCDD on corals**

363 Despite a rapid uptake of HBCDDs by the corals, the PS leachate had no significant effects on
364 measured physiological parameters (i.e., photosynthetic activity, respiration, symbiont density
365 and pigmentation) after a 5-d exposure (Figure 4). We did however observe a slight 1.4-fold
366 increase in symbiont density in corals exposed to the HBCDD alone compare to controls (One-
367 way ANOVA, $F(3, 12) = 7.51$, $p = 0.0043$). Differences in physiological responses between
368 the two treatments indicate that other constituents of the leachate are having an effect on coral.
369 Moreover, a consistent polyp retraction was observed in coral nubbins exposed to the PS
370 leachate, but not in those exposed to HBCDD alone further supporting that the corals were

371 likely stressed by another constituent of the leachate. Polyp retraction can be a mechanism to
372 reduce the available surface area for contaminant uptake as previously observed on marine
373 invertebrates after exposure to tributyltin (TBT) or heavy metals (Harland and Nganro, 1990;
374 Smith et al., 2003). Here, polyp retraction had no influence on HBCDD uptake by the corals as
375 similar HBCDD concentrations were detected in both corals either exposed to PS leachate and
376 HBCDD alone. Whether polyp retraction affects the uptake of other constituents leaching from
377 PS will require further investigations. In addition to a potential reduced contaminant uptake,
378 contracted polyps are not able to feed on zooplankton, have reduced photosynthetic rates on the
379 long term and are diffusion-limited for gas and dissolved nutrient exchange with the
380 surrounding seawater (Lasker, 1981; Patterson, 1992). Thus, pollution-induced polyp retraction
381 can have implications for the coral's energy reserves and colony growth over time and may
382 become particularly severe considering the chronic exposure of marine organisms to the
383 increasing amount of plastic debris and its chemical additives in the oceans. Although the
384 chemical composition of our tested leachate remains unknown except for HBCDD, other
385 studies have investigated the organic and metallic additives found in leachates (Capolupo et al.,
386 2020; Thaysen et al., 2018). These studies highlight their complexity and high variability
387 between polymer types and samples, and tentatively identified compounds including polymer
388 precursors, antimicrobials, lubricants, antioxidants, antifungals, plasticizers, dyes, UV
389 stabilizers, solvents but low levels of volatile compounds. Similarly, the leachates (including
390 PS's) were found to exhibit toxicity to microalgae, aquatic invertebrates or mussels, but the
391 attribution of effects to individual compounds remained challenging.

392

393 **Conclusion**

394 This study confirmed that PS debris collected from Mediterranean beaches can release high
395 amounts of HBCDD into the seawater and over relatively long periods of time. These leached

396 brominated flame retardants were readily bioaccumulated by corals, with a relative slow
397 depuration. The leaching, transfer and accumulation of HBCDD from the beached debris to the
398 coral was found to be isomer specific, with α -HBCDD prevailing over the other isomers.
399 Although, no significant effects on coral photosynthetic activity, symbiont concentration and
400 chlorophyll content were observed during the short-term exposure to the PS leachate, the polyp
401 retraction in nubbins exposed to this treatment indicates that corals were stressed by another
402 constituent of the leachate. This might include PS nanoparticles generated by breakdown of the
403 polymer or styrene polymerisation by-products from PS resin manufacturing. Further work is
404 needed to provide an exhaustive chemical characterisation of these complex leachates through
405 a Non-Target Screening approach. Given the recent reports of plastic pollution in coral
406 ecosystems (Connors, 2017; Lamb et al., 2018; UNEP, 2019), more research is needed to
407 elucidate the stress response of coral exposed to chemicals leached from PS foams.

408

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421

422 **Supplementary materials**

423 **Table S1 and S2.** Analytical method parameters.

424 **Figure S1.** Accuracies of the analytical methods.

425 **Figure S2.** FTIR spectra of the 17 plastic samples.

426 **Figure S3.** Results of preliminary HBCDD kinetics experiment showing the behaviour of the
427 compound within the test system in the presence or absence of coral.

428 **Figure S4.** Concentrations of HBCDD in corals after exposure to HBCDD in water for 24 h
429 (preliminary experiment).

430

431

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599

600 **Captions:**

601 Figure 1. (a) Beached plastic macrodebris collected along the French Riviera coast; (b) PS
602 leaching experimental design.

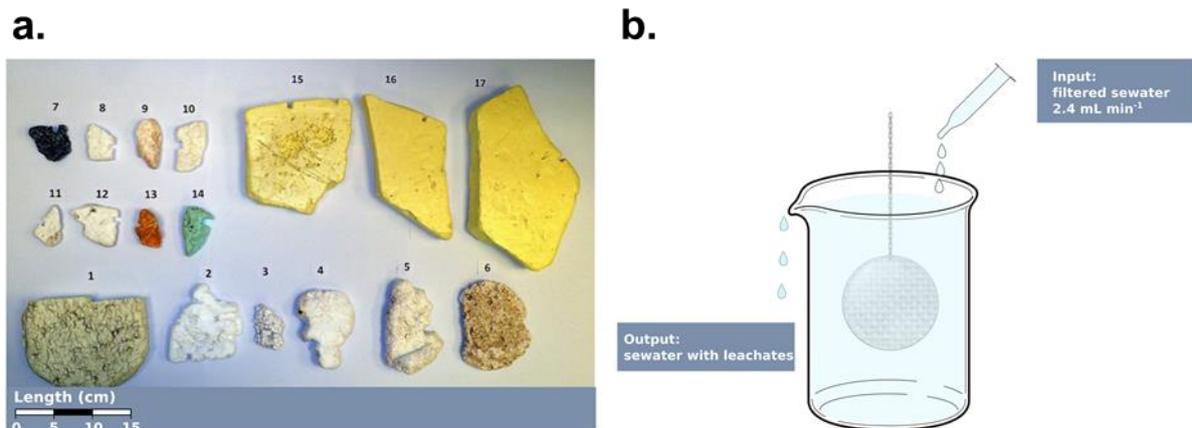
603 Figure 2. Concentrations and isomeric profiles of HBCDD (a) and concentrations of phenolic
604 compounds (b) found in the debris (n = 1 to 4).

605 Figure 3. Diastereoisomer compositions of HBCDD in coral after 5 days of exposure (left) and
606 retention after 9 days of depuration (right). Bars are mean \pm SD (n = 4). Asterisks indicates
607 significant differences between Σ HBCDD concentrations in coral post-exposure and post-
608 depuration.

609 Figure 4. Gross photosynthesis, chlorophyll content, respiration and symbiont density in the
610 coral *Stylophora pistillata* after 5 days of exposure to either control (C), MeOH solvent
611 control (SC), leachate or HBCDD spiked seawater. Boxplots show max and min (whiskers),
612 mean (+), median (line). Letters indicate significant differences between treatments.

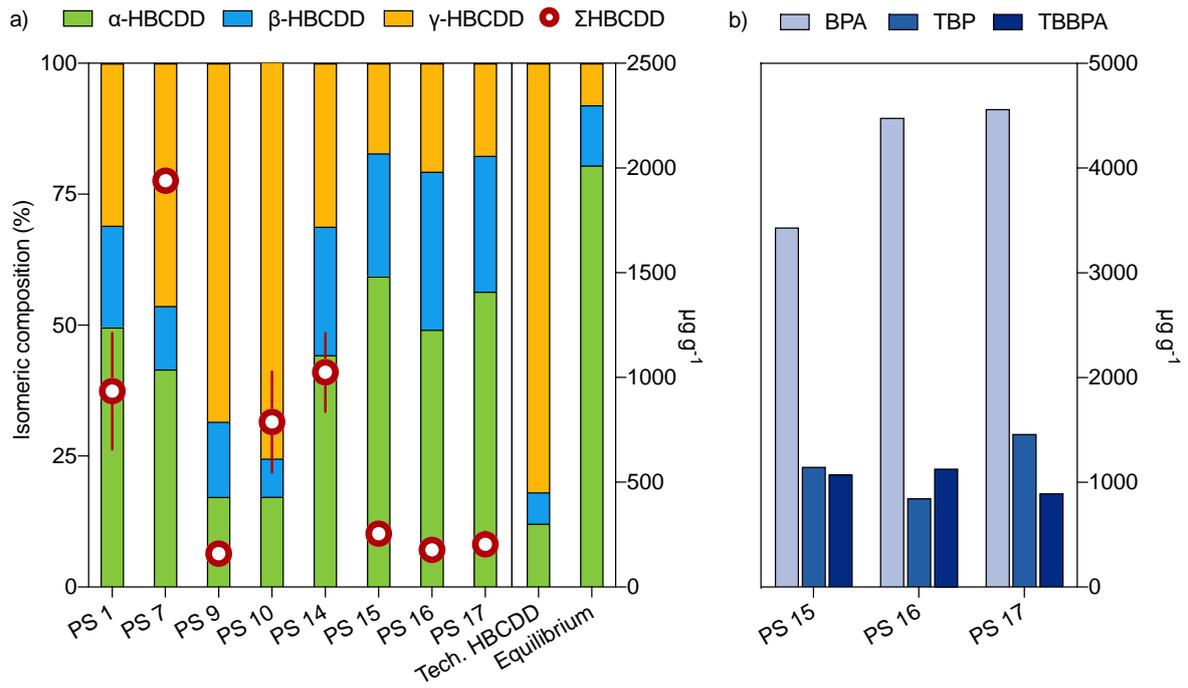
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614 **Figures:**



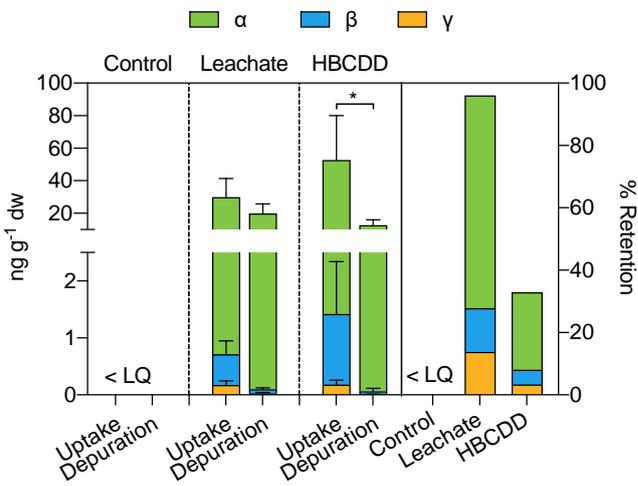
615
616 Figure 1.

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618
619 Figure 2.

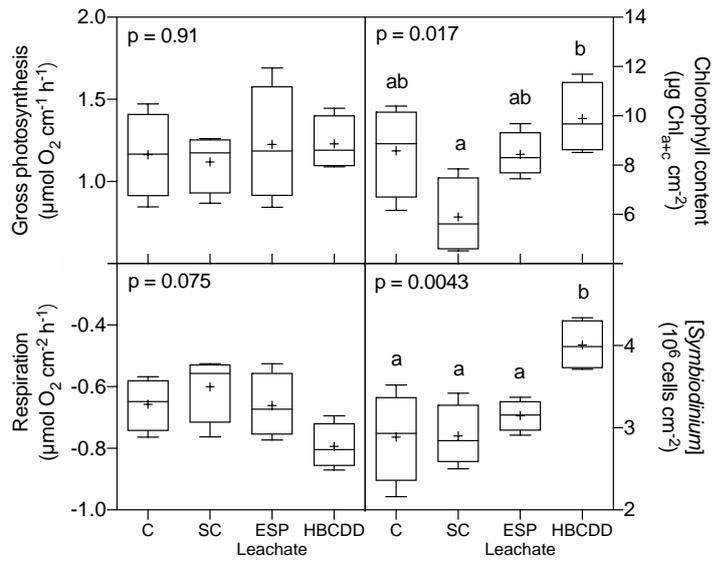
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622 Figure 3.

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626 Figure 4

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