Assessing bioaccumulation potential of personal care, household and industrial products in a marine echinoderm (*Holothuria tubulosa*)

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ABSTRACT A bioaccumulation study of 16 emerging contaminants including preservatives, UV-filters, biocides, alkylphenols, anionic surfactants and plasticizers, in *Holothuria tubulosa* Gmelin, 1791 specimens was developed. Water and sediments from their coastal habitat were also analyzed. Sediment-water distribution coefficients (log Kd) were in the range 0.78 to 2.95. A rapid uptake and bioaccumulation of pollutants was found. Compounds were detected in intestine and gonads of *H*. *tubulosa* after only eight days of exposure. Field-based bioconcentration (BCF) and biota-sediment accumulation factors (BSAF) were calculated. Log BCF > 1 were obtained for most of the compounds studied, indicating their tendency to accumulate in tissue of *H*. *Tubulosa*. BCF values decrease as follow: Triclocarban > anionic surfactants > benzophenone 3 > non-ionic surfactants > bisphenol A > parabens. These data provide a detailed accounting of the distribution patterns of some emerging contaminants in organisms at the lower trophic level, representing a potential source of contaminants for organisms in higher levels of the food chain.

Keywords: Personal Care products; Household and industrial chemicals; *Holothuria tubulosa*; Environmental partitioning; Bioaccumulation studies; Marine pollution

1. Introduction

Contaminants of emerging concern (CECs) such as preservatives, UV-filters, biocides, surfactants, and others are being increasingly detected in the world's marine environments (Negri and Marshal 2009; Richir et al., 2015; Ruhí et al., 2016; Díaz-Cruz et al., 2019). There are CECs from multiple sources including domestic, commercial, hospital and industrial wastewater discharges as well as from agriculture and aquaculture activities (Wilkinson et al., 2017). Although the consequences of these pollutants are becoming increasingly apparent (Rahman Kabir et al., 2015; Meador et al., 2018), their impact on ecosystems is poorly known, particularly concerning the possibility of bioaccumulation in lower trophic levels.

Research on the accumulation of contaminants, especially of CECs, in aquatic organisms are crucial to support and provide the Marine Strategy Framework Directive (MSFD, 2008/56/EC; OSPARCOM 2012) with the robust scientific knowledge. In this respect, in-depth research on the complex interactions between water, sediment, and biota is needed to better understand the fate of ~~EC~~CECs and their effects on the aquatic environment (Xu et al., 2014; Ruhí et al., 2016; Beyer et al., 2017; Koba et al., 2018; Sanganyado et al., 2018; Rocha et al., 2018; Wilkinson et al., 2018; Zhang and Kelly, 2018). Quantification of pollutant accumulation has been calculated in terms of several bioaccumulation metrics including the bioaccumulation factor (BAF), the biomagnification factor (BMF), trophic magnification factor (TMF), and biota-sediment accumulation factor (BSAF) (Diepens et al., 2015; Peng et al., 2017; Koba et al., 2018; Rocha et al., 2018).

Variations of these parameters are related to species specific characteristics such as diet, feeding habits, habitat, sizes, gender, metabolic capacity, and trophic levels (Diepens et al., 2015; Peng et al., 2017). Peng et al. (2017) studied the bioaccumulation and biomagnification of 13

organic UV absorbents in marine organisms and reported that direct uptake from the growth media was a key route of exposure to these absorbents (BSAF 0.003-2.152). They also found that bioaccumulation, in estuarine organisms, was partially related to the type of compound and the species, with higher bioaccumulation in fish than invertebrate species (except for benzophenone 3). Recently, Martín et al. (2019) reported log BAFs and BSAFs values > 1 for six perfluoroalkyl compounds in *Holothuria tubulosa* specimens. Wilkinson et al. (2018) investigated the accumulation of plasticisers, illicit drugs, pharmaceuticals and PFAS in river sediments, aquatic plants and benthic organisms and reported different levels of bioaccumulation indicating that there may be a need for a species-specific BCF/BSAF classification system.

Despite this, in-depth research on the bioaccumulation behaviour of some CECs like anionic surfactants such as linear alkylbenzene sulfonates (LAS) and alkylsulfates (AS) in organisms from the lower levels of the trophic system through the measurement of field-based factors is still limited. Holothurians can be used as a suitable proxy of CECs bioaccumulation and toxicity in marine ecosystems since these filter-feeders are found in coastal areas all over the world and are therefore highly likely to be exposed to anthropogenic xenobiotics. These species filter the water around and the sandy substrate. Also are edible species and threatened by illegal fishing (Xing and Chia, 1997; Warnau et al., 2006; Sugni et al., 2007; Jiang et al., 2015; Martín et al., 2017; Martín et al., 2019).

Against this background, the main purpose of the present study was to assess the bioaccumulation behaviour of selected emerging contaminants using sea cucumbers (*Holothuria tubulosa* Gmelin, 1791) in a laboratory model. A total of 16 target compounds including three preservatives, one UV-filter, two biocides, two non-ionic surfactants (alkylphenols), seven anionic surfactants (LAS and AS), and one plasticizer were chosen because of their environmental representativeness, occurrence, persistence and ecotoxicological risks. The

duration of the experimental studies was 197 days to determine the relationship between CECs concentrations in specimens of *Holothuria tubulosa*, water, and sediment. Partition coefficients (K_d) were used to determine the relative migration of pollutants between the aquatic and the solid phase; additionally, the BASF and BAF were also calculated using tissue from the *Holothuria* specimens and compared with measurements from other aquatic organisms.

2. Materials and methods

2.1. Standards and reagents

Analytical grade reagents were used for all experiments, unless otherwise specified. A commercially available mixture of LAS (C10 to C13) was provided by CEPSA Química (Spain). The sodium salts of AS (AS-C12; AS-C14 and AS-C16) were supplied by Alfa Aesar (Barcelona, Spain). 4-nonylphenol (4-NP) technical grade mixture of isomers; nonylphenol monoethoxylate (NP_1EO); the personal care products methylparaben (MeP), ethylparaben (EtP), propylparaben (PrP) and benzophenone 3 (BP3); the biocides triclosan (TCS) and triclocarban (TCB); the plasticizer bisphenol A (BPA); the solvents methanol, ethanol, water, acetone and acetonitrile; and octadecyl functionalized silica (C18) were from Sigma-Aldrich (Steinheim, Germany). Ammonium acetate was purchased from Panreac (Barcelona, Spain).). Finally, labelled bisphenol A-d_{16} (BPA-d_{16}), benzophenone-d_{10} (BP-d_{10}) and ethylparaben-d_5 (EtP-d_5) were obtained from Cambridge Isotope Laboratories (MA, USA). Working solutions were mixtures of compounds and were prepared by diluting stock solutions with ethanol:water 5:95 (v/v). Test kits from Hanna instruments® (3874 for nitrate, 3873 for nitrite, and 3826 for ammonium) were used for determination of nitrogen species in water.

118 *2.2. Specimen collection and acclimation*

119 The average weight of each *Holothuria tubulosa* Gmelin, 1791 specimen was approximately
120 200 g. The specimens were collected by SCUBA divers at 10-15 m depth along the southern
121 Mediterranean coast of Spain in May 2016. The animals were transported in containers at 4 ºC to
122 the laboratory where they were placed into the experimental tanks and left for two weeks as
123 depuration period and for acclimation to the experimental conditions.

124

125 *2.3. Experimental conditions*

126 The aquarium facilities are located in the Zoology Department of the University of Granada
127 (Spain) where three independent experimental units were placed. Each unit consisted of a closed
128 recirculating system of sea water with 9 animal housing tanks. Units were equipped with an
129 ultraviolet sterilization unit, a filter, a skimmer, a thermostat and air and water pumps. The
130 dimensions of the tanks were 42 x 36 x 25 cm, having a capacity of 25 L and a bottom surface of
131 0.15 m^2 which was covered with a 5 cm thick sediment layer of clean commercial silica sand
132 (Productos QP S.A.) of 0.4-0.8 mm particle size enriched with 10 g minced dry *Laminaria* algae
133 (ALGAMAR® Kombu). Once a week, this amount of algae was added to the sediment in order to
134 provide food for the animals, that were maintained in 12 h light-12 h dark cycles. During the
135 experiment, the physicochemical conditions of the water were daily monitored to ensure they
136 were stable [T = 20 ± 2ºC, pH = 8.3± 0.5, salinity = 37 ± 2 PSU (conductivity = 50.3 ± 0.9 mS
137 cm^{-1}), dissolved O_2 = 6.0 ± 0.5 mg L^{-1} (90 % saturation level), ammonium = 0.25± 0.04 mg L^{-1},
138 nitrites = 0.10 ± 0.01 mg L^{-1}, nitrates = 50 ± 4 mg L^{-1}]. The average organic carbon content of the
139 tank sediments was 0.5 ± 0.1 % without significant differences between tanks and treatments.

140 The experimental research was designed as follow. A total of 108 sea cucumbers were
141 randomly distributed in the three experimental units placing 4 specimens per tank (36 per unit).

142 After the two-week acclimation, the pollutants were added to the water of each unit at 1.0 μg mL^{-1}
143 (Unit/Batch-1), 0.5 μg mL^{-1} (Unit/Batch-2) and 0.1 μg mL^{-1} (Unit/Batch-3). The selection of
144 these concentration levels was based on two experimental findings from our previous experience
145 in working with these bioindicators: the amount of contaminants found in wild *Holothuria*
146 specimens (Martín et al., 2017) and the need to identify a tendency and clear differences between
147 the experimental batches. The experiments and pollutants exposure time lasted six months and
148 sampling was carried out immediately prior to the incorporation of contaminants to the water and
149 at days 1, 8, 15, 22, 27, 38, 53, 86 and 197 of the experiment. At the beginning of the
150 experiments, samples taken before the addition of the contaminants to the water tanks (control)
151 were analysed and the practical absence of analytes was demonstrated (lower concentration than
152 the detection limit of the method).
153
154 *2.4. Sample pretreatment and analysis*
155 Three specimens, 20 mL of water and 2 g of sediment were taken by hand with latex gloves,
156 from each of the units at each sampling. Sampled animals were dissected to separate gonads and
157 digestive track which were frozen at -20 ºC until analysis. Samples were treated according to a
158 method described elsewhere with slight modifications (Martin et al., 2017).
159 *Holothuria* (gonads and intestine) and sediment samples were freeze-dried, homogenized and
160 grounded into powder. Aliquots of the samples (0.5 g) were weighed into 12 mL glass vials,
161 containing 100 μL of a methanol solution (250 ng mL^{-1}) of a mixture of ISs. The samples were
162 vortexed twice for homogenization in 7 mL of acetonitrile for 2 min and centrifuged for 10 min at
163 4050 × g. For clean-up, disperse solid phase extraction (d-SPE) was applied. The supernatants
164 obtained from the two extractions of each sample were combined in a 50 mL polypropylene
165 conical tube containing 800 mg of C18 sorbent. The mixture was hand-shaken for 2 min and

centrifuged for 5 min at 4050 × g. The solvent was evaporated to dryness at room temperature under a nitrogen stream and the extract was reconstituted in 0.25 mL of methanol:water (50:50, v/v) and filtered through a 0.22 μm nylon filter. A 20 μL aliquot of the extract was injected into the LC instrument.

Water samples were collected in brown bottles pre-cleaned with acetone and methanol. Prior to analysis, water samples were filtered through a 0.45 μm membrane filter. Subsequently 2 mL of sample, containing 100 μL of a methanol solution (250 ng mL^{-1}), was evaporated to dryness at room temperature under a nitrogen stream and the extract was reconstituted in 0.25 mL of methanol:water (50:50, v/v) and filtered through a 0.22 μm nylon filter. A 20 μL aliquot of the extract was injected into the LC instrument.

Liquid chromatography-tandem mass spectrometry analysis was performed using a HALO C-18 Rapid Resolution (50 × 4.6 mm i.d., 2.7 μm particle size) column. The method used here was previously optimised and validated in-house (Wilkinson et al., 2016b). Full and detailed HPLC-MS/MS protocol and method quality control measures used in this work are described described by Martin et al. (2017) and summarized in the Supplementary material.

3. Results and discussion

3.1. Contaminants distribution in sediment and water

The concentration of each compound in each of the batches was determined. As example, the fate profile over the whole experiment (197 days) of selected compounds in Batch-1 is shown in figure 1. Higher concentrations in water samples were found for the NP group of surfactants (30-83 ng mL^{-1}) and the ionic surfactants LAS and AS (1.13-75 ng mL^{-1}). In the case of the personal

189 care products MeP and BP3 and the biocide TCB the concentration levels ranged 0.07-3.2 ng mL^{-1}, 0.05-14.4 ng mL^{-1} and 0.06-0.14 ng mL^{-1}), while BPA and TCS were not detected in any water sample.

192 Our results show that the selected compounds exhibit more affinity for solid phases than for water. In Batch 1 all pollutants were detected in the sediment phase, except BPA. Overall, the selected surfactants were most found in sediment, especially NP (380-4027 ng g^{-1} d.w.) followed by the biocides TCB (71-308 ng g^{-1} d.w.) and TCS (5.1-320 ng g^{-1} d.w.), and to a lesser extent parabens (0.1-9.4 ng g^{-1} d.w.) (see Fig. 1). A slight increase in concentration levels was observed during the 197 days.

198 K_d were calculated as C_{sed} (contaminant in solid phase, ng g^{-1}d.w.) divided by dissolved phase C_w (contaminant in water volume, ng mL^{-1}) at equilibrium (See Table 1). Calculated rates revealed that TCB and AS C14 have a strong tendency to adsorb onto sediments (log K_d, = 2.95 and 1.76, respectively, in Batch-1). These high partition ratios indicate the strong adsorption of these compounds onto sediments, despite the fact that to some extent they are dissolved in the water compartment. Parabens showed the lower log K_d values (0.78-1.02). Overall, no clear relationships between the K_d and log K_{ow} were observed (Figure 2).

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206 *3.2. Bioacumulation of compounds in Holothuria tubulosa specimens*

207 The contaminant distribution between water and sediments may result in the transfer of these chemicals to upper trophic level species that feed on benthic organisms (Díaz-Cruz et al., 2019). Concentrations in *Holothuria tubulosa* measured before exposure were similar to the background levels found in the coastal areas of Granada (Martín et al., 2017) and they were about 100-fold lower than those found at day 8 of exposure in Batch-1. *Holothuria tubulosa* showed a high capacity to bioaccumulate the targeted chemicals, 100% detection frequency. Based on the

213 concentrations of contaminants detected during the experiments, the order of the most abundant
214 family of selected compounds to the least abundant was surfactants (AS, LAS and ≃ TCB and
215 TCS > BP3 > BPA > parabens (MeP, EtP, PrP).

216 The concentration levels of compounds in *Holothuria* specimens were ≥50-fold higher than
217 in water and sediment for some chemicals (BP3 or TCS). Sea cucumbers are often exposed to
218 pollutants by a contaminated ambient environment via ingestion. They ingest and defecate large
219 amounts of sediment and water and digest and absorb only the organic and living material. Since
220 these species live on soft sediments, being therefore benthic, are mostly contaminated by
221 sediments rather than water. Nonetheless, the uptake via water could be also probable and cannot
222 be ruled out. For all selected compounds, accumulation occurred rapidly after only 8 days. For
223 example, concentration levels for AS C12 in intestine and gonads were of 5.9 and 2.1 mg kg^{-1}
224 d.w, respectively, after eight days exposure to 1 μg mL^{-1}. BPA was detected in the intestine and
225 gonads of the organisms but not in water or sediment samples.

226 The concentration profiles in intestine shown in Fig. 1 indicate a decrease in the
227 concentration probably due to a natural degradation process in the biota. For example, AS C12
228 decreased from 5.9 mg kg^{-1} d.w. at day 8 to 0.34 mg kg^{-1} d.w. at day 197; TCB from 2.9 mg kg^{-1}
229 d.w. at day 8 to 0.48 mg kg^{-1} d.w. at day 197; and BPA from 0.77 mg kg^{-1} d.w. at day 8 to 0.007
230 mg kg^{-1} d.w. at day 197. This effect was observed in Batch-1 (spiked with the higher
231 concentrations). Moreover, no significant differences were found between Batch-2 and -3 (spiked
232 with lower concentration) for most of compounds. A study of these characteristics is rather
233 complex since *H. Tubulosa* absorbs the pollutant from the ambient environment being the net
234 result of competing rates of chemical uptake at the respiratory surface and chemical elimination
235 including respiratory exchange, fecal egestion, metabolic biotransformation of the parent

236 compound, and growth dilution (Arnot and Gobas, 2006).

237 In the gonads of the *Holothuria* specimens this decrease was also observed but it seemed to
238 occur at random during the experiments, without a clear tendency. No significant differences
239 were found between female and male gonads. The general high concentrations of some target
240 pollutants at the end of the trial could be explained by the fact that the degradation of target
241 compounds from the intestine occurs more rapidly respect to gonads.

242 Literature on the bioaccumulation of environmental contaminants in benthic invertebrates, of
243 surfactants in particular, is rather scarce and this topic deserves further research. Gatiduo et al.
244 (2010) evaluated the bioconcentration potential of NP, TCS and BPA in Mediterranean mussel
245 (*Mytilus galloprovincialis*) and found concentrations between <LOD and 914 ng g^{-1} d.w. for NP,
246 <LOD and 2578 ng g^{-1} d.w. for TCS and <LOD and 612 ng g^{-1} d.w. for BPA. During the intake
247 period (28 days of exposure to 300 μg L^{-1}) tissue concentrations of the target compounds
248 increased. Subsequently the concentration decreased during a depuration period in clean water.
249 However, at day 56 (end of depuration) the residual concentrations of chemicals were higher than
250 their initial concentrations. Similar results were reported by Vidal-Liñán et al. (2017) who
251 assessed the bioaccumulation of UV filters in mussel tissues. The uptake and accumulation
252 occurred rapidly with concentrations of up to 418 ng g^{-1} d.w. 24 h after exposure to 1 ng mL^{-1} to
253 the contaminants. This concentration naturally decreased during the experiments. More recently,
254 Ademollo et al. (2018) studied the bioaccumulation of NP, NP1EO and BPA in the Greenland
255 shark (*Somniosus microcephalus*) and found higher contamination levels in muscle (mean
256 content 20.3, 171.1 and 7.9 ng g^{-1} wet weight for NP, NP1EO and BPA, respectively) and liver
257 (mean content 43.5, 288.5 and 8.2 ng g^{-1} wet weight for NP, NP1EO and BPA, respectively).
258 Maulvault et al. (2018) assessed the effects of seawater temperature and pH on the
259 bioaccumulation of CECs (flame retardants, inorganic arsenic, and PFAS) in marine bivalves.

Their results showed that when both environmental conditions act in combination with the before bivalves' capacity to accumulate contaminants may be time-dependent. Changes in the bioaccumulation and removal patterns of some contaminants also suggest an increase of adverse effects on humans if global warming, ocean acidification and other environmental conditions linked to climate change persist. Álvarez-Muñoz et al. (2015) studied the occurrence of parabens, BPA and TCS in macroalgae, bivalves, and fish from coastal areas and found no contaminants in macroalgae samples, however, the analysis revealed their presence in bivalves and fish in concentrations up to 16.4, 12.5, and 1.5 ng g^{-1} d.w. for MeP, BPA, and TCS respectively.

Holothuria tubulosa is an extremely valuable sentinel of environmental quality as this species meets the criteria set by the OSPAR Commission for the selection of species that can be monitored for determining contaminant concentration in the surrounding ecosystem.

3.3. Field-based bioconcentration (BCFs) and biota-sediment accumulation factors (BSAFs)

BCF (Equation (1)) and BSAF (Equation (2)) values were calculated based on that described by Arnot and Gobas (2006). Bioaccumulation was classified using three categories: 'not bioaccumulative,' 'bioaccumulative' and 'very bioaccumulative.' Classifications were selected based upon criteria established by different government guidelines/acts (see Wilkinson et al. (2018): 1). A bioaccumulative BCF was considered a log value between 3.0 (1000 L kg^{-1}) and 3.7 (5000 L kg^{-1}) while a 'very bioaccumulative' log BCF value was considered anything ≥ 3.7 (5000 L kg^{-1}).

The BCF was calculated as the ratio of the contaminant concentration in the intestine or gonads of holothurians (C_{hol}, expressed in ng g^{-1} d.w.) and the contaminant concentration in the water (C_w, expressed in ng mL^{-1}) at equilibrium. The BSAF is calculated as the ratio between the chemical concentration in intestine or gonads of specimens (C_{hol}, ng g^{-1} d.w.), and the

284 concentration in sediments (C_{sed}, ng g^{-1} d.w.). Concentrations of studied analytes in the studied
285 water and sediment were determined at the same time and at the same locations that all biota
286 samples were collected as part of a concurrent study. The range and mean of both factors
287 calculated in the three batches are shown in the Table 1.

288 The pollutants with the highest BCF values to those with the lowest values are TCB >
289 anionic surfactants > BP3 > non-ionic surfactants > BPA > parabens. For most of the target
290 compounds Log BCF > 1 were obtained, except for EtP–(0.51-0.57) and PrP (0.38-0.47),
291 indicating their tendency to accumulate in tissue of *H. Tubulosa*. Nevertheless, according with
292 the bioaccumulation categories guides, of mean log BCFs (Table 1), a total of 15 out of 16
293 compounds can be classified as "non bioaccumulative" (log BCF < 3) while TCB can be
294 classified as a very bioaccumulative compound in the intestine (4.14).

295 For parabens, the estimated log BCF was dependent on the corresponding concentration in
296 water. In particular, BCF values in Batch-1 were sometimes significantly lower or higher than
297 those found in Batch-2 or 3. A similar behaviour has been reported in the literature when assessed
298 the bioaccumulations of PFCs in similar studies using varied species such as H. tubulosa (Martín
299 et al., 2019), fishes, bivalves, crabs, gastropods, shrimps, starfish, and polychaetes (Hong et al.,
300 2015). In batches at lower concentration of target substances, BCF values might be
301 underestimated because of the possible overestimation of freely dissolved compound in water.

302 The log K_{ow} gives a theoretical proxy for bioaccumulation potential (MacKay, 1982). For
303 example, the high BCF values found for TCB (4.14) and BP3 (2.35) correlate with their
304 respective log K_{ow}: 4.93 for TCB and 3.79 for BP3 (3.79). However, no clear relationships were
305 found between BCF values and log K_{ow} for the rest of before analytes (Figure 2). Overall, the log
306 K_{ow} ranged from 1.7 for MeP to 6.2 for AS C16 when both chemicals showed a similar
307 experimental BCF in *H. tubulosa*. A similar situation has been reported for UVA in marine

308 wildlife (fish, cephalopoda and crustaceans) (Peng et al., 2017), pharmaceuticals, BPA and TCS
309 in aquatic macrophyte and macroinvertebrates (Ruhí et al., 2016; Pi et al., 2017), benzophenones
310 in mussels (Vidal-Liñán et al., 2018). Vidal-Liñán et al. (2018) reported rapid uptake and
311 accumulation of organic UV filters in mussels and found that measured bioaccumulation of BP-4
312 was much higher than predicted by K_{ow}-based models. This demonstrates the necessity of using
313 experimental models in order to prevent underestimation of risk.

314 Since *Holothuria tubulosa* is a benthic bioindicator of pollution that feeds on the organic
315 matter absorbed onto sediments, BSAF would be the most useful parameter to describe and
316 predict bioaccumulation in biota through sediments (Labadie and Chevreuil, 2011; Zhao et al.,
317 2014). As shown in Table 1, log BSAF values vary considerably, from -0.13 (NP) to 1.75 (BP3)
318 and from -0.61 (NP) to 1.55 (EtP) in Batch-1 for intestine and gonads, respectively. In general,
319 BSAF-values were lower than BAF-values in this work indicating that water concentrations are
320 simply too low in these study compared to those in the sediment for accurate estimation. For
321 example, the biocides TCB and TCS, once of the selected compounds detected at higher
322 concentrations in *H*. *Tubulosa*, were not found or detected at very low concentrations in the water
323 samples.

324 The use of BCFs, BAFs and BSAFs for the evaluation of bioaccumulation of organic
325 contaminants in marine environments is still limited and measured primarily fish. Table 2 lists
326 the BAF and BSAF values compiled from database sources for different marine species. In
327 addition im comparison with BAF, BSAF reported values are very limited. Reported results
328 revealed the differential bioaccumulation behaviour of contaminants occurring in different biota.
329 While the evaluated data still contains sources of error, it is important to acknowledge there are
330 also sources of inherent variability. Some possible sources resulting in observed variability in the
331 BAF for individual chemicals among experiments are: lipid content, organism size, metabolic

transformation, organic carbon in water, temperature, pH. According with a complete review study carried out by Arnot and Gobas (2006) on BCF and BAF assessments for organic chemicals in aquatic organisms, those organisms with higher lipid contents have a greater capacity to store hydrophobic organic chemicals and therefore can exhibit a higher BAF. Larger organisms have slower elimination rates and may feed at higher trophic levels. Trophic position is a key factor influencing the BAF as observed.

Most of the studies available focus on NP and BPA determination. Surfactants are amongst the most common contaminants of water bodies, being LAS one of the most used anionic surfactant on the market (Könnecker et al., 2011; Alves et al., 2016; Martín et al., 2017). Its acute and chronic toxicity are well documented, however, to the best of our knowledge, there are no data regarding BAF and BSAF estimations for the anionic surfactants (LAS and AS mixtures). Procter and Gamble (2002–2004) investigated the bioconcentration of AS C14-C15 in fish (fathead minnow, channel catfish) and invertebrates (*Corbicula fluminea* [Asiatic clam]) and found bioconcentration factors ranging 180-422, 402-972 and 81-400 L kg^{-1} for fathead minnows, catfish and clams respectively, with values for AS C15 being 6.5 times greater than for AS C14. On the determination of bioconcentration factors in fathead minnow specimens exposed acutely (4 days) and chronically (33 days) to C12 AS, the detected values were 1-4 L kg^{-1}. These results suggest some chain length dependency and inter-species differences associated ~~to~~with the bioaccumulation of AS.

There are only a few studies that have investigated the bioaccumulation behaviour of TCS and TCB with inconsistent results. For example, Vimalkumar et al., (2018) reported log BAF values for TCB ranging from -1.70 to 1.19 in fish species from Indian rivers while Yao et al. (2018) reported log BAF values from 1.31 (Tilapia sp.) to 5.35 (common carp sp.) in different species of wild fish from Chinese rivers. These values are in the range of those obtained in our

356 study (3.52-4.48 in intestine and 1.71-4.11 in gonads). Nevertheless, as mentioned above,
357 comparison of results is difficult because BAF can be affected by the test conditions, the species
358 used or the exposure concentration of the target compounds (Gatidou et al., 2010). Moreover,
359 BAF values reported in some studies are expressed as dry weight, while other studies express
360 their data as wet weight. Pi et al. (2017) found in their laboratory experiments conducted on two
361 aquatic macrophyte species that TCS showed high uptake and bioaccumulation potential (log
362 BAF 3.64) but relatively low BPA (0.30-2.17).

363 Salgueiro González et al. (2015) reported a high affinity of NP for sediments and the
364 molluscan species *C. fluminea* (BSAF=1.63-3.57) while Zhang et al. (2011) reported a relatively
365 low bioaccumulation of NP (BSAF=0.05-0.40) in short-necked clam and black seabream.
366 Bioaccumulation of NP was not as relevant in our study where mean BSAF values of -0.23 and -
367 0.48 were detected in intestine and gonads of *H. Tubulosa* specimens.

368 These results have revealed the differential bioaccumulation behaviour of contaminants
369 occurring in different biota, which may suggest the need for a species-specific BAF/BSAF
370 classification system as demonstrated recently elsewhere (e.g., Peng et al., 2017; Zhang and
371 Kelly, 2018) warranting further research.

372

373 ## 4. Conclusions

374

375 This study provides new insights on the distribution and bioaccumulation potential of
376 different classes of CECs like preservatives, UV-filters, biocides, surfactants, and plasticizers.
377 This is the first application of bioaccumulation model in marine ecosystem for some of the target
378 analytes such as LAS or AS.

379 Higher levels of contaminants were found in the intestine and gonads of *Holothuria tubulosa*
380 specimens than in water or sediments, which highlights the need for special attention to the
381 potential biomagnification effects on these species. However, according with the
382 bioaccumulation categories guides, a total of 15 out of 16 compounds can be classified as "non
383 bioaccumulative" (log BCF < 3) while TCB can be classified as a very bioaccumulative
384 compound in the intestine (4.14). The CECs concentration profiles in intestine showed a natural
385 decrease over the duration of the experiments. No clear relationship was observed between
386 physicochemical properties (log K_{ow}) and bioaccumulation behaviour, suggesting the need to
387 validate the modelled bioaccumulation values with experimental data in order to provide an
388 effective risk assessment. It is our hope that these findings would shed some insights into the
389 behaviour of ~~EC~~CECs in environmental and biological systems and provide essential information
390 to support risk assessment and management of the large and increasing number of chemicals that
391 may enter the environment. Future work may focus on the modelling of contaminant uptake and
392 transformation mechanisms in aquatic organisms and communities.
393

394 ## Acknowledgements

395

396 The present study has been supported by Project No. CTM2017-82778-R of the Spanish Ministry
397 of Economy and Competitiveness.
398

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Figure Captions

Fig. 1. Fate profile of EC in the high exposure tank during the 197 day experiments. ♦: water samples: □; sediment samples; ▲: intestine samples; ○: female gonad samples; ●: male gonad biota samples.

Fig. 2. Relationships between log K_d (left), log BAF (middle), and log BSAF (right) with log K_{ow} of the selected EC.

Table

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Table 1. K_d (n=27), BCF (n=15) and BSAF (n=15) values of CEC measured in *Holothuria tubulosa* at three exposure concentrations.

Batch	**log Kd**		**log BCF (intestin)**		**log BCF (gonads)**		**log BSAF (intestin)**		**log BSAF (gonads)**	
Batch-1 (1 mg L^{-1})	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**
ASC12	0.86-1.94	1.32	0.65-2.86	2.17	0.90-2.83	2.09	-0.21-1.61	0.99	0.04-1.73	0.89
ASC14	1.41-2.13	1.76	1.26-3.06	2.36	1.41-3.04	2.36	-0.23-1.49	0.73	-0.32-1.45	0.62
ASC16	1.24	1.24	1.71	1.71	1.59	1.59	0.05-1.85	0.98	0.01-1.50	0.57
LAS C10	0.78-1.54	1.13	0.81-2.66	1.99	0.89-2.70	1.91	-0.73-1.88	0.84	-0.65-1.92	0.90
LAS C11	0.74-1.81	1.24	0.96-2.55	2.01	1.03-2.61	1.96	-0.60-1.64	0.78	-0.53-1.70	0.90
LAS C12	0.68-1.49	1.07	0.84-1.80	1.50	1.02-2.31	1.67	-0.65-1.07	0.44	-0.47-1.33	0.76
LAS C13	-	-	1.64	1.64	1.31-2.41	1.95	-	-	-	-
NP	0.73-1.69	1.11	0.22-1.56	1.01	-0.18-1.53	0.59	-1.47-0.55	-0.23	-1.58-0.69	-0.48
NP1EO	0.82-2.22	1.24	-2.62	1.52	-0.54-2.21	1.04	-0.99-1.22	0.37	-1.36-0.97	-0.03
MeP	0.36-1.40	1.02	0.02-3.34	1.73	-0.03-3.06	1.90	-0.34-1.94	1.01	-0.39-1.93	0.68
EtP	0.91	0.91	0.51	0.51	0.57	0.57	-0.41-2.22	1.44	-0.34-3.55	1.49
PrP	0.78	0.78	0.38	0.38	0.47	0.47	-0.40-1.64	0.95	-0.31-2.77	0.92
TCB	2.48-3.39	2.95	3.52-4.48	4.14	1.71-4.11	2.93	0.67-1.96	1.19	-1.34-1.59	0.02
TCS	-	-	-	-	-	-	0.07-2.12	1.13	-0.99-2.23	0.55
BPA	-	-	-	-	-	-	-	-	-	-
BP3	0.32-1.42	0.98	0.21-3.58	2.35	-0.13-2.81	1.90	-0.14-2.93	1.71	-0.48-2.66	1.15
Batch-2 (0.5 mg L^{-1})	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**
ASC12	0.58-1.58	1.21	1.73-2.31	2.07	1.59-2.51	1.91	0.65-1.66	1.03	0.33-1.23	0.77
ASC14	0.23-2.20	1.42	1.95-2.56	2.19	1.73-2.33	2.03	0.36-2.09	0.98	0.13-1.21	0.59
ASC16	0.12	0.12	2.07	2.07	-	-	0.22-1.95	0.90	0.09-0.84	0.46
LAS C10	0.54-1.41	1.00	1.23-2.54	1.78	1.32-1.91	1.53	0.31-1.51	0.89	0.44-0.75	0.57
LAS C11	0.16-1.52	1.00	1.30-2.21	1.69	1.27-1.97	1.57	0.51-1.34	0.85	0.48-0.80	0.59
LAS C12	0.08-1.21	0.82	0.84-1.58	1.17	1.10-1.55	1.31	0.16-1.11	0.49	0.41-0.74	0.51
LAS C13	1.66	1.66	1.67-2.88	2.35	2.19-2.54	2.37	1.10-1.22	1.16	0.88-1.13	1.01
NP	0.62-1.06	0.90	0.31-1.79	0.82	0.20-0.60	0.36	-0.52-0.76	-0.13	-0.86--0.26	-0.61
NP1EO	0.29-1.49	0.90	0.62-1.49	1.02	0.47-1.34	0.89	-0.11-0.60	0.18	-0.32-0.15	-0.04
MeP	1.27-1.84	1.55	1.89	1.89	-	-	0.34-1.04	0.63	0.30-0.98	0.65
EtP	1.49	1.49	1.83	1.83	-	-	0.34-2.04	1.15	1.02-2.30	1.55
PrP	1.19	1.19	1.69	1.69	-	-	0.17-0.90	0.49	0.58-1.49	0.94
TCB	1.45-3.45	2.52	3.12-3.91	3.52	2.08-3.91	3.04	-0.05-1.55	0.87	0.15-1.38	0.67
TCS	-	-	-	-	-	-	0.56-1.63	0.91	-0.10-0.73	0.28
BPA	-	-	-	-	-	-	-	-	-	-
BP3	-0.17-1.91	1.04	0.92-4.83	2.79	1.85-3.38	2.54	0.51-3.45	1.74	0.59-2.00	1.29

Table 1 cont. K_d (n=27), BCF (n=15) and BSAF (n=15) values of CEC measured in *Holothuria tubulosa* at three exposure concentrations.

Batch-3 (0.1 mg L^{-1})	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**
ASC12	0.47-1.81	1.26	0.65-2.62	1.89	1.66-3.07	2.15	0.18-0.81	0.59	0.03-1.40	0.68
ASC14	0.19-2.45	1.52	0.65-2.52	1.87	1.04-2.90	2.09	-0.05-0.57	0.30	-0.23-1.29	0.24
ASC16	-0.70-0.74	0.02	0.59-1.14	0.87	0.65-2.46	1.56	-0.57-1.29	0.30	-1.27-1.72	0.04
LAS C10	0.37-1.56	0.98	0.46-2.35	1.42	1.19-2.12	1.69	0.07-1.13	0.46	0.00-1.27	0.60
LAS C11	-0.02-1.57	0.96	0.43-2.31	1.39	0.95-1.96	1.62	0.02-0.92	0.47	-0.08-1.25	0.52
LAS C12	-0.17-1.37	0.84	0.02-1.69	0.90	0.63-1.68	1.39	-0.27-0.43	0.12	-0.14-1.20	0.42
LAS C13	-0.17-3.19	1.13	0.35-3.86	1.70	0.23-3.50	1.96	0.44-1.23	0.78	-0.15-1.02	0.36
NP	0.64-1.06	0.85	0.23-1.04	0.60	-0.23-1.33	0.57	-0.64-0.02	-0.29	-1.29-0.38	-0.34
NP1EO	0.14-1.55	0.78	-0.65-1.67	0.70	-0.20-1.83	0.92	-0.80-0.46	-0.08	-1.75-0.95	0.02
MeP	0.97-3.08	1.57	0.53-4.06	2.02	1.38-3.89	2.51	-0.45-1.45	0.64	0.14-1.44	0.78
EtP	1.01	1.01	0.08	0.08	-	-	-0.93-2.01	1.09	1.02-3.22	2.11
PrP	0.91	0.91	-0.42	-0.42	-	-	-1.33-1.55	0.33	0.28-2.27	1.16
TCB	1.48-2.33	1.90	1.90-3.50	2.85	1.68-3.89	2.75	0.04-1.54	0.84	-0.45-1.90	0.71
TCS	-	-	-	-	-	-	-0.44-1.76	0.76	-0.78-1.92	0.83
BPA	-	-	-	-	-	-	-	-	-	-
BP3	0.34-1.66	1.09	-0.43-4.27	2.40	1.60-3.93	2.98	-0.77-2.68	1.23	-0.05-2.96	1.38

Kd: partition factor between marine sediment and water; BCF: Bioconcentration factor; BSAF: biota-sediment accumulation factor

Table
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Table 2. Log BAF and log BSAF values reported in the literature for contaminants measured in aquatic organisms.

Contaminant	Organisms	log BAF	log BSAF	Reference
AS C12	Fish *(Pimephales promelas)*	0-0.60*		Procter and Gamble, 2004
	Fish *(Proterorhinus marmoratus)*	0.85*		Topcuoglu and Birol, 1982
	Fish *(Cyprinus carpio)*	0.59-0.72*		Wakabayashi et al., 1981
	Fish *(Cyprinus carpio)*	0.32*		Wakabayashi et al., 1980
	Fish *(Cyprinus carpio)*	0.60*		Wakabayashi et al., 1978
	Fish *(Carassius auratus)*	0.17*		Tovell et al., 1975
AS C14	Fish *(Cyprinus carpio)*	1.04*		Wakabayashi et al., 1980
AS C14-C15	Fish *(Pimephales promelas)*	2.25-2.62*		Procter and Gamble, 2002
	Fish *(Ictalurus punctatus)*	2.60-2.99*		Procter and Gamble, 2002
AS C16	Fish *(Cyprinus carpio)*	1.86*		Wakabayashi et al., 1980
LAS C12	Shrimp (*Palaemonetes varians*)	2.07*		Renaud et al., 2014
	Oligochaeta *(L. variegatus)*	-0.30-0.67	0.75-1.55	Mäenpää and Kukkonen, 2006
	Fish (*Abramis brama*)	0.09-4.64		Tolls and Sijm, 2003
LAS C13	Fish (*Abramis brama*)	0.26-6.67		Tolls and Sijm, 2003
NP	Mussel (*Mytilus galloprovincialis*)	3.84		Vidal-Liñán et al., 2015
	Oreochromis miloticus	2.99-4.44		Chen et al., 2014
	Algae	2.12-2.87		Yang et al., 2014
	Carp (*Cyprinuscarpio linnaeus*)	3.22-4.05		Yang et al., 2014
	Corbicula fluminea	3.45-3.56	1.63-3.57	Salgueiro-González et al., 2012
	Copepod (*Eurytemora affinis*)	2.51		Cailleaud et al., 2011
	Ruditapes pjilippinarum, Sparus latus, Acanthopagrus schlegel		0.05-0.40	Zhang et al., 2011
	D. Magna	2.67-4.67		Preuss et al., 2008
	Mussel (*Mytilus galloprovincialis*)	1.72-1.86		Ricciardi et al., 2008
	Zebra fish	3.16		Huang et al., 2007
	Clam (*T. philippinarum*)	0.78-1.05[a]		Lietti et al., 2007
	Oligochaeta *(L. variegatus)*	0.17-1.52	1.14-1.74	Mäenpää and Kukkonen, 2006
	Amphipod species (*Eohaustrius estuaries, Grandidierella japonica, Corophium salmonis*)		4.6-33.9	Hecht et al., 2004
	Fathead minnow	246–434[a]		Snyder et al., 2001
	Killifish (*Oryzias latipes*)	0.653-0.863		Tsuda et al., 2001
	Mussel (*M. Edulis*)	2.75- 4.1[a]		Ekelund et al., 1990
MeP	Tilapia (muscle)	1.08-2.08		Yao et al., 2018
	Tilapia (liver)	2.77-6.47		Yao et al., 2018
	Common carp (muscle)	1.23-2.82		Yao et al., 2018
	Common carp (liver)	2.61-4.88		Yao et al., 2018
	Bream (muscle)	1.19-1.75		Yao et al., 2018
	Bream (liver)	3.85-6.12		Yao et al., 2018
	Crucian (muscle)	1.60-2.10		Yao et al., 2018
	Crucian (liver)	4.58		Yao et al., 2018
	Chub (muscle)	1.37-2.17		Yao et al., 2018
	Chub (liver)	3.78		Yao et al., 2018
	Grass carp (muscle)	1.33-2.08		Yao et al., 2018
	Grass carp (liver)	6.33		Yao et al., 2018
	Catfish (muscle)	1.35-1.68		Yao et al., 2018
	Bullhead (muscle)	1.37, 1.75		Yao et al., 2018
	Bullhead (liver)	4.38		Yao et al., 2018
	Snapper (muscle)	1.24		Yao et al., 2018
	Snapper (liver)	4.31		Yao et al., 2018
	Snakehead (muscle)	1.61		Yao et al., 2018
	Mud carp (muscle)	1.46		Yao et al., 2018
	Mullet (muscle)	1.56		Yao et al., 2018

EtP	Tilapia (muscle)	1.59		Yao et al., 2018
	Tilapia (liver)	1.12-3.37		Yao et al., 2018
	Common carp (muscle)	1.47-3.15		Yao et al., 2018
	Common carp (liver)	1.99-3.06		Yao et al., 2018
	Bream (muscle)	2.40-2.70		Yao et al., 2018
	Bream (liver)	1.26-2.68		Yao et al., 2018
	Crucian (liver)	1.66-2.67		Yao et al., 2018
	Chub (muscle)	2.48, 2.56		Yao et al., 2018
	Chub (liver)	2.15-2.40		Yao et al., 2018
	Grass carp (muscle)	2.54, 2.73		Yao et al., 2018
	Grass carp (liver)	2.23-2.54		Yao et al., 2018
	Catfish (muscle)	2.67		Yao et al., 2018
	Catfish (liver)	2.14-2.43		Yao et al., 2018
	Bullhead (liver)	2.02-2.07		Yao et al., 2018
	Snapper (liver)	1.79		Yao et al., 2018
	Snakehead (liver)	2.29		Yao et al., 2018
	Mud carp (liver)	2.45		Yao et al., 2018
PrP	Tilapia (muscle)	3.60-4.15		Yao et al., 2018
	Common carp (muscle)	1.24-4.08		Yao et al., 2018
	Common carp (liver)	2.85-3.44		Yao et al., 2018
	Bream (muscle)	1.60-3.60		Yao et al., 2018
	Bream (liver)	2.90-3.29		Yao et al., 2018
	Crucian (muscle)	3.60, 4.25		Yao et al., 2018
	Chub (muscle)	1.55-3.60		Yao et al., 2018
	Chub (liver)	3.17		Yao et al., 2018
	Grass carp (muscle)	1.63-4.17		Yao et al., 2018
	Grass carp (liver)	2.87		Yao et al., 2018
	Catfish (muscle)	1.55-4.18		Yao et al., 2018
	Catfish (liver)	2.94		Yao et al., 2018
	Bullhead (muscle)	1.29		Yao et al., 2018
	Snapper (muscle)	1.61		Yao et al., 2018
	Snakehead (muscle)	3.6		Yao et al., 2018
	Mud carp (muscle)	3.6		Yao et al., 2018
	Mullet (muscle)	3.6		Yao et al., 2018
TCB	Fish	-1.70-1.19[a]	-0.79-2.37	Vimalkumar et al., 2018
	Tilapia (muscle)	1.31-3.75		Yao et al., 2018
	Tilapia (liver)	4.14-5.42		Yao et al., 2018
	Common carp (muscle)	1.77-3.13		Yao et al., 2018
	Common carp (liver)	2.09-5.35		Yao et al., 2018
	Bream (muscle)	1.59-1.74		Yao et al., 2018
	Bream (liver)	2.37-4.40		Yao et al., 2018
	Crucian (muscle)	3.28-5.49		Yao et al., 2018
	Crucian (liver)	4.49		Yao et al., 2018
	Chub (muscle)	2.34		Yao et al., 2018
	Chub (liver)	2.37-4.61		Yao et al., 2018
	Grass carp (muscle)	1.71		Yao et al., 2018
	Grass carp (liver)	2.51-4.74		Yao et al., 2018
	Catfish (muscle)	2.7		Yao et al., 2018
	Catfish (liver)	2.42-2.89		Yao et al., 2018
	Bullhead (liver)	2.23-4.47		Yao et al., 2018
	Snapper (liver)	2.39		Yao et al., 2018
	Snakehead (liver)	4.55		Yao et al., 2018
	Mud carp (liver)	5.11		Yao et al., 2018
	Fish	4.94[b]	0.97	Zhang and Kelly, 2018
	Common carp; Crucian carp	0.47-1.54[b]		Tanoue et al., 2015
	Earthworm		0.07-0.70	Macherius et al., 2014
	Fish (*Gibelion catla*)	2.91	0.32	Shanmugam et al., 2014
	Algae (*Cladophora spp*)	3.20-3.43[a]		Coogan et al., 2007

	Snail	3.2		Coogan and La Point, 2008
	Fish (*coregonus sp.)* and roach (*rutilus rutilus*)	5.0-5.4[b]		Balmer et al., 2004
TCS	Tilapia (muscle)	1.64-5.23		Yao et al., 2018
	Tilapia (liver)	1.26-3.20		Yao et al., 2018
	Common carp (muscle)	1.80-4.09		Yao et al., 2018
	Common carp (liver)	1.95-2.58		Yao et al., 2018
	Bream (muscle)	1.31-3.48		Yao et al., 2018
	Bream (liver)	1.84-2.44		Yao et al., 2018
	Crucian (muscle)	2.24		Yao et al., 2018
	Crucian (liver)	4.3		Yao et al., 2018
	Chub (muscle)	1.93-2.46		Yao et al., 2018
	Chub (liver)	1.53		Yao et al., 2018
	Grass carp (muscle)	1.9		Yao et al., 2018
	Catfish (liver)	2.79		Yao et al., 2018
	Bullhead (liver)	2.09		Yao et al., 2018
	Snapper (liver)	2.32		Yao et al., 2018
	Mud carp (muscle)	1.7		Yao et al., 2018
	Mullet (muscle)	1.71		Yao et al., 2018
	Fish	3.02[b]	0.4-0.6	Zhang and Kelly, 2018
	Macrophite *(Echinodorus horemanii* and *Eichhornia crassipes*).	3.64[*]		Pi et al., 2017
	Common carp; Crucian carp	1.23-2.41[b]		Tanoue et al., 2015
	Earthworm		0.62-1.14	Macherius et al., 2014
	Amphipod (*Ampelisca abdita*), and shrimp (*Americamysis bahia*)		-0.92-(-0.63)	Perron et al., 2012
	Snail	2.7-3.5[a]		Coogan and La Point, 2008
	Algae (*Cladophora spp*)	2.95-3.32[a]		Coogan et al., 2007
BPA	Plants *(Callitriche sp.)*	0.71[*]	1.90	Wilkinson et al., 2018
	Crustaceans (*Gammarus pulex*)	0.10[*]	1.70	Wilkinson et al., 2018
	Aquatic snails (*Bithynia tentaculata*)	1.29[*]	2.78	Wilkinson et al., 2018
	Macrophite (*Echinodorus horemanii* and *Eichhornia crassipes*)	0.30-2.17[*]		Pi et al., 2017
	Phytoplankton, parva, carp, crucian, snake fish, Zooplankton, snail, white shrimp, lobster, catfish	-0.40-1.32[a]		Wang et al., 2017
	Macroinvertebrate (*Hydropsyche*)	4.735		Ruhí et al., 2016
	Algae	3.25-4.09		Yang et al., 2014
	Carp (*Cyprinuscarpio linnaeus*)	3.55-4.15		Yang et al., 2014
	Mussel (*Mytilus galloprovincialis*)	3.65		Gatidou et al., 2010
	Stephanidiscus hantzschii	2.00-2.70		Li et al., 2009
	Clam (*Pisidium amnicum*)	0.110-0.144[a]		Heinonen et al., 2002
	Salmon eggs	0.025-0.066		Honkanen et al., 2001
	Rainbow trout	0.23-1.58		Lindholst et al., 2000
BP3	Fish, cephalopoda and crustaceans		0.0-2.3[b]	Peng et al., 2017
	Fish		0.04-0.3[b]	Gago-Ferrero et al., 2015

BAF (L kg^{-1}); [a]: wet weight; [b]: lipid weight; [*]: BCF (Bioconcentration factor).

Figure

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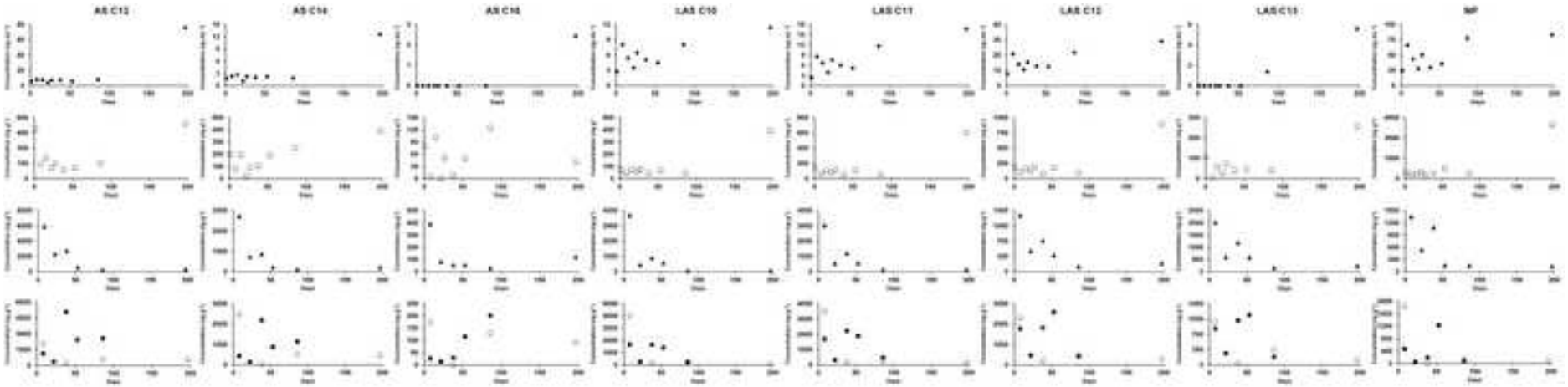

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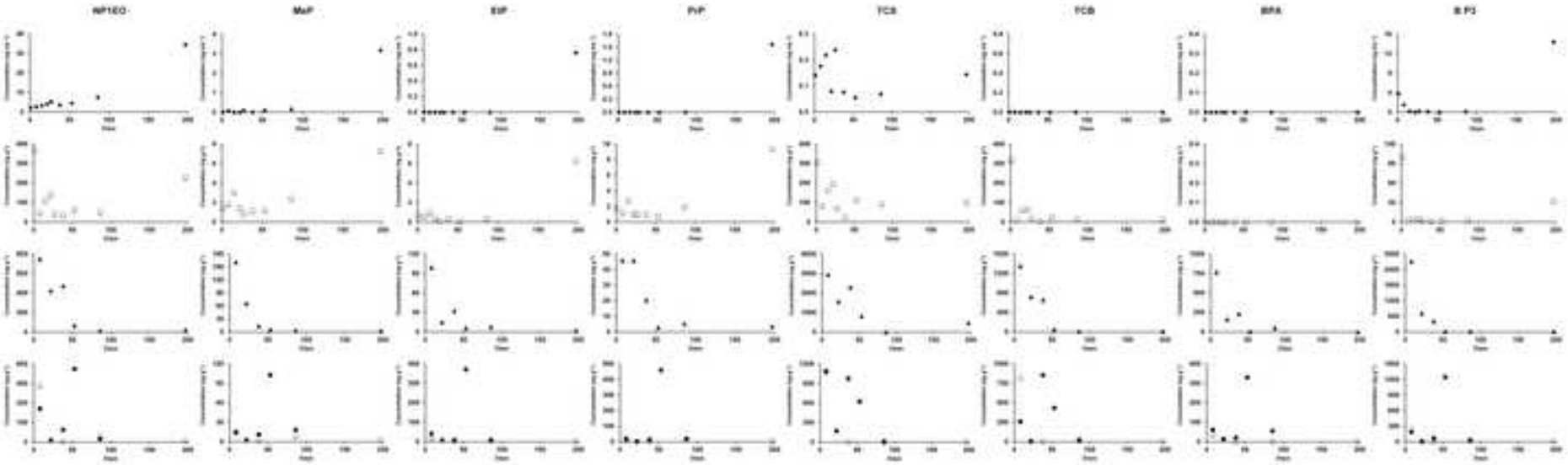

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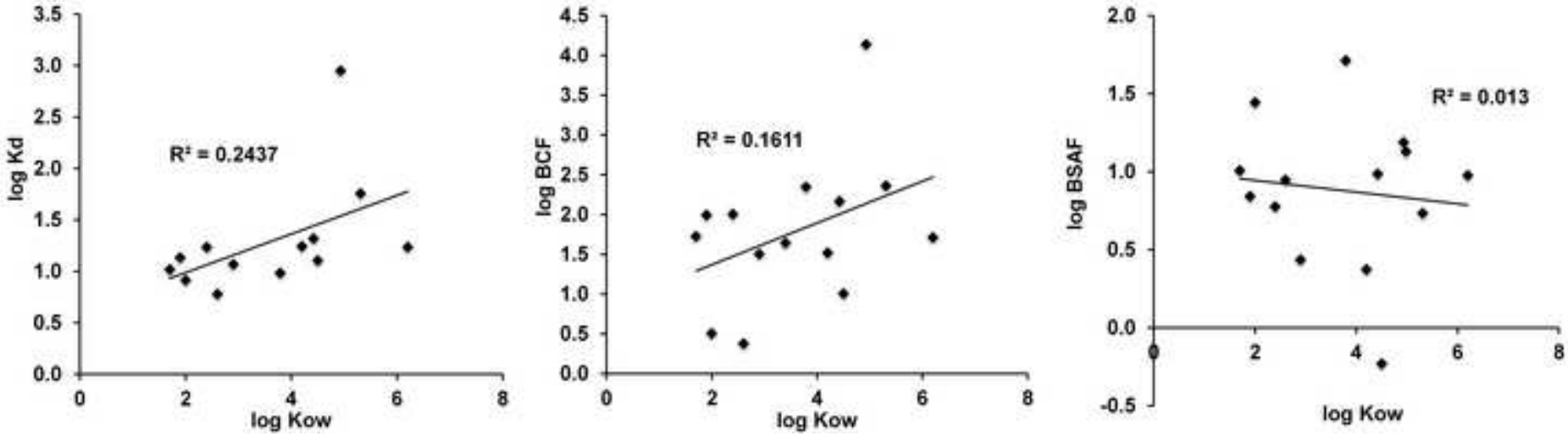

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***conflict of Interest Statement**

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:
