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Bioaccumulation of additives and chemical contaminants from environmental microplastics in European seabass (*Dicentrarchus labrax*)

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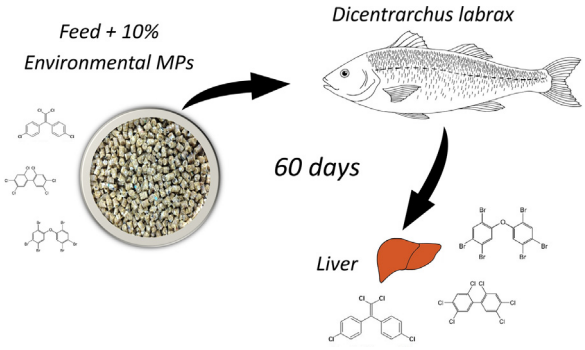
HIGHLIGHTS

- Only fish fed with 10% environmental microplastics (EMPs) bioaccumulated PBDEs in the liver.
- PCBs bioaccumulated in the liver in all treatments, but with higher values in EMP treatment.
- DDE were found in the liver of MP and EMP treatments, with significant higher concentration in EMP.
- Additives and chemical pollutants bioaccumulate in the liver by long-term ingestion of EMPs.

GRAPHICAL ABSTRACT



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ABSTRACT

Marine microplastic pollution is one of the biggest environmental problems we face. The growth of plastic production has not ceased since the 1950s and it is currently estimated that 368 tons of plastic were produced in 2019 (PlascticsEurope, 2020). Geyer et al. (2017) estimate that 79% of the plastic produced in the world still remains in the environment; this plastic due to the effect of degradation and subsequent fragmentation, is present in the form of microplastics in all oceans and, due to its small size can be ingested by fish and filter-feeding organisms. In addition, microplastics have additives and chemical contaminants associated with them, and the potential effect of microplastic ingestion on marine organisms, and through them, the potential risk to humans, is unknown. In the present study, European seabass (*Dicentrarchus labrax*) were fed for 60 days with three treatments: Control (feed), MP (feed with 10% virgin microplastics) and EMP (feed with 10% environmental microplastics), being the first study to evaluate long-term accumulation of contaminants due to ingestion of environmental microplastics (EMP) in fish. Both plastic additives such as PBDEs, and chemical contaminants adsorbed from the environment such as PCBs and DDE, were analyzed in the EMP, feed and liver.

The concentration of microplastics in the feed was calculated based on the MPs/zooplankton wet weight (WW) ratio of 0.1 found in an area of maximum accumulation in the Canary Islands. Therefore, it is an experiment that simulates real conditions, but in the worst-case scenario, using both, concentrations based on data obtained in oceanographic campaigns and microplastics collected from the environment. Our results show that in this scenario, additives and chemical contaminants adsorbed on EMPs bioaccumulate in fish liver due to long-term ingestion of microplastics.

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1. Introduction

Plastic pollution is an issue of growing concern to the citizens, environmental policy makers and the scientific community because of the potential dangers to ecosystems and organisms, including humans (Carbery et al., 2018; Galgani et al., 2019; Lim, 2021; Rainieri and Barranco, 2019). The increasing levels of pollution detected in all environments and the unstoppable growth of plastic production worldwide (PlascticsEurope, 2020) make it one of the major environmental problems facing humanity. It is estimated that 79% of all plastics produced to date remain in the environment (Geyer et al., 2017). The degradation process due to UV radiation causes them to fragment into smaller pieces, these, together with other plastics manufactured smaller than 5 mm in size and synthetic fibres derived from textiles, constitute what are known as microplastics (MPs). Although the definition is still debated, in this study we will refer to the fraction between 5 mm-1um as microplastics (MPs) (Frias and Nash, 2019; Hartmann et al., 2019). These MPs pose a danger to marine organisms. A recent review indicates that among marine vertebrates, turtles are the group with the highest incidence of MPs in their stomach, with an average of 122 particles per individual and present in the 88% of the total studied (Ugwu et al., 2021). They are followed by marine mammals, among which MPs were found in 59% of the animals studied, with an average of 10 particles per individual; seabirds, with polymers in 50% of the specimens studied and with an average of 7 particles per individual; and fish, with MPs found in 42% of the animals investigated and with an average of 3 particles per individual (Ugwu et al., 2021). For these animals there is not only the physical damage associated with the ingestion of MPs, but also the toxicological danger associated with the chemical pollutants present in MPs (Rainieri et al., 2018a). Several studies show high concentrations of chemical contaminants that adsorb MPs from surrounding environment (Camacho et al., 2019; Heskett et al., 2012; Hirai et al., 2011; Ogata et al., 2009; Teuten et al., 2009). On the other hand, plastics have additives that are intentionally added during their manufacture to give them color, or properties such as flexibility, termoresistance, UV resistance or fire resistance (Campanale et al., 2020).

The study by Camacho et al. (2019) in the Canary Islands (Spain), reported chemical pollutants in beach MPs as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), semi-persistent contaminants such as polycyclic aromatic hydrocarbons (PAHs), as well as bromodiphenyl ethers (BDEs) and emerging contaminants, such as organophosphorus flame retardants (OPFRs), chemical sunscreens (UV-filters), and the widely used pesticide chlorpyriphos. In this study, high levels of DDE-p,p′ were found in the pellets collected in Las Canteras beach, which reached median levels of 56.0 ng/g and with an extreme value of 13,488 ng/g of ΣDDTs (Camacho et al., 2019). Concentrations of ΣPCBs ranged between 0.9 and 2286 ng/g. In the case of polybrominated diphenylethers (PBDEs) (congeners #28, 47, 85, 99, 100, 153, 154 and 183) frequently used as flame retardants in plastic polymers, concentrations between 0 and 3924 ng/g were found, with significantly higher concentrations in fragments than in pellets. These additives are persistent and lipophilic, bioaccumulating in fatty tissues and thus entering the trophic webs (Shanmuganathan et al., 2011).

The Canary Islands are an area particularly affected by pollution by plastics and MPs that are carried by the Canary Current and deposited on its coasts. Many studies corroborate the high incidence of MPs on the north and north-east facing coasts (Baztan et al., 2014; Hernández-Sánchez et al., 2021; Herrera et al., 2018; Rapp et al., 2020; Reinold et al., 2020). Recent studies also indicate areas of accumulation at the surface (Herrera et al., 2020) and in the water column (Vega-Moreno et al., 2021). The study by Herrera et al. (2020) showed areas of maximum accumulation of MPs between 1 and 5 mm in the coastal area off Las Canteras beach. In these areas, the MPs/zooplankton ratio in dry weight reached 2, i.e. twice as much plastic as zooplankton, and in wet weight 0.1, which represents 10% of the zooplankton sample (Herrera et al., 2020). Furthermore, recent studies reveal the presence of MPs in fish of commercial interest in the Canary Islands such as Atlantic chub mackerel (*Scomber colias*) (Herrera et al., 2019), and in farmed sea bass (*Dicentrarchus labrax*) grown in near-shore cages (Reinold et al., 2021).

Other studies on wild fish have found the presence of MPs in tissues, causing neurotoxicity and oxidative damage (Barboza et al., 2020b; Zitouni et al., 2020). Barboza et al. (2020b) estimates that an adult could ingest about 842 MPs/year from fish consumption. Furthermore, in wild fish, a correlation has been found between the ingestion of MPs and the presence plastic additives in tissues such as bisphenols (Barboza et al., 2020a). The presence of PBDEs in fish tissues has been correlated with greater plastic debris densities, suggesting that the presence of PBDEs in fish is an indicator of plastic pollution in marine ecosystems (Rochman et al., 2014).

Many laboratory studies investigating the effect of ingestion of microplastics and associated chemical contaminants in fish have recently been conducted, many of them in zebrafish (see the review by Bhagat et al. (2020)). Most studies are conducted with microbeads or commercial MPs (Barboza et al., 2018a, 2018b, 2018c; Cousin et al., 2020; Ding et al., 2020; Grigorakis and Drouillard, 2018; Mazurais et al., 2015; Mizukawa et al., 2009; Rainieri et al., 2018b; Rummel et al., 2016; Zhu et al., 2020). Some authors have experimented with MPs deployed in harbors or bays (Müller et al., 2020; Rochman et al., 2013), and only a few studies have been conducted with environmental microplastics (Bucci et al., 2021; Pannetier et al., 2019; Zitouni et al., 2021). However, the effect of MPs and their associated chemical contaminants on wild marine organisms is so far unknown, as most experiments are conducted in the laboratory and with one particular type of MPs, small polyethylene spheres in much higher concentrations than actual concentrations (Lim, 2021). However, more environmentally realistic experiments are needed to determine effects in wild organisms, as MPs in the environment have different shapes and polymers, and are associated with different types of chemical contaminants (Wang et al., 2020). In view of the above, the main objective of this study is to determine whether there is trophic transfer of chemical contaminants (DDT, PCBs and PBDEs) to the liver of sea bass (*Dicentrarchus labrax*) by testing two different feeding treatments with realistic concentrations (in the worst-case scenario) of virgin MPs, and with environmental MPs.

2. Material and methods

2.1. Ethics statement

All procedures involving fish complied with the guidelines of the European Union Council (86/609/EU) and Spanish legislation (RD 53/2013) and were approved by Bioethical Committee of the University of Las Palmas de Gran Canaria (Ref. 06/2021 CEBA ULPGC).

2.2. Experimental diets

As is shown in Fig. 1, three diets were proposed in the experimental design: Control, MP and EMP. Control diet consistent only in feed, without MPs or added pollutants. To test the individual effects of MPs content, a diet was formulated including only 10% virgin MPs following the wet weight (WW) MP/zooplankton ratio found in the study in surface sea waters around the Canary Islands (Herrera et al., 2020) (Diet MP). Finally diet EMP was formulated with feed with a 10% of "environmental MPs" collected from Las Canteras beach.

For all the diets, commercial feed was used (D-2 Optibream AE 1P, Skretting Spain Spa crude protein 48%, fat 18%, ash 6.3%, and fiber 3.6%). A total of 5 kg of experimental feed were produced for each diet.

2.2.1. Virgin microplastics

The "virgin" microplastics were resins pellets, low density polypropylene (LDPP), (Sigma-Aldrich, ref. 428116), listed as free of bioaccumulative additives that are toxic and persistent or very bioaccumulative and very persistent at levels of 0.1% or higher (Fig. 2a).

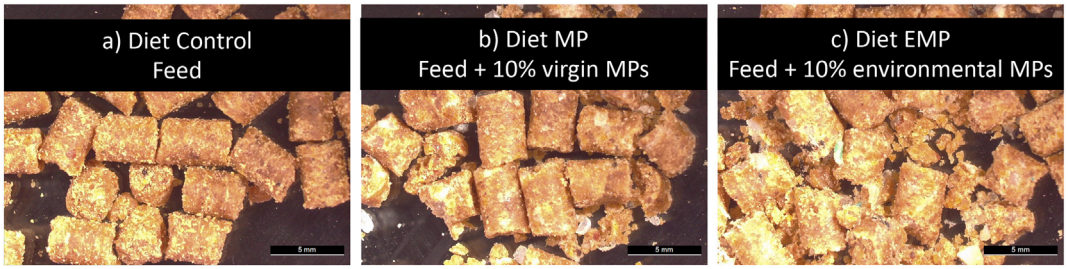

Fig. 1. Photograph of each diet a) Control, b) MP and c) EMP.

2.2.2. Environmental microplastics

MPs "naturally" contaminated were collected from the sand of Las Canteras beach with a 1 mm diameter net. Once in the laboratory, the sample was washed with bidistilled water and 1 kg of 1-5 mm MPs fraction were separated manually with tweezers.

Both virgin and environmental MPs were shredded using a cutting mill (Retsch-SM100, Haan, Germany), to obtain a size fraction between 0.7 and 1 mm.

A total of 0.5 kg of each, shredded virgin MPs (Fig. 2a) and shredded environmental MPs (Fig. 2b), were obtained. Ten kg of commercial feed was shredding too following the same process, then 5 kg were mixed with each of the MPs (virgin and environmental MPs), and re-pelletize to the corresponding grain size, resulting in both MP and EMP diets (Fig. 2c). Once the diets were mixed were put into the granulator with a 3 mm output diameter. In order to no affect the stability of the pollutants, the granulator was kept cold with ice to avoid the temperature rising up 50 °C, and the process was done under stable pressure of 2–3 atm. The feed pellets were dried in an oven at 37–39 °C. The humidity of the diets at the end of the process was between 6.75 ± 0.75%.

2.3. Experimental conditions

The experiment was carried out during September and October 2019. One hundred and eighty European seabass (*Dicentrarchus labrax*) (mean weight 80.91 ± 13.28 g and mean length 17.98 ± 1.06 cm) cultured in the facilities of the ECOAQUA Institute, ULPGC (Canary Islands, Spain) were subjected to the experimental conditions.

We have chosen the species *Dicentrarchus labrax* because they are produced from eggs in our aquaculture facilities and, therefore, have not been in contact with microplastics in the natural environment. In addition, there are a large number of studies on ingestion of microplastics and associated chemical contaminants on this species (Barboza et al., 2018a, 2018b; Espinosa et al., 2019; Granby et al., 2018a; Mazurais et al., 2015; Pedà et al., 2016; Zeytin et al., 2020); as well as metabolic studies by our research group (Torrecillas et al., 2021, 2015), which allow us to compare the results with previous studies. The experiment was carried out for 60 days, as this is the time required for this species of fish to double their weight.

The fish were distributed in nine tanks (three per each diet) with a flow-through system with aerators to provide sufficient oxygen to the tank (20 fish/tank). The experimental tanks are those usually used in aquaculture species and standardized in the aquaculture experimental facilities at ECOAQUA. The approximately half cubic meter tanks are cylindroconical with a height of 1 m and a diameter of 1.07 m. Real water column height is around 89 cm, since there is a surface drain (plus a drain at the bottom) to permit the renovation of the whole amount of water in the tank. Water conditions were monitored daily, maintaining salinity at 37 mg L^{-1}, oxygen values at 6.0 ± 0.5 ppm O_2 and temperature at 22 ± 1 °C. Fish were fed by hand 3 times per day to apparent satiety.

2.4. Sampling procedures

At day 60, 12 fish per dietary treatment (4 fish/tank) were euthanized with an overdose of clove oil anesthesia by immersion to obtain samples of different tissues after dissection. Liver samples were obtained for analysis of chemical contaminants. Pooling of 4 fish livers per tank was performed.

2.5. Analysis of chemical contaminants

2.5.1. Reagents and chemicals

Analytical-grade acetonitrile (ACN), acetone (Ac), and formic acid (FA, HCOOH) were purchased from Honeywell (Morristown, NJ, USA). Mass spectrometry grade cyclohexane (CHX) and Ethyl Acetate (AE) were obtained from Merck (Darmstadt, Germany). Salts for extraction based on the AOAC QuEChERS method (Quick, Easy, Cheap, Effective, Rugged, and Safe) (Lehotay et al., 2007) were purchased in commercial premixes from Agilent Technologies (6 g $MgSO_4$ and 1.5 g CH_3COONa) (Palo Alto, CA, USA).

The standards of the selected organic compounds were purchased from CPA Chem (Stara Zagora, Bulgaria) in a mix solution at 20 mg/mL in Ac. PCB 200 was used as procedural internal standard (P-IS) and was acquired from Dr. Ehrenstorfer (Augsburg, Germany) at 10 ng/mL. A working solution at 1 mg/mL in Ac was prepared for the analytes and another for the PCB 200. All standard stock and working mix solutions were stored in glass amber vials at −20 °C and checked periodically for stability.

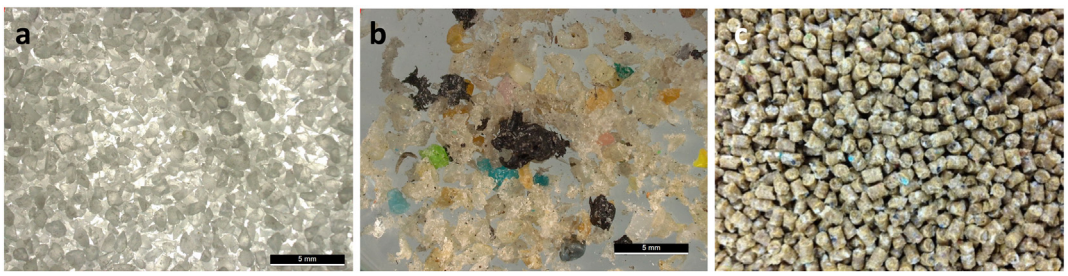

Fig. 2. a) Shredded virgin microplastics. b) Shredded environmental microplastics. c) Feed plus 10% of shredded environmental microplastics (diet EMP).

2.5.2. Extraction of chemical pollutants in environmental and virgin microplastics

The analysis of DDE-p-p′, PCBs (congeners 153, 138) and PBDEs (congeners 47, 99, 100, 153, 154) in the microplastic samples was performed following the analysis methodology presented in Camacho et al. (2019). The limits of detection (LOD) for the compounds analyzed in this study by this method have been added as supplementary data in Table S1.

For solid-liquid extraction, 5 mL of a CHX:AE mixture (1:1, *v*/v) was added to an amber glass vial containing 1 ± 0.05 g of pulverized MPs. After that, the vials were shaken vigorously and subjected to ultrasound for 20 min (Selecta, Barcelona, Spain). The samples were then shaken for 24 h on a rotary shaker (Ovan, Barcelona, Spain). Next, the 5 mL of solvent was transferred to a new vial and the process was repeated two more times until a final volume of 15 mL of the extraction mixture was reached after 72 h. Finally, 1 mL of this mixture was filtered through a 0.20 μm Chromafil® PET filters (Macherey-Nagel, Düren, Germany) to an amber glass chromatography vial and evaporated in a vacuum concentrator RVC 2–25 CD plus (Christ, Germany) to dryness. The samples were resuspended in 100 μL CHX and analyzed by GC–MS/MS.

2.5.3. Extraction of chemical pollutants in liver

Liver and feed samples were extracted by a modified QuEChERS (Anastassiades et al., 2003) procedure optimized and validated in our laboratory (Rial-Berriel et al., 2021). The limits of quantification (LOQ) and validation results for the compounds analyzed in this study by this method have been added as supplementary data in Table S2.

Into a 7 mL plastic hermetic tube containing 1 ± 0.05 g of liver or feed diet, 4 mL of ultrapure water was added along with a few stainless-steel beads and samples were subjected to homogenization for 2 min in a sample preparation equipment (Precellys Evolution, Bertin Technologies, Montigny-le-Bretonneux, France). Then, 1 g of the homogenate was taken into a 5 mL Eppendorf tube and the internal standard solution was added to all samples and blanks to yield a concentration of 5 ng/g. Likewise, blank matrix samples for a matrix-matched 10-point calibration curve were spiked with the appropriate volume for each calibration point in the same step, and all samples were mixed and left to age for 1 h. Next, 2 mL of extraction solvent (ACN 1%FA) was added to the samples, and they were shaken vigorously for 1 min. Then, they were subjected to ultrasound for 20 min (Selecta, Barcelona, Spain). After that, 480 mg of $MgSO_4$ and 120 mg of CH_3COONa were added to each tube and they were shaken for another minute. Finally, samples were centrifuged for 10 min at 4200 rpm (3175.16 ×*g*) (5804 R, Eppendorf, Hamburg, Germany) and 1 mL of supernatant was filtered into chromatography glass amber vials through a 0.20 μm Chromafil® PET filters (Macherey-Nagel, Düren, Germany).

2.5.4. Instrumental analyses

The analyses of the MPs, liver and diet samples were performed with a gas chromatograph coupled to a mass spectrometer GC System 7890B and Triple Quad 7010 (Agilent Technologies, Palo Alto, CA, USA). The chromatographic separations were performed using two 15 m columns (Agilent J&WHP-5MS, 0.25 mm inner diameter and 0.25 μm film thickness each) joined together using a purged junction that allowed the use of the backflushing technique (reversal of the carrier gas flow to remove matrix components once all analytes of interest have passed to the second column). The flow rate of the carrier gas (helium, 99.999%) was adjusted by means of the retention time locking function (RTL), using chlorpyrifos methyl (tR = 9.143 min) as a reference. The temperature ramp was programmed as follows: (a) 80 °C—1.8 min; (b) 80 °C to 170 °C at a rate of 40 °C min^{-1}; (c) 170 °C to 310 °C at a rate of 10 °C min^{-1}; (d) 310 °C for 3 min. The total time for each analysis was 20.75 min. For each analysis, 1.5 μL of the extract were injected in splitless mode. A 4 mm ultra-inert liner with glass wool was used. The injector temperature was programmed at 250 °C. Helium backflushing at 5.8 mL min^{-1} at a temperature of 315 °C for 5 min was used to clean the injector after each analysis.

The MS/MS analyses were performed in multiple reaction monitoring (MRM). The ionization source (electron impact, 70 eV) was maintained at a temperature of 280 °C. Nitrogen gas of the highest purity available (99.9999%, Linde, Dublin, Ireland) was used for Q2 fragmentation of the parent ions at a flow rate of 1.5 mL min^{-1}. The transfer line temperature was 280 °C. For data acquisition, a delay of 3.7 min was programmed to allow the solvent front to pass. The precursor and product ions, retention times, and the collision energies of all the compounds have been previously published (Henríquez-Hernández et al., 2016). The quantification was based on peak areas, using matrix-matched calibration for the liver, muscle, and diet samples and as for the MPs, calibration curves prepared in solvent (CHX) with 1% olive oil to emulate the matrix load in the samples.

The retention times, precursor, fragment ions, and collision energies for each compound and equipment have been previously published (Rial-Berriel et al., 2021). In addition, the analysis parameters for the compounds analyzed in this study have been added as supplementary data in Table S3.

2.6. Statistical analyses

Normality and homoscedasticity of data were checked by Shapiro–Wilk's and Levene's test, respectively. Statistical analysis was performed using the R Version 4.0.5 with RStudio Version 1.4.1106. One-way *ANOVA Test* was applied to determine if there were significant differences (*P*-value <0.05), and *Tukey's* post hoc for multiple comparisons when *ANOVA Test* indicated significant differences among treatments. Graphics were performed in RStudio using ggplot2 package.

3. Results

3.1. Chemical pollutants present in environmental microplastics (EMPs) from Las Canteras beach used in diet EMP

DDE-p-p′, PCBs (congeners 153, 138) and PBDEs (congeners 47, 99, 100, 153, 154) were analyzed in the microplastics collected at Las Canteras beach. In the case of DDE-p-p′, mean concentration was 0.3 ± 0 ng/g. For PBDEs, the highest values found were for BDE-47 (3.3 ± 0.6 ng/g) and BDE-99 (2.7 ± 0.2 ng/g), followed by BDE-100, BDE-153 and BDE-154 which were in the range between 0.6 and 0.2 ng/g mean values (Table 1, Fig. 3). Mean concentration of PCB-153 and PCB-138 were 1.4 ± 0.1 and 0.9 ± 0 ng/g respectively (Table 1, Fig. 3).

3.2. Chemical pollutants present in diets

In feed, PCBs and PBDEs were found only in diet EMP. The mean values of PBDEs follow the same trend as for environmental MPs, with the highest

Table 1
Concentration in ng/g of chemical pollutants present in environmental microplastics collected in Las Canteras beach (mean, standard deviation, median, minimum and maximum values). A color scale was used, ranging from yellow for the lowest values, followed by green to blue to highlight the highest values.

Pollutants in environmental microplastics (EMPs)
Mean, sd, median, minimum an maximum values by beach replicates

pollutant	mean	sd	median	min	max
BDE 100	0.6	0.1	0.6	0.55	0.79
BDE 153	0.6	0.6	0.2	0.20	1.31
BDE 154	0.2	0.0	0.2	0.15	0.24
BDE 47	3.3	0.6	3.2	2.88	3.97
BDE 99	2.7	0.4	2.6	2.38	3.22
DDE	0.3	0.0	0.3	0.24	0.28
PCB 138	0.9	0.0	0.9	0.92	0.99
PCB 153	1.4	0.1	1.3	1.31	1.41

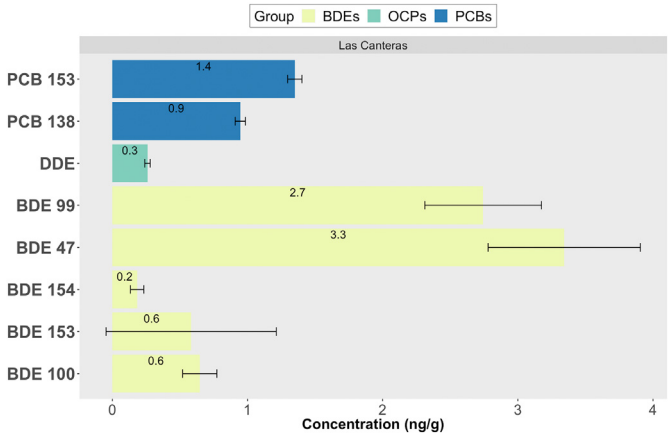

Fig. 3. Concentration of chemical pollutants in environmental microplastics (EMPs) collected from Las Canteras beach.

values being in BDE-99 (63.8 ± 25.2 ng/g) and BDE-47 (47.2 ± 16.1 ng/g); followed by BDE-100 (13.6 ± 5.4 ng/g), and BDE-153 and BDE-154 with mean concentrations of 5.8 ± 1.9 and 4.0 ± 1.6 ng/g respectively (Table 2, Fig. 4). Mean concentration of PCB-153 and PCB-138 were 4.3 ± 0.9 and 3.7 ± 0.4 ng/g respectively (Table 2, Fig. 4).

DDE-p-p′ appeared in significantly lower concentrations in diet Control (3.0 ± 0.2 ng/g) which consisted only of feed, than in diet MP consisting of feed + virgin MPs (20.8 ± 0.9 ng/g) (P-value < 0.05). On the other hand, in diet EMP (feed + environmental MPs) its mean concentration was significantly lower than in the diet with virgin MPs (7.1 ± 1.3 ng/g) (P-value < 0.05) (Table 2, Fig. 4).

3.3. Chemical pollutants in Dicentrarchus labrax liver after 60 days of treatment

At day 60 of the experiment, no PBDEs were detected in sea bass liver fed either the Control or the MP diet. The congeners BDE-47, BDE-154 and BDE-100 were found only in the liver of sea bass fed the EMP diet, at concentrations of 8.9 ± 0.8 ng/g, 4.1 ± 3.4 ng/g and 1.8 ± 0.8 ng/g respectively (Table 3, Fig. 5).

Liver bioaccumulation of PCB-153 and PCB-138 was observed in all treatments, but with significantly higher values for the EMP treatment (P-value < 0.05), with concentrations of 4.8 ± 0.5 ng/g and 4.1 ± 0.5 respectively (Table 3, Fig. 5).

Regarding DDE, no accumulation was detected in the liver of sea bass fed the control diet, but it was detected in those fed the MP and EMP diets with concentrations of 0.2 ± 0.2 ng/g and 9.1 ± 1.6 ng/g respectively, being significantly higher in EMP diet (P-value < 0.05), (Table 3, Fig. 5).

4. Discussion

There are several studies on the effect of ingestion of MPs and chemical contaminants in *Dicentrarchus labrax* (see Table S4). Many studies confirm the effect of MPs causing oxidative stress (Barboza et al., 2018a, 2018b) and histopathological alterations in liver and intestine (Espinosa et al., 2019; Pedà et al., 2016), however, there are few studies on the trophic transfer of POPs (Granby et al., 2018a; Zitouni et al., 2021).

To our knowledge, this is the first long-term study on fish fed a diet with environmental MPs, being more realistic in terms of the composition and form of MPs, as well as using actual concentrations of chemical contaminants associated with MPs.

In the present study, both plastic additives such as PBDEs and chemical contaminants adsorbed from the environment such as PCBs and DDE were analyzed. PBDEs are used as flame retardant in plastic polymers, the comercial products are industrial mixtures under the names pentabromodiphenyl ethers (penta-BDE), octabromodiphenyl ethers (octa-BDE) and decabromodiphenyl ethers (deca-BDE) (la Guardia et al., 2006). Penta-BDEs have been shown to affect neurobehavioural development and alter hormone levels; and produce adverse effects at lower doses than octa and deca-BDEs (Darnerud, 2003). While PBDEs are used in plastics as additives, also are presents in the environment and can be adsorbed from them. In our study we used environmental MPs, mainly plastic fragments derived from larger plastics containing additives, flame retardants, plasticizers, UV filters, etc. Although some commercial mixtures of penta-BDEs and octa-BDEs were banned in 2004 in the European Union (Official Journal of the European Union, 2003), the MPs we use may have been in the sea for much longer and contain these mixtures as additives, as well as adsorbing PBDEs from surrounding waters.

A study conducted on seafood in Australia, showed that the BDE-209 was the most dominant congener followed by BDE-47, 99 and 100; silver fish (*Lespisma sacchrina*) from Vietnam recorded the highest ΣPBDEs concentration (45.10 ng/g fresh weight) (Shanmuganathan et al., 2011). In addition, in the study by Tanaka et al. (2013), found presence of PBDEs lower-brominated congeners in wild seabirds, and associates it with the presence of these pollutants in prey as they are found at various levels of the marine food chain. However, predominance of higher-brominated congeners and their profile were similar to those in the deca-BDE commercial product, suggesting the transfer of these contaminants from plastic to tissues (Tanaka et al., 2013). Unfortunately, due to

Table 2
Concentration in ng/g of chemical pollutants present in each diet: control, MP and EMP (mean, standard deviation, median, minimum and maximum values). A color scale was used, ranging from yellow for the lowest values, followed by green to blue to highlight the highest values.

Pollutants in feed
Mean, sd, median, minimum and maximum values by treatment

treatment	pollutant	mean	sd	median	min	max
Control	BDE 100	0.0	0.0	0.0	0.00	0.00
Control	BDE 153	0.0	0.0	0.0	0.00	0.00
Control	BDE 154	0.0	0.0	0.0	0.00	0.00
Control	BDE 47	0.0	0.0	0.0	0.00	0.00
Control	BDE 99	0.0	0.0	0.0	0.00	0.00
Control	DDE	3.0	0.2	3.0	2.81	3.25
Control	PCB-138	0.0	0.0	0.0	0.00	0.00
Control	PCB-153	0.0	0.0	0.0	0.00	0.00
MP	BDE 100	0.0	0.0	0.0	0.00	0.00
MP	BDE 153	0.0	0.0	0.0	0.00	0.00
MP	BDE 154	0.0	0.0	0.0	0.00	0.00
MP	BDE 47	0.0	0.0	0.0	0.00	0.00
MP	BDE 99	0.0	0.0	0.0	0.00	0.00
MP	DDE	20.8	0.9	20.8	19.82	21.66
MP	PCB-138	0.0	0.0	0.0	0.00	0.00
MP	PCB-153	0.0	0.0	0.0	0.00	0.00
EMP	BDE 100	13.6	5.4	15.6	7.51	17.72
EMP	BDE 153	5.8	1.9	6.7	3.56	7.08
EMP	BDE 154	4.0	1.6	4.1	2.29	5.58
EMP	BDE 47	47.2	16.1	54.1	28.74	58.72
EMP	BDE 99	63.8	25.2	74.8	35.01	81.62
EMP	DDE	7.1	1.3	7.8	5.61	7.89
EMP	PCB-138	3.7	0.4	3.9	3.26	3.91
EMP	PCB-153	4.3	0.9	4.3	3.41	5.17

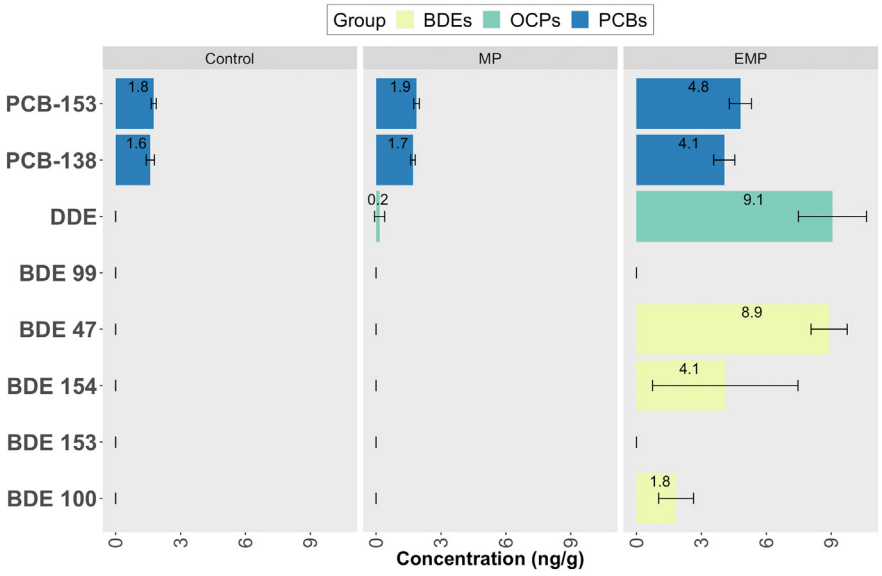

Fig. 4. Concentration of chemical pollutants in each feed treatment: control, MP and EMP.

technical limitations the concentration of BDE-209 could not be analyzed in the present study.

Our results show that PBDEs that were present in the environmental MPs, were found only in the liver of sea bass fed with feed + 10% EMPs, demonstrating that transfer was from the EMPs to the liver of the fish. However, from analyzed PBDEs (congeners 47, 99, 100, 153, 154), BDE-99 and BDE-153 were present in EMPs and in diet EMP in relatively high concentrations but not in *Dicentrarchus labrax* liver. In the case of BDE-99 is remarkable, because was found in the highest concentration in diet EMP (63.8 ng/g) but did not bioaccumulate in the liver. These results are consistent with those obtained by Granby et al. (2018b) who found that in sea bass fed with diet containing MPs and contaminants, fillets did not accumulate BDE-99 as expected, finding low concentrations and an accumulation efficiency between 9 and 12%, and was also eliminated much faster than the rest of the contaminants. On the other hand, the concentration of BDE-47 was much higher than expected, the authors attribute this to the fact that sea bass metabolizes 2,2,4,4,4,5-pentabromodiphenyl ether (BDE-99) by a reductive debromination to 2,2,4,4,4,-tetrabromodiphenyl ether (BDE-47) (Granby et al., 2018b). In the cited study, a high elimination coefficient of BDE-153 and a lower assimilation efficiency than its structural analogue BDE-154 was also observed, indicating that also in this case debromination may contribute to metabolism. These results could explain why in our study BDE-99 and BDE-153 did not appear in the liver of sea bass fed diet EMP.

Other authors have studied the bioconcentration and biomagnification of PBDEs and PCBs, demonstrated that congeners with higher octanol-water partition coefficient (Kow) have more bromine atoms, which increases their molecular weight, inhibiting their passage through membranes and reducing their partitioning in tissues. The bioconcentration factors (BCF) of PBDEs and PCBs congeners increase as the octanol-water partition coefficient (Kow) increases up to a threshold (log Kow = 7), above which, this trend is reversed and BCF decreases as Kow increases (Mizukawa et al., 2009). (Wardrop et al., 2016) have found similar results to those obtained in the present study regarding the bioaccumulation of PBDEs in rainbow fish fed with PBDEs-contaminated microbeads. The concentration of BDE-47 in the tissue was significantly higher than for the rest of congeners, followed by BDE-23, 100, 154 and 153. As in our study, they found that BDE-99 is not transferred to the tissues of exposed fish, o more probably BDE99 was transformed to BDE47 by the removal of Br at the meta position as proposed by other authors (Granby et al., 2018b; Mizukawa et al., 2013; Noyes et al., 2010; Roberts et al., 2011). Furthermore, (Roberts et al., 2011) showed that there are differences in the metabolism of PBDEs among different fish species.

Other laboratory studies showed that PBDEs can be transferred to amphipod tissue, both in the presence and absence of MPs (Chua et al., 2014). MPs apparently inhibited the uptake of PBDEs in the amphipods, possibly because the PBDEs were strongly adsorbed to the MPs, making them less bioavailable. However, the study demonstrates that MPs can transfer BDEs to amphipod tissue by acting as vectors of chemical contaminants (Chua et al., 2014). In the cited study BDE-99 had the highest proportional uptake. Authors hypothesize that could be due to digestive fluids in the gut that removing a greater proportion of adsorbed BDE-99 from the MPs, or marine organisms may have a greater propensity for assimilating adsorbed BDE-99 compared to other PBDE congeners when ingesting contaminated MPs (Chua et al., 2014). These results do not agree with ours and Granby et al. (2018b) as BDE-99 appeared in high concentration in the diet but did not bioaccumulate in the liver. However, it should be noted that the study by Chua et al. (2014) was carried out in crustaceans and only worked with polyethylene microspheres obtained from Nivea facial soap, whereas our study used beach MPs containing various types of polymers.

Here, PCBs congeners 153 and 138 were found in the MPs collected on beaches (EMP) at concentrations of 1.4 ng/g and 0.9 ng/g respectively. PCBs were not detected in the feed without environmental MPs (Control and MP) but were detected in the feed with 10% of environmental MPs (EMP feed) with values of 3.7 ng/g for PCB-153 and 4.3 ng/g for PCB-138. After 60 days of treatment, both congeners were detected in the liver of sea bass exposed to the three treatments, although in significant higher concentrations for the 10% EMP feed, which reached values of 4.8 ng/g for congener 153 and 4.1 ng/g for congener 138.

In the case of PCBs that are ubiquitous in the environment, they are adsorbed on MPs in the surrounding water and may contribute, via MPs ingestion, to bioaccumulation in marine organisms. However, chemical partitioning models indicate that POPs bioaccumulation through MPs is unlikely to occur, or that their contribution would be negligible compared to uptake through water or diet (Bakir et al., 2016; Koelmans et al., 2016).

This explains what occurred in the sea bass exposed to the Control and MP treatments, that PCBs may have bioaccumulated through the feed or water (Berntssen et al., 2010; di Bella et al., 2018). Environmental MPs, however, have contributed to this bioaccumulation in addition to the contribution from water and diet.

Respect to the PCBs bioaccumulation, the results of the different experimental studies can be contradictory. Some studies such as those carried out

Table 3
Concentration of chemical pollutants (ng/g) in *Dicentrarchus labrax* liver after 60 days feeding each treatment: control, MP and EMP (mean, standard deviation, median, minimum and maximum values). A color scale was used, ranging from yellow for the lowest values, followed by green to blue to highlight the highest values.

Pollutants in Dicentrarchus labrax liver
Mean, sd, median, minimum and maximum values by treatment

treatment	pollutant	mean	sd	median	min	max
Control	BDE 100	0.0	0.0	0.0	0.00	0.00
Control	BDE 153	0.0	0.0	0.0	0.00	0.00
Control	BDE 154	0.0	0.0	0.0	0.00	0.00
Control	BDE 47	0.0	0.0	0.0	0.00	0.00
Control	BDE 99	0.0	0.0	0.0	0.00	0.00
Control	DDE	0.0	0.0	0.0	0.00	0.00
Control	PCB-138	1.6	0.2	1.7	1.38	1.72
Control	PCB-153	1.8	0.1	1.8	1.65	1.88
MP	BDE 100	0.0	0.0	0.0	0.00	0.00
MP	BDE 153	0.0	0.0	0.0	0.00	0.00
MP	BDE 154	0.0	0.0	0.0	0.00	0.00
MP	BDE 47	0.0	0.0	0.0	0.00	0.00
MP	BDE 99	0.0	0.0	0.0	0.00	0.00
MP	DDE	0.2	0.2	0.2	0.00	0.33
MP	PCB-138	1.7	0.1	1.7	1.63	1.78
MP	PCB-153	1.9	0.1	1.9	1.78	1.96
EMP	BDE 100	1.8	0.8	1.6	1.17	2.73
EMP	BDE 153	0.0	0.0	0.0	0.00	0.00
EMP	BDE 154	4.1	3.4	3.0	1.44	7.88
EMP	BDE 47	8.9	0.8	9.2	7.95	9.51
EMP	BDE 99	0.0	0.0	0.0	0.00	0.00
EMP	DDE	9.1	1.6	9.6	7.29	10.31
EMP	PCB-138	4.1	0.5	4.2	3.51	4.45
EMP	PCB-153	4.8	0.5	4.6	4.41	5.38

by Devriese et al. (2017) in lobsters (*Nephrops norvegicus*), or those of Besseling et al. (2013) in lugworms (*Arenicola marina*), observe a small contribution of PCBs spiked on MPs to the bioaccumulation. In the other hand, van der Hal et al. (2020) observed traces of PCB-194 in rabbitfish (*Siganus rivulatus*) muscle after feeding it for two weeks with a diet containing MPs pre saturated with 11 PCB congeners at concentration of 5000 ng/g. Other long-term studies, in contrast, demonstrate the transfer of chemical pollutants from MPs to fish, as well as that they induce hepatic stress (Rochman et al., 2013).

Rochman et al. (2013) with an experimental design similar to the present study but using a diet containing 10% marine MPs (low-density polyethylene (LDPE) pellets deployed in an urban bay), obtained results according to ours. Their findings showed that fish exposed to marine plastics bioaccumulate PAH, PCBs and PBDEs and suffer liver toxicity at 60 days of treatment, however, at 30 days exposure no significant differences were observed between treatments, suggesting that short periods of exposure to 10% marine plastics are not a significant source of these contaminants (Rochman et al., 2013).

Finally, regard to DDE, the concentration in the environmental MPs was 0.3 ng/g. In the analysis of the diets, DDE was found in the control, denoting that the feed had DDE residues, as well as in the MPs diet and the diet with EMPs. In the liver, DDE was not detected in the control treatment, while in sea bass consuming 10% EMPs, liver concentrations were significantly higher (9.1 ng/g) than the MP treatment (feed + virgin MPs) (0.2 ng/g). Other studies also demonstrate the presence of DDE in feed and in wild and farmed fish (Schnitzler et al., 2008), as this contaminant, a residue of DDT, is ubiquitously present in all environments, and especially in fish oils (Ginés et al., 2018). Similar to that occurs with PCBs, although DDE were found in the liver of sea bass exposed to control and MP diets, a higher bioaccumulation was observed in the diet containing EMPs, which confirms that environmental MPs increase the concentration of chemical pollutants, adding to the contribution through water and diet.

The effect of exposure to a complex mixture of plastics and chemical additives or chemicals adsorbed from the marine environment is currently unknown. The type of polymer, the type of chemical pollutant, the exposure time and the physiological processes of digestion, assimilation and metabolization of each organism can substantially modify the levels of bioaccumulation.

While the composition was not analyzed in the present study, Edo et al. (2019) have studied the composition of MPs in Lambra beach (La Graciosa, Canary Islands) and showed that they correspond mainly to PE (63%), PP (32%) and PS (3%). Another study on the beaches of Tenerife (Canary Islands) also reported similar percentages of polymers PE (69%), PP (18%) and PS (4%) (Álvarez-Hernández et al., 2019). Since that microplastics are of exogenous origin and are deposited on beaches carried by the Canary Current, it can be assumed that the composition is similar in the microplastics collected in Las Canteras beach.

It was not the objective of this study to analyze the effect of the different types of polymers and the aging of the MPs on the transfer of chemical contaminants. However, other studies have shown that the physicochemical properties of the different plastics can affect the adsorption, release and transport of chemical contaminants, and different types of additives are added depending on the type of polymer (Gouin, 2021). On the other hand, the aging of plastics increases the mobility capacity of chemical contaminants, since it increases the negative charge on the surface and fundamentally the hydrophilicity of the materials (Liu et al., 2019).

The novelty of the present study is that the experiment was conducted with "real" microplastics collected from the environment, which have been floating in the sea and are aged by the effect of UV radiation; these microplastics are the ones to which marine organisms are exposed, and the health effects are currently unknown.

The present study provides evidence that both, additives present in environmental MPs (EMPs), and those adsorbed from the surrounding ocean have been transferred to the liver of fish after 60 days of feeding with feed + 10% EMPs.

5. Conclusions

Our results show that additives present in environmental microplastics bioaccumulated in the liver after 60 days of treatment with a diet consisting of feed plus 10% microplastics collected from beaches. This shows that in a real, but in the worst-case scenario, as they are areas of maximum accumulation, fish could be bioaccumulating additives from microplastic ingestion. In the case of contaminants that were found adsorbed to microplastics, such as DDT and PCBs that are ubiquitous in all environments, these contaminants also appeared in control diet and diet with virgin MPs, but for both contaminants bioaccumulation was significantly higher in the liver of sea bass fed 10% environmental microplastics with the diet, so ingestion of microplastics in the environment may increase exposure to these contaminants, adding to that of the surrounding water and diet.

It is important to continue with studies carried out with environmental microplastics to understand both the physical effects of ingestion as well as the trophic transfer of chemical contaminants that could cause long-term health effects. Studies over longer periods of time are also needed to assess the effect of chronic exposure to environmental microplastics on fish.

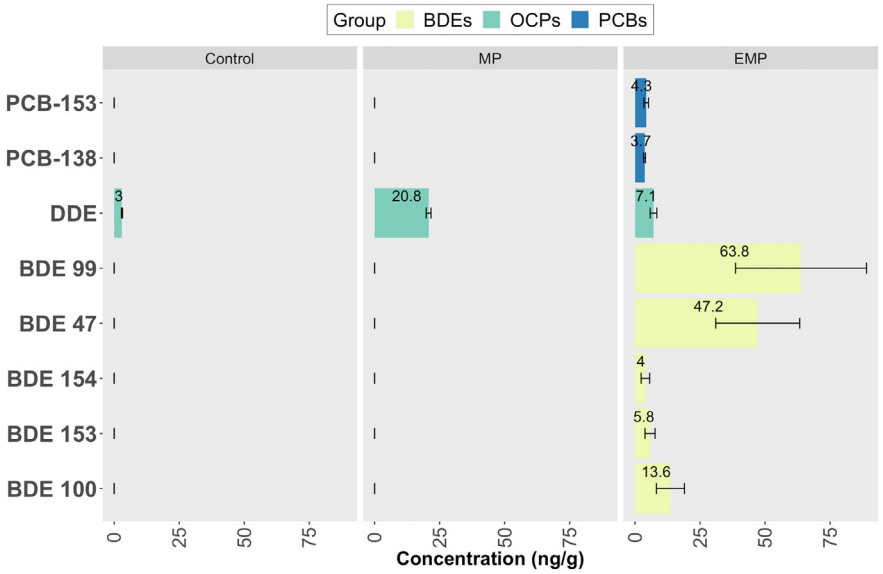

Fig. 5. Concentration of chemical pollutants (ng/g) in *Dicentrarchus labrax* liver after 60 days feeding each treatment (control, MP and EMP).

CRediT authorship contribution statement

Alicia Herrera: conceptualization, experimental investigation, data curation and writing. **Andrea Acosta-Dacal, Octavio Pérez Luzardo** and **Sarah Montesdeoca-Esponda**: analysis of chemical pollutants, experimental investigation, methodology. **Jorge Rapp** and **Stefanie Reinold**: experimental investigation, methodology. Ico Martínez, Daniel Montero and May Gómez: conceptualization, experimental investigation. All authors contributed to the acquisition of the data, review the manuscript and approved the final version.

Declaration of competing interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.153396.

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