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PII:	S0048-9697(19)33219-X
DOI:	https://doi.org/10.1016/j.scitotenv.2019.07.106
Reference:	STOTEN 33300
To appear in:	*Science of the Total Environment*
Received date:	2 May 2019
Revised date:	3 July 2019
Accepted date:	7 July 2019

Please cite this article as: R. Beiras, S. Muniategui-Lorenzo, R. Rodil, et al., Polyethylene microplastics do not increase bioaccumulation or toxicity of nonylphenol and 4-MBC to marine zooplankton, Science of the Total Environment, https://doi.org/10.1016/j.scitotenv.2019.07.106

Polyethylene microplastics do not increase bioaccumulation or toxicity of nonylphenol and 4-MBC to marine zooplankton.

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Declarations of interest: none

Abstract

Global production of synthetic polymers, led by polyethylene (PE), rose steadily in the last decades, and marine ecosystems are considered as a global sink. Although PE is not biodegradable, in coastal areas it fragments into microplastics (MP) readily taken up by biota, and have been postulated as vectors of hydrophobic chemicals to marine organisms. We have tested this hypothesis using two organisms representative of the marine plankton, the holoplanktonic copepod *Acartia clausi*, and the meroplanktonic larva of the *Paracentrotus lividus* sea-urchin, and two model chemicals with similar

hydrophobic properties, the 4-n-Nonylphenol and the 4-Methylbenzylidene-camphor used as plastic additive and UV filter in cosmetics. Both test species actively ingested the MP particles. However, the presence of MP never increased the bioaccumulation of neither model chemicals, nor their toxicity to the exposed organisms. Bioaccumulation was a linear function of waterborne chemical disregarding the level of MP. Toxicity, assessed by the threshold (EC_{10}) and median (EC_{50}) effect levels, was either independent of the level of MP or even in some instances significantly decreased in the presence of MPs. These consistent results challenge the assumption that MP act as vectors of hydrophobic chemicals to planktonic marine organisms.

Keywords: plastics, bioaccumulation, nonylphenol, 4-MBC, toxicity bioassays.

1. Introduction

Due to the multiple advantages of plastics for house-hold and industrial applications, the global production of synthetic polymers such as polyethylene (PE), polypropylene (PP) polystyrene (PS), and polyvinylchloride (PVC), rose steadily from the middle of the last century up to the current 380 Mt per year (Geyer et al., 2017). More than a half of the total plastic production is intended to packaging, bags and non-durable objects (Barnes et al., 2009). However, none of these polymers are biodegradable, and marine ecosystems are considered as the global sink for plastic litter, where it shows a tendency towards fragmentation, sinking and ingestion by biota (Cózar et al., 2014; Eriksen et al., 2014). The average size of plastic particles in the sea seems to be decreasing, and the abundance and global distribution of microplastic (MP) fragments have increased over the last few decades (Barnes et al., 2009). Since the first observations of plastic particles

in the sea, the ingestion of those particles by marine fauna was documented, and they were proposed as potential vectors of sorbed hydrophobic organic pollutants into the marine food webs (Carpenter et al., 1972). Laboratory experiments confirmed the trophic transfer of MP across planktonic food webs (Setälä et al., 2014), and also the egestion of the ingested MP (Mazurais et al., 2015). However, evidence remains scant that during gut passage MP-sorbed hydrophobic organic compounds (HOC) detach from the polymer, and thus that MP are important transfer vectors of HOC to biological tissues of marine organisms (Lohmann, 2017; Ziccardi et al., 2016). Therefore, there is currently limited scientific evidence to suggest that MP are increasing the uptake of HOC by aquatic organisms (Burns and Boxall, 2018).

This work is aimed to test the hypothesis that the ingestion of polyethylene MP (hereafter PE-MP) increases the bioavailability of HOC to marine zooplanktonic organisms, acting as vectors of those chemicals from the water column into the organisms. The experiments were conducted with the meroplanktonic larvae of the *Paracentrotus lividus* sea-urchin , and with the holoplanktonic larvae of the copepod *Acartia clausi*. Both are common biological models used in standard methods adopted by international institutions such as ISO and ICES (Beiras et al., 2012; ISO14669, 1999), and adaptations for toxicity testing with plastic microparticles are available (Beiras et al., 2018), PE was used as model polymer since it is the most frequent component of marine microplastics (Burns and Boxall, 2018), and the polymer with the highest sorption capacity for HOC according to the polymer-water partition coefficients (O'Connor et al., 2016).

4-Nonylphenol (4-n-NP, log K_{ow} 5.76 (ECHA, 2012)) and 4-Methylbenzylidene-camphor (4-MBC, log K_{ow} 5.1 (ECHA, 2018)) were used as model HOC. Nonylphenols (NP) are released in the environment by degradation of the most common non-ionic

surfactants used in detergents and cleaning products. Moreover, NP is used as pesticide, as a monomer in phenol/formaldehyde resins and mainly as plasticizer for high-density PE, polyethyleneterephthalate, and PVC (Loyo-Rosales et al., 2004). 4-n-NP was detected in estuaries at levels ranging between 0.005 µg L^{-1} (MQL) and 0.337 µg L^{-1} (Salgueiro-González et al., 2015b), and found in aquatic organisms (*Corbicula fluminea* and *Mytilus galloprovincialis*) at concentrations ranging from 9 to 24.5 ng g^{-1}dw and 6.7 to 175 ng g^{-1}dw, respectively, indicating potential bioaccumulation ability (Salgueiro-González et al., 2015a; Salgueiro-González et al., 2016). NP stands out as an endocrine disruptor compound (EDC) because it mimics the female hormone 17-*β*-estradiol and inhibits the aromatase enzyme essential for the synthesis of testosterone, according with a variety of both *in vitro* and *in vivo* assays (David et al., 2009), and because of its toxicity on sensitive aquatic organisms it poses a remarkable environmental risk in coastal ecosystems (Tato et al., 2018).

4-MBC is an aromatic compound used as UV filter in sunscreens and other common personal care products because it adsorbs UV radiation. The use of 4-MBC is not allowed as a sunscreen component in USA because it is considered an EDC, but cosmetic products in Europe can contain up to 4% (Regulation (EC) No 1223/2009). 4-MBC has been found in coastal ecosystems at concentrations up to 0.8 µg L^{-1} in Oslo Fjord (Langford and Thomas, 2008), and up to 1.04 µg L^{-1} in beach water of the Canary Islands (Spain) (Sánchez Rodríguez et al., 2015), and it bioaccumulates in bivalves (Vidal-Liñán et al., 2018) and fish (Balmer et al., 2005). Reported toxicity thresholds for marine organisms (5.4 µg L^{-1} for microalgae, 71.6 µg L^{-1} for invertebrates (Paredes et al., 2014)) are only one order of magnitude above maximum concentrations in coastal ecosystems, which translates into concerning levels of environmental risk.

2. Materials and Methods

2.1. Exposure media

4-n-NP (CAS number 104-40-5) and 4-MBC (CAS number 36861-47-9) analytical grade standards were obtained from Sigma-Aldrich. Stock solutions were prepared in dimethyl sulfoxide (DMSO), >99.9% purity, stored in darkness and discarded after a maximum of 48 h. PE-MP with a nominal size range of 4-6 µm and a density of 0.96 g cm^{-3} were obtained from Micropowders Inc. (NY-USA). Actual particle size distribution was recorded by using a Multisizer 3 Coulter Counter (Beckman), resulting a median of 5.5 µm and a distribution with 10^{th} and 90^{th} percentiles of 3.4 and 9.9 µm respectively.

Exposure media were made up in 500 mL glass bottles with Teflon-lined caps by diluting the 4-n-NP and 4-MBC stocks in organic matter free artificial sea-water (Lorenzo et al., 2002). Two levels of MP, 1 mg L^{-1} and 10 mg L^{-1}, plus a control with no MP, were tested. After 48 h of rotatory mixing (1 rpm) at 20±1ºC in the dark, exposure media were transferred into glass vials for the bioassays.

2.2. Biological material and bioassays

The obtaining of biological materials and general bioassay procedures have been previously described (Beiras et al., 2012; Beiras and Tato, 2019). Briefly, mature oocytes obtained by dissection of ripe adults were fertilized with a few µL of sperm in 50 mL measuring cylinders under gentle stirring. For the bioassays, fertilized eggs (40 per mL) were transferred before the first cleavage into glass vials with airtight Teflon-lined caps containing 27 mL of the exposure media. Four replicates per treatment plus seawater and solvent controls were carried out. The vials were placed in a rotatory wheel set at 1 rpm at a temperature of 20±1 °C in the dark. After 48 h, vials were fixed with a few drops of concentrated formalin, observed in a Leica DMI 4000B inverted

microscope, and length (maximum linear dimension) was recorded by using Leica LAS image analysis software (Leica Microsystems, Germany). Controls run in all experiments were DMSO solutions at the highest concentration used in each test, always at values ≤ NOEC (no observed effect concentration) previously calculated for this test species.

Acute lethal toxicity tests with copepods followed standard methods (ISO14669, 1999) using nauplius larvae. From 48 to 72 h before the start of the test, *A. clausi* mature adults obtained from a laboratory stock maintained by ECIMAT (University of Vigo) were transferred to a 300 µm mesh submerged in a 2 L plastic jar with bubbling filtered air maintained in an isothermal room at 20 º C, and fed with a *Rhodomonas lens* cell suspension. Using a 40 µm mesh, ≤ 24 h larvae were collected and placed in a 250 mL baker. Under binocular stereoscope, 10 larvae were delivered by glass pipette in each of the 25 mL glass vials filled with the testing solutions. A total of 4 vials per treatment were used. The vials were placed in a rotatory wheel set at 1 rpm at 20±1 °C and under an 18/6 h light/dark photoperiod. Copepod survival was recorded after 48 h.

Photographs reporting ingestion of MP were made using bright light microscopy with polarized light (Leica DMI 4000B). PE birefringence properties (Wedgewood and Seferis, 1984) allow PE-MP to appear green under polarized light.

In order to study ingestion of both MP and microalgae, forty-two hours post-fertilization sea-urchin larvae were incubated in the presence of 1 mg L^{-1} of PE-MP or *Isochrysis galbana* microalgae, in 250 mL glass bottles placed in a rotatory wheel, and particle density was recorded during 6 h every 2 h by using a Multisizer 3 Coulter Counter (Beckman). Clearance rates (CR, mL min^{-1}) were calculated following the expression:

$$CR = \frac{(\text{Ln } C_{tc} - \text{Ln } C_t) \times V}{t}$$

Where V is the volume of the incubation vessels (mL), t is the incubation time (min), and C_{tc} and C_t are the particle concentrations at the end of the incubation period for the control and the treatment respectively.

2.3. Chemical analyses

Actual concentrations of both chemicals in the bioassay containers were analytically checked to assess their stability along the exposure period. Water samples were taken at the beginning (t=0 h), at 48 h (only for the intermediate concentration), and at the end of the incubation period (96 h). Larvae were also analyzed at the end of the exposure period. PE-MP were removed from water samples by centrifugation (2000 rpm/18 ˚C/5 min), since PE-MP remain afloat and water was collected with a pipette excluding the surface layer. Larvae were then retained in an 80 µm nylon mesh. All samples were stored frozen in darkness until chemical analysis.

2.3.1. 4-n-NP

Analysis of 4-n-NP was performed by the Applied Analytical Chemistry Group (University of A Coruña). Water samples were extracted according to Salgueiro-González et al. (2012). Briefly, water samples (30 mL) were extracted by dispersive liquid–liquid microextraction (DLLME) with 100 µL of 1-octanol.

Larva samples were extracted by sonication with 25 mL of MeOH at 30ºC during 30 min. The extract was concentrated in Syncore® Analyst evaporator at 40ºC and rise up to 1 mL with MeOH.

In both cases determination was performed by liquid chromatography tandem mass spectrometry (LC–MS/MS) using an Agilent HP-1200 Series LC system coupled to a mass spectrometer with a triple quadrupole detector (API 3200, Applied Biosystems,

Carlsbad, CA, USA) with ESI interface operating in negative mode. The chromatographic separation was carried out with a column Hypersil Gold C18 (150 x 2.1 mm), 3 µm Thermo Fisher Scientific Inc. (Waltham, MA), using as mobile phase A (water) and B (methanol) with 0.05% of ammonia as modifier. According conditions optimized by Salgueiro-González et al. (2012). The limit of quantification of the method and trueness were 5 ng L^{-1} and 103% for water samples, 20.3 ng g^{-1} and 113% for larvae samples, respectively.

2.3.2. 4-MBC

The analysis of 4-MBC was performed at the Institute for Food Analysis and Research, (University of Santiago de Compostela) LC-MS/MS. Water samples were filtered and directly injected in the LC-MS/MS system. In the case of larvae samples, these were previously submitted to a sample preparation protocol based on matrix-solid phase dispersion (MSPD). Thus, 20-50 mg of larvae were dispersed in a glass mortar with 0.5 g of anhydrous sodium sulfate and 0.5 g of Florisil and placed into a cartridge containing 0.5 g of C_{18}. Then, 5 mL of acetonitrile were passed through the cartridge, evaporated to dryness under N_2 stream and reconstituted with 0.5 mL of acetonitrile.

Finally, 10 µL of water or larvae extract were injected in a Varian (Walnut Creek, CA, USA) LC-tripe quadrupole-MS system. This instrument consists of a ProStar 210 dual pump coupled to a Varian 320MS with an ESI interface. The column was a Supelco Ascentis Express C8 (50 mm × 2.1 mm, 2.7 µm) from Sigma-Aldrich. A dual eluent system of water (A) and methanol (B), both acidified with 0.1% of formic acid, was used. The gradient was as follows: 0-1 min (5% B), 5-10 min (95% B), 10-15 min (5% B). The column flow rate was set at 0.2 mL min^{-1}. 4-MBC was measured in the multiple reaction monitoring (MRM) ESI positive mode. The transition m/z 255>105 was used

for quantification and 255>97 and 255>195 were used for confirmation purposes. Quantification was made using the standard addition method for both water (10-1000 µg L^{-1}; R^2 0.994) and larvae (10-1000 µg g^{-1}; R^2 0.999) since a recovery around 90% was achieved after the MSPD protocol. The limits of quantification were 10 µg L^{-1} and 13 ng g^{-1} for water and larvae, respectively.

2.4. Statistical analysis

Normal distribution of data was checked using the Kolmogorov-Smirnov test, and homoscedasticity of the data was checked using the Levene's test. Mortality data was angular transformed (arcsine of the square root) prior to analysis. Two-way ANOVA was used for normal and homoscedastic datasets. When significant differences ($p < 0.05$) among groups were found using ANOVA then each treatment was compared to the control using Dunnett's post hoc test. Non-parametric post hoc tests were used for normal but heteroscedastic data. Non-parametric Mann–Whitney U tests were used when data clearly departed from normal distribution. Effective concentrations reducing by 10% and 50% *P. lividus* larval size (EC_{10} and EC_{50} respectively), and causing 10% and 50% mortality of *A. clausi* larvae (LC_{10} and LC_{50} respectively), were calculated by fitting the data to a probit dose–response model. Statistical analyses were performed using IBM SPSS (v.22) and SigmaPlot (v.12.5) software.

3. Results and Discussion

3.1. Stability of the chemicals in solution and sorption to PE-MP

The analytical results show that during the 48 h incubations without PE-MP, the concentrations of 4-n-NP in solution showed remarkable stability and a moderate decrease down to 56–93% of initial values, higher at the lower 4-n-NP levels (Table 1). This suggests non-linear adsorption processes more relevant at lower solute

concentrations. As expected, when PE-MP were present (1 and 10 mg L^{-1}) more pronounced decrease in dissolved 4-NP concentration was observed, presumably due to sorption to the polymer particles.

Likewise, 4-MBC showed an even higher stability in solution than 4-n-NP, with 48 h concentrations in the absence of PE-MP within the same range than initial values (96 to 105%). When high levels of PE-MP (10 mg L^{-1}) were present, dissolved 4-MBC decreased down to 81-90% of initial values, but in the presence of 1 mg L^{-1} there was no clear trend (79-110%).

MP offer a hydrophobic moiety with high surface: volume ratio, ideal conditions to concentrate HOC from the polar aquatic environment by adsorption to the surface and diffusion into the polymeric matrix. In fact, PE membranes are used in passive sampling devices intended to monitor HOC pollution in aquatic habitats. Because of that, the equilibrium partition between PE and water ($K_{PEW=}$ C_{PE} / C_W ; where C_{PE} is the concentration of HOC in g per Kg of PE, and C_W is the dissolved concentration in g per L of water) for many HOC is well known, and log K_{PEW} results to be a linear function of log K_{ow} within the range of log K_{ow} values up to 6 (Lohmann, 2011; Müller et al., 2001) (see also Fig. 3 in (Fries and Zarfl, 2012)). Since the model HOC here selected show log K_{ow} values of 5.76 for 4-n-NP (ECHA, 2012) and 5.1 for 4-MBC (ECHA, 2018), high accumulation of both chemicals in the particulate phase is expected. Therefore, the spontaneous transfer of HOCs will occur in such a direction that the actual concentration ratio (C_{PE}/C_W) approaches the value of the equilibrium partitioning constant (Koelmans et al., 2016). In fact 4-n-NP concentrations as high as 3,936 (Hirai et al., 2011) and 16,000 ng g^{-1} (Mato et al., 2001) were reported in marine plastic particles.

The proportions of the total HOC amount dosed sorbed to PE-microplastics (C_{PE}) in the exposure media can be estimated from the difference in the dissolved concentrations measured at 48 h between samples not containing PE-MP and those containing 1 and 10 mg $_{PE}$ L^{-1}. For example, for the Low 4-n-NP treatment with 1 mg $_{PE}$ L^{-1}, the percentage of 4-n-NP sorbed to PE can be estimated as:

$$\% \text{ sorbed} = [(C_{48} (0 \text{ mg}_{PE} L^{-1}) - C_{48} (1 \text{ mg}_{PE} L^{-1})] \times 100 / (C_{48} (0 \text{ mg}_{PE} L^{-1}))$$

i.e.,

$$\% \text{ sorbed} = [3.9 - 2.1] \times 100 / 3.9 = 46\%$$

Concentrations at t=48 h without PE-MP were considered a better reference than concentrations at t=0 because the former already account for HOC decrease due to sorption to the sample vessels. The resulting values are shown in the last two columns of Table 1.

Thus, the estimated sorption of 4-n-NP to PE-particles after 48h incubation for the concentration of 1 mg $_{PE}$ L^{-1} are 46, 48 and 30% for the Low, Medium and High treatments (Table 1), which correspond to 4-n-NP concentrations in the PE-MP of 1.8 g kg^{-1} PE (Low), 8.2 g kg^{-1} PE (Medium) and 17.8 g kg^{-1} PE (High) . For 10 mg $_{PE}$ L^{-1} the percentages sorbed are similar (41%, 60 % and 37% for Low, Medium and High treatments respectively), resulting concentrations in the PE (C_{PE}) lower than those estimated for 1 mg $_{PE}$ L^{-1} C_{PE}= 0.2 g kg^{-1} PE (Low), C_{PE}= 1.0 g kg^{-1} PE (Medium) and C_{PE}= 2.1 g kg^{-1} PE (High).

Regarding 4-MBC, for the 10 mg $_{PE}$ L^{-1} treatment, a sorption of C_{PE}= 1.1 g kg^{-1} (16% sorption) was obtained for the Low concentration level, C_{PE}= 1.9 g kg^{-1} (12% sorption) for the Medium level, and 7.9 g kg^{-1} (22% sorption) for the High level. In the case of 1_{PE} mg L^{-1}, adsorption could only be estimated for the Low concentration level (C_{PE}= 12 g kg^{-1}, 17% sorption).

Although the experiments where not specifically designed for that purpose, K_{PEW} can be estimated from the data shown in Table 1 as $K_{PEW=} C_{PE} / C_{W.}$ The log K_{PEW} values obtained would be 5.4 ±0.5 (mean±SD) in the case of 4-NP and 4.5 ±0.5 (mean±SD) in the case of 4-MBC, in line with the higher K_{ow} of 4-NP (see above).

Table 1. Actual concentrations (mean±SD) of dissolved 4-n-NP and 4-MBC measured in the water at t=0 h, and after 48 h incubation in absence or presence (1 and 10 mg L^{-1}) of PE-MP. Values without SD correspond to single measurements with no replicates. Estimated sorption (%) of chemicals to PE-MP in the exposure media (see text).

Chemical	Level	Measured concentration (solution) (µg L^{-1})				Estimated sorption to PE-MP (%)	
		Initial (0 h) C_0	Final (48 h) C_{48}			Final (48 h)	
			0	1 mg_{PE} L^{-1}	10 mg_{PE} L^{-1}	1 mg_{PE} L^{-1}	10 mg_{PE} L^{-1}
4-n-NP	Low	4.2±0.6	3.9±0.5	2.1±0.3	2.3±0.3	46	41
	Medium	30±2.4	17±1.4	8.9±0.7	6.9±0.6	48	60
	High	77±6	58±5	41±3	37±3	30	37
4-MBC	Low	72±7	69	57	58	17	16
	Medium	149±23	153±11	150±3	134	nd	12
	High	338±18	354	370	275	nd	22

nd: not determined

3.2. Ingestion of MP by the larvae

Both the sea-urchin and the copepod larvae actively ingested the MP particles. The 2 dpf (days post fertilization) plutei larvae incubated in the absence of MP showed transparent empty stomachs (Fig. 1a), whereas larvae from treatments where MP were present showed dark full stomachs (Fig. 1b). However, the MP CR (mean ± SD; n=3) was 0.5±0.13 mL min^{-1}, less than a half than the *I. galbana* CR measured in the same conditions: 1.2±0.54 mL min^{-1}.

Copepod larvae (Fig. 1c-d) show a less transparent body surface, but ingestion could be documented using and polarized light microscopy. MP particles accumulated in the medium and posterior parts of the digestive track (Fig. 1d).

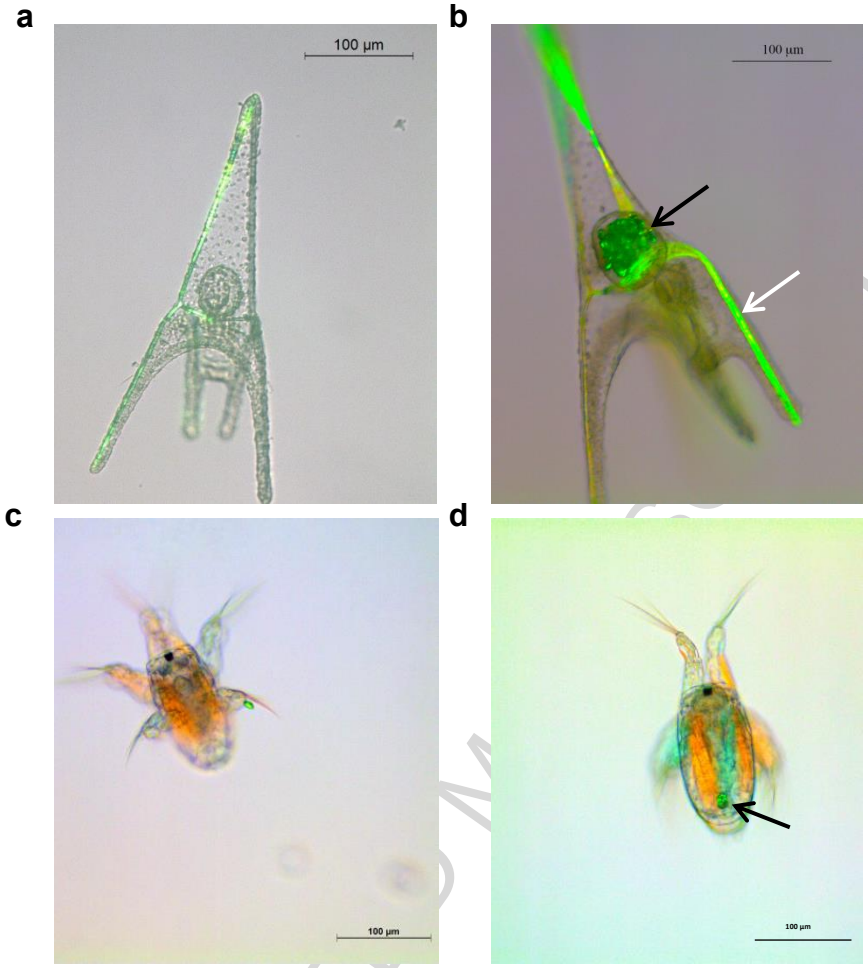


Fig.1 Ingestion of PE-MP. Photographs of 2 dpf *P. lividus* plutei (a, b) and *A. clausi* nauplii (c, d) using bright field and polarized light. Notice the empty digestive system of control treatments (a, c) contrasting with stomachs and gut full of MP in treatments with 10 mg L^{-1} PE-MP (b, d). MP (black arrows) and skeleton rods in plutei (white arrow) appear green under polarized light. Images taken at x100 magnification; scale bars 100 µm.

3.3. Effects of the PE-MP on the uptake of chemicals by the larvae

For both HOC tested, the uptake by the sea-urchin larvae was dependent on the water dissolved concentration ($p<0.001$), and independent of the presence (and levels) of PE-MP ($p= 0.848$ for 4-n-NP, and $p= 0.082$ for 4-MBC). As illustrated in Fig. 2, for both chemicals the uptake, either directly measured in the larvae or indirectly estimated from the decrease in dissolved concentrations during the period of larval exposure, was an approximately linear function of the concentration in water, and the presence of increasing densities of PE-MP (grey and black symbols) did not affect the uptake. These results are in line with those obtained by Besseling et al. (2017), who performed a similar analysis for *Arenicola marina* and also found that bioconcentration and bioaccumulation were only driven by aqueous phase concentration and not ingestion of microplastics.

The mean bioconcentration factor (BCF) obtained using the HOC concentrations measured in the larvae and nominal water concentrations was 6.5±4.8 L kg^{-1} fresh weight for 4-n-NP, and of 482±181 L kg^{-1} fresh weight for 4-MBC. Balmer et al. (2005), reported that different chemicals with similar log K_{ow} showed experimental BCF values differing in more than one order of magnitude. Thus, it is not surprising that 4-n-NP and 4-MBC, despite having similar log K_{ow} result in remarkably different BCF values. The fact that these two HOCs do not belong to the same class of chemicals, one being a phenol and the second being an aldehyde, with very different structures and putative metabolization pathways can contribute to explain the difference in experimental BCF values.

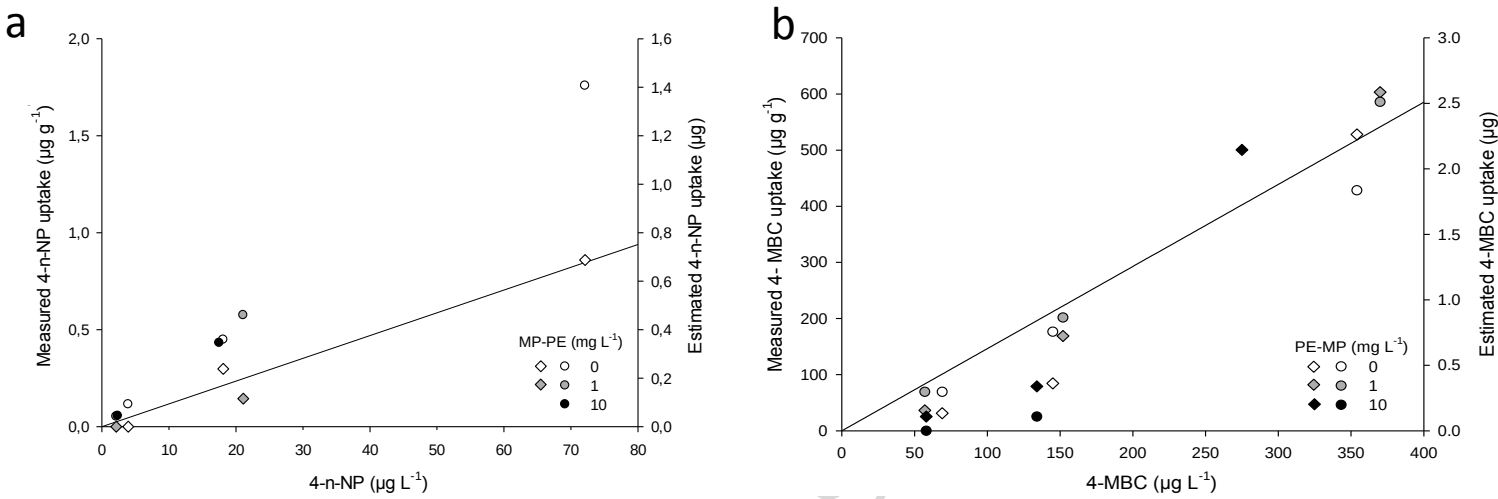


Fig. 2 Measured (diamonds in µg g^{-1}) and estimated (circles in µg) uptake of (a) 4-n-NP and (b) 4-MBC by sea-urchin larvae at different levels of PE-MP in suspension. Measured uptake was fit to linear regression models: y = 0.0.0117x (R^2 = 0.959, p<0.01) and y = 1.464x (R^2 = 0.878, p <0.001) respectively.

In the case of 4-n-NP, the effect of *I. galbana* microalgae on uptake was also tested. Interestingly, the interaction of 4-n-NP with either PE-MP or microalgae, in terms of effects on accumulation in the larvae, was very similar. When larvae were exposed to these two kind of particles of similar size at the same levels (1 mg L^{-1}), the 4-n-NP uptake was independent of the presence of particles (p= 0.791) but dependent on the dissolved water concentration only (p<0.001) (Fig. 3).

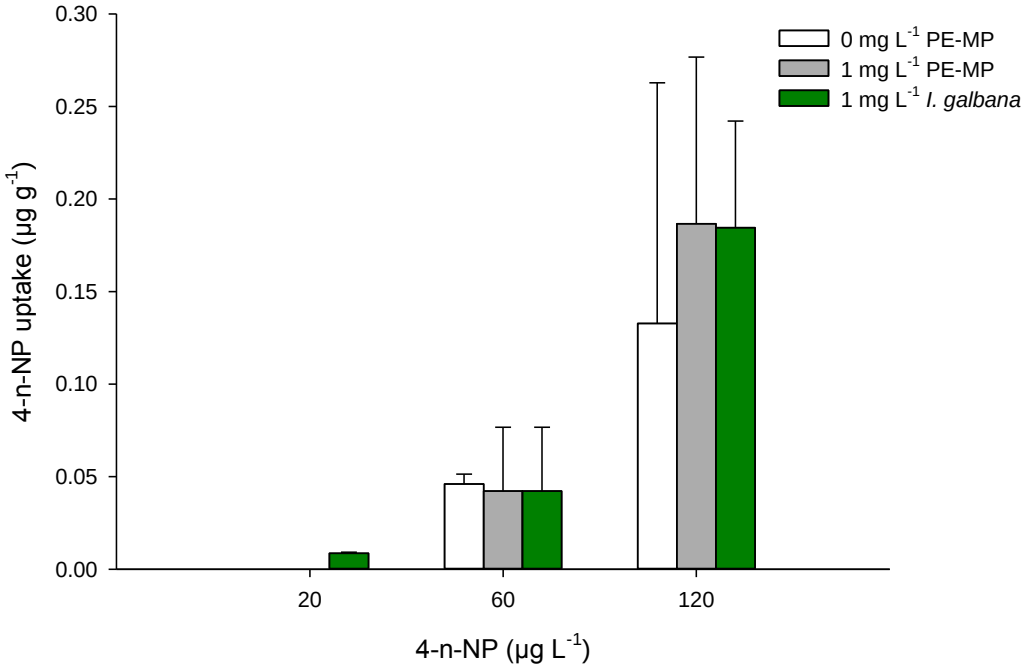


Fig. 3. Comparison of 4-n-NP uptake by sea-urchin larvae exposed to PE-MP and *I. galbana*. Bars represent mean ± SD, N=3.

3.4. Effects of the MP on the toxicity of chemicals to the larvae

For both chemicals tested, the presence of PE-MP did not increase the toxicity on any of the biological models used (Table 2, Fig. 4). In fact, for *A. clausi*, PE-MP tended to reduce the toxicity of 4-n-NP, with LC_{10} values increasing from 15.5 µg L^{-1} in the absence of PE-MP to 43.7 µg L^{-1} at the highest MP level tested. Moreover, survival of *A. clausi* larvae at medium levels of 4-n-NP was significantly (p= 0.020) reduced in the absence of MP, whereas no reduction in survival was detected when MP were present (Fig. 4a). Similarly, at intermediate 4-MBC levels, no *A. clausi* survival was observed in the absence of MP while 6.4 % and 9.2 % survival were recorded at 1 and 10 mg L^{-1} of PE-MP (Fig. 4c). A reduction in the bioaccumulation and toxicity of another HOC, the 17 α-ethynylestradiol, in the presence of MP (but not nanoplastics) was previously reported by Chen et al. (2017) for zebrafish larvae.

Table 2. Lethal or effective concentrations reducing by 10% and 50% the response (LC_x and EC_x, respectively) for 4-n-NP and 4-MBC. All concentrations are µg L^{-1}. The 95% confidence intervals are given in brackets. n.c. not calculable.

		A. clausi		*P. lividus*	
	PE-MP (mg L^{-1})	LC_{10}	LC_{50}	EC_{10}	EC_{50}
4-n-NP	0	15.5 (4.2-24.7)	48.5 (32.6-86.0)	27.6 (13.9-38.3)	61.7 (46.4-83.0)
	1	25.5 (0-46.8)	54.8 (22.0-485)	60.1 (n.c.)	82.5 (n.c.)
	10	43.7 (29.6-54.0)	63.8 (51.1-80.4)	24.1 (7.7-36.7)	59.4 (39.9-90.9)
4-MBC	0	85.2 (n.c.)	107 (n.c.)	116 (4.8 - 229)	1309 (588-10814)
	1	73.8 (53.2-90.6)	131 (110-169)	216 (156-269)	738 (602-985)
	10	89.2 (63.9-109.8)	150 (125-180)	158 (47-252)	858 (555-2454)

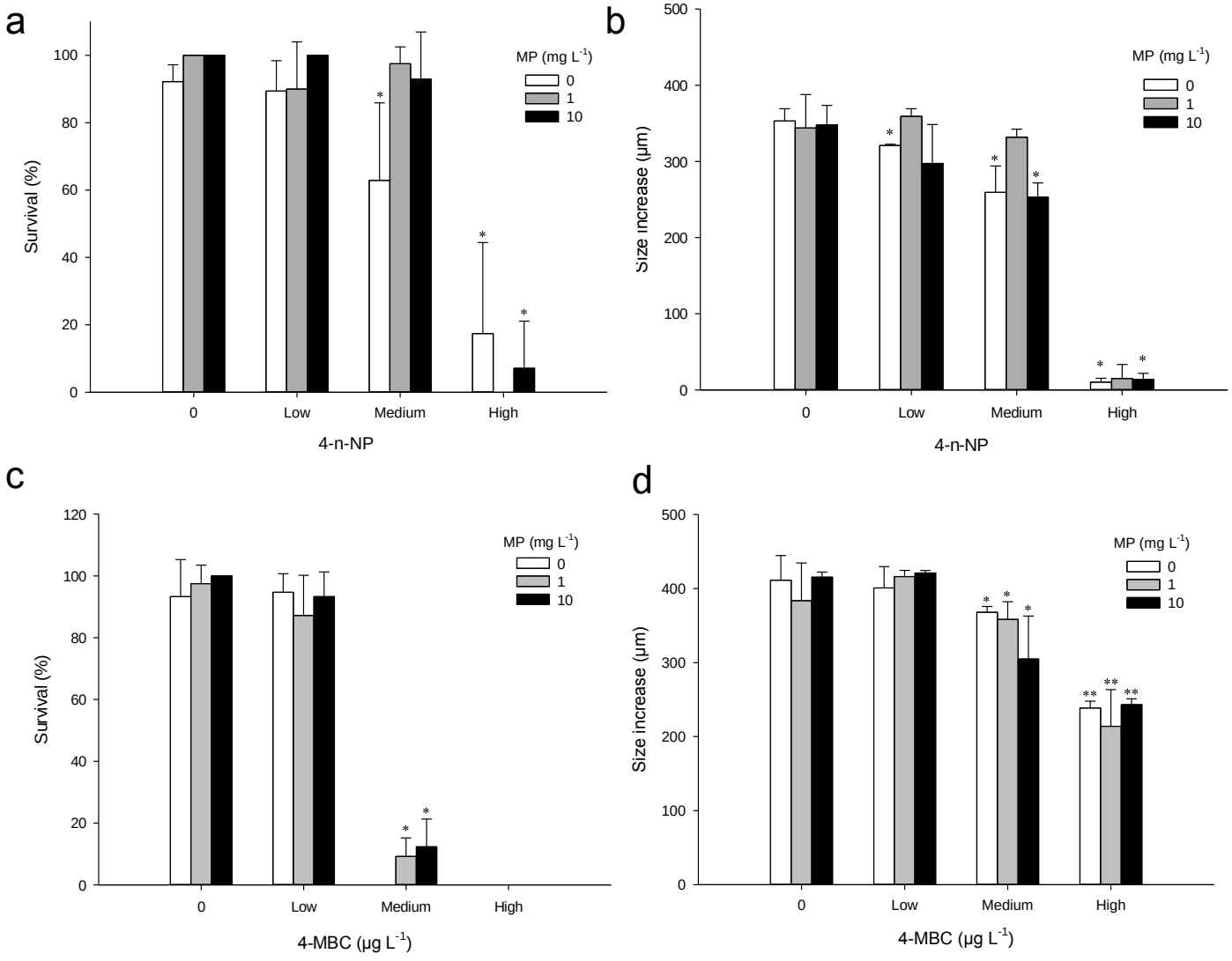


Fig. 4. Toxicity of 4-n-NP to *A. clausi* (a) and *P. lividus* larvae (b), and toxicity of 4-MBC to *A. clausi* (c) and *P.lividus* larvae (d). Toxicity was assessed as reduction in larval size in *P. lividus* and survival in *A. clausi*. Bars represent mean ± SD, N=3-4. Absence of bars indicate 0% survival. Asterisks refer to significant differences to the control treatment. * $p<0.05$, ** $p<0.01$.

It is well known that the most hydrophobic organic chemicals show the slower release from polymers. Experimental results reported in the literature indicate that desorption kinetics from PE-particles in marine environment is generally slow for highly hydrophobic compounds, concluding that the higher the log K_{PEW} (> 5-6), the lesser the fraction desorbed within a given time (Endo et al., 2013; León et al., 2018). In fact, the most recent studies concluded that MP in the environment are expected to act as sinks for HOC (e.g. PAHs, PCBs, PBDEs) and not sources to organisms post ingestion (Bakir et al., 2016; Besseling et al., 2017; Burns and Boxall, 2018; Herzke et al., 2016; Koelmans et al., 2016; Kwon et al., 2017; Lee et al., 2017; León et al., 2019).

Indeed, desorption rate can be crucial for estimating the potential transfer of contaminants from plastic to each marine compartment. Sorption and diffusion of an organic molecule through a polymeric structure depends on the degree of crystallinity of the polymer, with diffusive coefficients increasing as glass transition temperature decreases (George and Thomas, 2001). It is noteworthy, that results presented here refer to HOC with log K_{ow} > 5 and specifically to PE. This polymer has a very low glass transition temperature (T_g= -120 °C) and thus shows mostly amorphous regions at environmental temperatures. This holds true also for PP (T_g= -10 °C). This structural property increases the sorption capacity of PE and PP for hydrophobic chemicals compared to more crystalline polymers showing higher T_g values, such as PVC (T_g = 83 °C) or PS (T_g= 100 °C). Chua et al. (2014) provided strong experimental evidence that PE has a higher affinity for HOC than the gut of marine invertebrates, showing that PCB accumulation in an amphipod was ca. ten times reduced when PCBs were dosed along with PE-MP, compared to seawater-only treatments. Similarly, Albentosa and coworkers, using the same PE-MP that those used in the present study, found a significantly reduced bioavailability of toxicants to both microalgae (Garrido et al.,

2019) and filter feeders (Rivera-Hernández et al., 2019) when MP where present in the exposure media compared to waterborne exposures. Beckingham and Ghosh (2017) found that uptake of PCBs in aquatic worms was lower by 76% when PCBs were associated with PP compared to sediment. Working with fish, Oliveira et al. (2013) found that the addition of PE beads significantly delayed the mortality of juveniles exposed to lethal concentrations of pyrene.

However, the behavior of the PE or PP-sorbed organics along the digestive tract of the aquatic invertebrates may not be representative of that corresponding to other crystalline polymers with a lower affinity for HOC, and hypothetically a higher tendency to desorb the organics during passage through the gut. This was studied through experimental data and modeling in an infaunal worm, *Arenicola marina*, by Koelmans and coworkers. They concluded that, whilst PS particles slightly but significantly increased PCB bioaccumulation in the *A. marina* (Besseling et al., 2013), PE particles were expected to decrease bioaccumulation of organics with log log K_{ow} above 5, approximately (Koelmans et al., 2013a; Koelmans et al., 2013b).

In filter feeders, Avio et al. (2015) found a slightly higher transfer of pyrene to mussel tissues from PS compared to PE, Paul-Pont et al. (2016) reported that PS microparticles had a minor role in transferring fluoranthene to tissues in comparison with waterborne and foodborne exposures, whereas Pittura et al. (2018) showed that BaP bioaccumulation was similar when comparable levels were dosed either via precontaminated PE-MP or waterborne.

The experimental design used in this study was intended to maximize the relevance of the particulate phase for the HOC uptake in the test species. MP levels chosen (1 and 10 mg L^{-1}) are above the range of maximum levels reported for marine waters (between 0.0002 and 0.32 mg L^{-1} (Beiras, 2018)). The MP size range (10^{th} and 90^{th} percentiles of

3.4 and 9.9 µm respectively) is suitable for ingestion for both test species (Helenius and Saiz, 2017; Martínez-Gómez et al., 2017), and smaller –and thus with a higher surface: volume ratio- than MP usually monitored in the sea using manta trawls (>330 µm). Even under those conditions, we found lack of effects of PE-MP as vectors enhancing HOC uptake in zooplankton, and even instances of reduced uptake compared to pure waterborne exposure. In addition, the clearance of plastic particles from water by larvae was markedly lower than that for microalgae of similar size. This contributes to limit the environmental relevance of MP as vectors of HOC in environmentally relevant conditions.

In conclusion, despite active ingestion of PE-MP particles by zooplankton, the presence of these particles did not increase the bioavailability nor the toxicity of the hydrophobic organic pollutants 4-n-NP and 4-MBC compared to waterborne exposures, even at MP concentrations well above those recorded in the marine environment. These findings challenge the current paradigm of MP as vectors of organic pollutants to oceanic trophic webs.

Acknowledgements

This study has received funding by the Spanish Government (MINECO/AEI) through Projects PCIN-2015-187-C03-03 (JPI Oceans EPHEMARE), PCIN-2015-170-C02-01 (JPI Oceans BASEMAN), CTM2016-77945-C3, and CTM2017-84763-C3-2-R, by Xunta de Galicia through "Program of Consolidation and structuring of competitive research groups in the University system of Galicia" (Refs GRC2013-004, ED431C 2017/28 and ED431C 2017/36), and by the European Union through the European Regional Development Fund (ERDF). N. S.-G. was granted with a postdoctoral fellowship Mod. A (2016) by Axencia Galega de Innovación (GAIN), Xunta de Galicia.

The authors acknowledge Damián Costas, Sergio González and all the staff of ECIMAT for their helpful technical support.

References

Avio, C.G., Gorbi, S., Milan, M., Benedetti, M., Fattorini, D., d'Errico, G., et al. 2015. Pollutants bioavailability and toxicological risk from microplastics to marine mussels. Environ. Pollut. 198, 211-222. https://doi.org/10.1016/j.envpol.2014.12.021

Bakir, A., O'Connor, I.A., Rowland, S.J., Hendriks, A.J., Thompson, R.C. 2016. Relative importance of microplastics as a pathway for the transfer of hydrophobic organic chemicals to marine life. Environ. Pollut. 219, 56-65. https://doi.org/10.1016/j.envpol.2016.09.046

Balmer, M.E., Buser, H.-R., Müller, M.D., Poiger, T. 2005. Occurrence of Some Organic UV Filters in Wastewater, in Surface Waters, and in Fish from Swiss Lakes. Environ. Sci. Technol. 39, 953-962. https://doi.org/10.1021/es040055r

Barnes, D.K.A., Galgani, F., Thompson, R.C., Barlaz, M. 2009. Accumulation and fragmentation of plastic debris in global environments. Philo. Trans. R. Soc. 364, 1985-1998. https://doi.org/10.1098/rstb.2008.0205

Beckingham, B., Ghosh, U. 2017. Differential bioavailability of polychlorinated biphenyls associated with environmental particles: Microplastic in comparison to wood, coal and biochar. Environ. Pollut. 220, 150-158. https://doi.org/10.1016/j.envpol.2016.09.033

Beiras, R., 2018. Marine Pollution. Sources, fate and effects of pollutants in coastal ecosystems. Elsevier, Amsterdam.

Beiras, R., Bellas, J., Cachot, J., Cormier, B., Cousin, X., Engwall, M., et al. 2018. Ingestion and contact with polyethylene microplastics does not cause acute toxicity on marine zooplankton. J. Hazard. Mater. 360, 452-460. https://doi.org/10.1016/j.jhazmat.2018.07.101

Beiras, R., Durán, I., Bellas, J., Sánchez-Marín, P. Biological effects of contaminants: *Paracentrotus lividus* sea urchin embryo test with marine sediment elutriates. ICES Tech. Mar. Environ. Sci. Num 51. International Council for the Exploration of the Sea, Copenhagen, Denmark, 2012, pp. 13 pp.

Beiras, R., Tato, T. 2019. Microplastics do not increase toxicity of a hydrophobic organic chemical to marine plankton. Mar. Pollut. Bull. 138, 58-62. https://doi.org/10.1016/j.marpolbul.2018.11.029

Besseling, E., Quik, J.T.K., Sun, M., Koelmans, A.A. 2017. Fate of nano- and microplastic in freshwater systems: A modeling study. Environ. Pollut. 220, 540-548. https://doi.org/10.1016/j.envpol.2016.10.001

Besseling, E., Wegner, A., Foekema, E.M., van den Heuvel-Greve, M.J., Koelmans, A.A. 2013. Effects of Microplastic on Fitness and PCB Bioaccumulation by the Lugworm *Arenicola marina* (L.). Environ. Sci. Technol. 47, 593-600. https://doi.org/10.1021/es302763x

Burns, E.E., Boxall, A.B.A. 2018. Microplastics in the aquatic environment: Evidence for or against adverse impacts and major knowledge gaps. Environ. Toxicol. Chem. 37, 2776-2796. https://doi.org/10.1002/etc.4268

Carpenter, E.J., Anderson, S.J., Harvey, G.R., Miklas, H.P., Peck, B.B. 1972. Polystyrene Spherules in Coastal Waters. Science. 178, 749. https://doi.org/10.1126/science.178.4062.749

Cózar, A., Echevarría, F., González-Gordillo, J.I., Irigoien, X., Úbeda, B., Hernández-León, S., et al. 2014. Plastic debris in the open ocean. PNAS. 111, 10239-10244. https://doi.org/10.1073/pnas.1314705111

Chen, Q., Gundlach, M., Yang, S., Jiang, J., Velki, M., Yin, D., et al. 2017. Quantitative investigation of the mechanisms of microplastics and nanoplastics toward zebrafish larvae locomotor activity. Sci. Total Environ. 584-585, 1022-1031. https://doi.org/10.1016/j.scitotenv.2017.01.156

Chua, E.M., Shimeta, J., Nugegoda, D., Morrison, P.D., Clarke, B.O. 2014. Assimilation of Polybrominated Diphenyl Ethers from Microplastics by the Marine Amphipod, *Allorchestes Compressa*. Environ. Sci. Technol. 48, 8127-8134. https://doi.org/10.1021/es405717z

David, A., Fenet, H., Gomez, E. 2009. Alkylphenols in marine environments: distribution monitoring strategies and detection considerations. Mar. Pollut. Bull. 58, 953-960. https://doi.org/10.1016/j.marpolbul.2009.04.021

ECHA, 2012. Substance Name: 4-Nonylphenol, branched and linear. https://echa.europa.eu/documents/10162/3024c102-20c9-4973-8f4e-7fc1dd361e7d (accessed 11 May 2018)

ECHA, 2018. REACH Dossier 25426. https://echa.europa.eu/registration-dossier/-/registered-dossier/25426 (accessed 30 November 2018)

Endo, S., Yuyama, M., Takada, H. 2013. Desorption kinetics of hydrophobic organic contaminants from marine plastic pellets. Mar. Pollut. Bull. 74, 125-131. https://doi.org/10.1016/j.marpolbul.2013.07.018

Eriksen, M., Lebreton, L.C.M., Carson, H.S., Thiel, M., Moore, C.J., Borerro, J.C., et al. 2014. Plastic Pollution in the World's Oceans: More than 5 Trillion Plastic

Pieces Weighing over 250,000 Tons Afloat at Sea. PLOS ONE. 9, e111913. https://doi.org/10.1371/journal.pone.0111913

Fries, E., Zarfl, C. 2012. Sorption of polycyclic aromatic hydrocarbons (PAHs) to low and high density polyethylene (PE). C. Environ. Sci. Pollut. Res. 19, 1296-1304. https://doi.org/10.1007/s11356-011-0655-5

Garrido, S., Linares, M., Campillo, J.A., Albentosa, M. 2019. Effect of microplastics on the toxicity of chlorpyrifos to the microalgae *Isochrysis galbana*, clone t-ISO. Ecotoxicol. Environ. Saf. 173, 103-109. https://doi.org/https://doi.org/10.1016/j.ecoenv.2019.02.020

George, S.C., Thomas, S. 2001. Transport phenomena through polymeric systems. Prog. Polym. Sci. 26, 985-1017. https://doi.org/10.1016/S0079-6700(00)00036-8

Geyer, R., Jambeck, J.R., Law, K.L. 2017. Production, use, and fate of all plastics ever made. Science Advances. 3, e1700782. https://doi.org/10.1126/sciadv.1700782

Helenius, L.K., Saiz, E. 2017. Feeding behaviour of the nauplii of the marine calanoid copepod *Paracartia grani* Sars: Functional response, prey size spectrum, and effects of the presence of alternative prey. PLOS ONE. 12, e0172902. https://doi.org/10.1371/journal.pone.0172902

Herzke, D., Anker-Nilssen, T., Nøst, T.H., Götsch, A., Christensen-Dalsgaard, S., Langset, M., et al. 2016. Negligible Impact of Ingested Microplastics on Tissue Concentrations of Persistent Organic Pollutants in Northern Fulmars off Coastal Norway. Environ. Sci. Technol. 50, 1924-1933. https://doi.org/10.1021/acs.est.5b04663

Hirai, H., Takada, H., Ogata, Y., Yamashita, R., Mizukawa, K., Saha, M., et al. 2011. Organic micropollutants in marine plastics debris from the open ocean and remote and urban beaches. Mar. Pollut. Bull. 62, 1683-1692. https://doi.org/10.1016/j.marpolbul.2011.06.004

ISO14669, 1999. Water quality: Determination of acute lethal toxicity to marine copepods (Copepoda, Crustacea)Genève, Switzerland.

Koelmans, A.A., Bakir, A., Burton, G.A., Janssen, C.R. 2016. Microplastic as a Vector for Chemicals in the Aquatic Environment: Critical Review and Model-Supported Reinterpretation of Empirical Studies. Environ. Sci. Technol. 50, 3315-3326. https://doi.org/10.1021/acs.est.5b06069

Koelmans, A.A., Besseling, E., Wegner, A., Foekema, E.M. 2013a. Plastic as a Carrier of POPs to Aquatic Organisms: A Model Analysis. Environ. Sci. Technol. 47, 7812-7820. https://doi.org/10.1021/es401169n

Koelmans, A.A., Besseling, E., Wegner, A., Foekema, E.M. 2013b. Correction to Plastic as a Carrier of POPs to Aquatic Organisms: A Model Analysis. Environ. Sci. Technol. 47, 7812-7820. https://doi.org/10.1021/es401169n

Kwon, J.-H., Chang, S., Hong, S.H., Shim, W.J. 2017. Microplastics as a vector of hydrophobic contaminants: Importance of hydrophobic additives. Integr. Environ. Assess. Manage. 13, 494-499. https://doi.org/10.1002/ieam.1906

Langford, K.H., Thomas, K.V. 2008. Inputs of chemicals from recreational activities into the Norwegian coastal zone. J. Environ. Monit. 10, 894-898. https://doi.org/10.1039/B806198J

Lee, H., Chang, S., Kim, S.-K., Kwon, J.-H. 2017. Fugacity analysis of polycyclic aromatic hydrocarbons between microplastics and seawater. Ocean Sci. J. 52, 43-55. https://doi.org/10.1007/s12601-017-0004-9

León, V.M., García-Agüera, I., Moltó, V., Fernández-González, V., Llorca-Pérez, L., Andrade, J.M., et al. 2019. PAHs, pesticides, personal care products and plastic additives in plastic debris from Spanish Mediterranean beaches. Sci. Total Environ. 670, 672-684. https://doi.org/10.1016/j.scitotenv.2019.03.216

León, V.M., García, I., González, E., Samper, R., Fernández-González, V., Muniategui-Lorenzo, S. 2018. Potential transfer of organic pollutants from littoral plastics debris to the marine environment. Environ. Pollut. 236, 442-453. https://doi.org/10.1016/j.envpol.2018.01.114

Lohmann, R. 2011. Critical review of low-density polyethylene's partitioning and diffusion coefficients for trace organic contaminants and implications for its use as a passive sampler. Environ. Sci. Technol. 46, 606-618. https://doi.org/10.1021/es202702y

Lohmann, R. 2017. Microplastics are not important for the cycling and bioaccumulation of organic pollutants in the oceans—but should microplastics be considered POPs themselves? Integr. Environ. Assess. Manage. 13, 460-465. https://doi.org/10.1002/ieam.1914

Lorenzo, J.I., Nieto, O., Beiras, R. 2002. Effect of humic acids on speciation and toxicity of copper to *Paracentrotus lividus* larvae in seawater. Aquat. Toxicol. 58, 27-41. https://doi.org/10.1016/S0166-445X(01)00219-3

Loyo-Rosales, J.E., Rosales-Rivera, G.C., Lynch, A.M., Rice, C.P., Torrents, A. 2004. Migration of nonylphenol from plastic containers to water and a milk surrogate. J. Agric. Food Chem. 52, 2016-2020. https://doi.org/10.1021/jf0345696

Martínez-Gómez, C., León, V.M., Calles, S., Gomáriz-Olcina, M., Vethaak, A.D. 2017. The adverse effects of virgin microplastics on the fertilization and larval development of sea urchins. Mar. Environ. Res. 130, 69-76. https://doi.org/10.1016/j.marenvres.2017.06.016

Mato, Y., Isobe, T., Takada, H., Kanehiro, H., Ohtake, C., Kaminuma, T. 2001. Plastic Resin Pellets as a Transport Medium for Toxic Chemicals in the Marine Environment. Environ. Sci. Technol. 35, 318-324. https://doi.org/10.1021/es0010498

Mazurais, D., Ernande, B., Quazuguel, P., Severe, A., Huelvan, C., Madec, L., et al. 2015. Evaluation of the impact of polyethylene microbeads ingestion in

European sea bass (*Dicentrarchus labrax*) larvae. Mar. Environ. Res. 112, 78-85. https://doi.org/10.1016/j.marenvres.2015.09.009

Müller, J.F., Manomanii, K., Mortimer, M.R., McLachlan, M.S. 2001. Partitioning of polycyclic aromatic hydrocarbons in the polyethylene/water system. Fresenius J. Anal. Chem. 371, 816-822. https://doi.org/10.1007/s002160101025

O'Connor, I.A., Golsteijn, L., Hendriks, A.J. 2016. Review of the partitioning of chemicals into different plastics: Consequences for the risk assessment of marine plastic debris. Mar. Pollut. Bull. 113, 17-24. https://doi.org/10.1016/j.marpolbul.2016.07.021

Oliveira, M., Ribeiro, A., Hylland, K., Guilhermino, L. 2013. Single and combined effects of microplastics and pyrene on juveniles (0+ group) of the common goby *Pomatoschistus microps* (*Teleostei, Gobiidae*). Ecol. Indicators. 34, 641-647. https://doi.org/10.1016/j.ecolind.2013.06.019

Paredes, E., Perez, S., Rodil, R., Quintana, J.B., Beiras, R. 2014. Ecotoxicological evaluation of four UV filters using marine organisms from different trophic levels *Isochrysis galbana*, *Mytilus galloprovincialis*, *Paracentrotus lividus*, and *Siriella armata*. Chemosphere. 104, 44-50. https://doi.org/10.1016/j.chemosphere.2013.10.053

Paul-Pont, I., Lacroix, C., González Fernández, C., Hégaret, H., Lambert, C., Le Goïc, N., et al. 2016. Exposure of marine mussels *Mytilus spp*. to polystyrene microplastics: Toxicity and influence on fluoranthene bioaccumulation. Environ. Pollut. 216, 724-737. https://doi.org/10.1016/j.envpol.2016.06.039

Pittura, L., Avio, C.G., Giuliani, M.E., d'Errico, G., Keiter, S.H., Cormier, B., et al. 2018. Microplastics as Vehicles of Environmental PAHs to Marine Organisms: Combined Chemical and Physical Hazards to the Mediterranean Mussels, *Mytilus galloprovincialis*. Frontiers in Marine Science. 5. https://doi.org/10.3389/fmars.2018.00103

Rivera-Hernández, J.R., Fernández, B., Santos-Echeandia, J., Garrido, S., Morante, M., Santos, P., et al. 2019. Biodynamics of mercury in mussel tissues as a function of exposure pathway: natural vs microplastic routes. Sci. Total Environ. 674, 412-423. https://doi.org/10.1016/j.scitotenv.2019.04.175

Salgueiro-González, N., Concha-Graña, E., Turnes-Carou, I., Muniategui-Lorenzo, S., López-Mahía, P., Prada-Rodríguez, D. 2012. Determination of alkylphenols and bisphenol A in seawater samples by dispersive liquid–liquid microextraction and liquid chromatography tandem mass spectrometry for compliance with environmental quality standards (Directive 2008/105/EC). J. Chromatogr. 1223, 1-8. https://doi.org/10.1016/j.chroma.2011.12.011

Salgueiro-González, N., Turnes-Carou, I., Besada, V., Muniategui-Lorenzo, S., López-Mahía, P., Prada-Rodríguez, D. 2015a. Occurrence, distribution and bioaccumulation of endocrine disrupting compounds in water, sediment and biota samples from a European river basin. Sci. Total Environ. 529, 121-130. https://doi.org/10.1016/j.scitotenv.2015.05.048

Salgueiro-González, N., Turnes-Carou, I., Viñas-Diéguez, L., Muniategui-Lorenzo, S., López-Mahía, P., Prada-Rodríguez, D. 2015b. Occurrence of endocrine disrupting compounds in five estuaries of the northwest coast of Spain: Ecological and human health impact. Chemosphere. 131, 241-247. https://doi.org/10.1016/j.chemosphere.2014.12.062

Salgueiro-González, N., Turnes-Carou, I., Viñas, L., Besada, V., Muniategui-Lorenzo, S., López-Mahía, P., et al. 2016. Occurrence of alkylphenols and bisphenol A in wild mussel samples from the Spanish Atlantic coast and Bay of Biscay. Mar. Pollut. Bull. 106, 360-365. https://doi.org/10.1016/j.marpolbul.2016.03.003

Sánchez Rodríguez, A., Rodrigo Sanz, M., Betancort Rodríguez, J.R. 2015. Occurrence of eight UV filters in beaches of Gran Canaria (Canary Islands). An approach to environmental risk assessment. Chemosphere. 131, 85-90. https://doi.org/10.1016/j.chemosphere.2015.02.054

Setälä, O., Fleming-Lehtinen, V., Lehtiniemi, M. 2014. Ingestion and transfer of microplastics in the planktonic food web. Environ. Pollut. 185, 77-83. https://doi.org/10.1016/j.envpol.2013.10.013

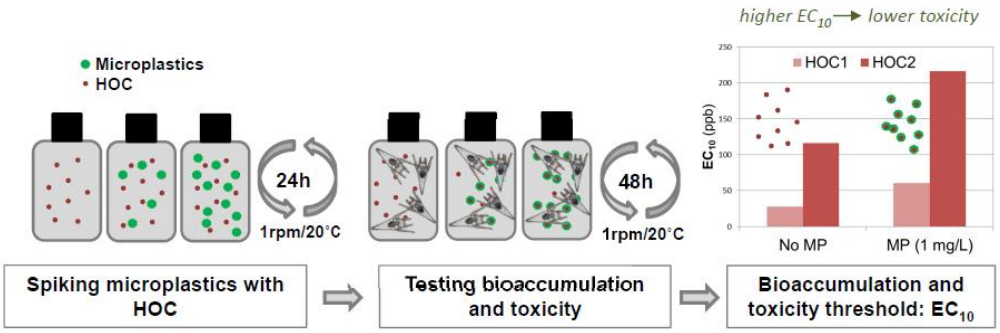
Tato, T., Salgueiro-González, N., León, V.M., González, S., Beiras, R. 2018. Ecotoxicological evaluation of the risk posed by bisphenol A, triclosan, and 4-nonylphenol in coastal waters using early life stages of marine organisms (*Isochrysis galbana*, *Mytilus galloprovincialis*, *Paracentrotus lividus*, and *Acartia clausi*). Environ. Pollut. 232, 173-182. https://doi.org/10.1016/j.envpol.2017.09.031

Vidal-Liñán, L., Villaverde-de-Sáa, E., Rodil, R., Quintana, J.B., Beiras, R. 2018. Bioaccumulation of UV filters in *Mytilus galloprovincialis* mussel. Chemosphere. 190, 267-271. https://doi.org/10.1016/j.chemosphere.2017.09.144

Wedgewood, A.R., Seferis, J.C. 1984. A quantitative description for the optical properties of crystalline polymers applied to polyethylene. Polym. Eng. Sci. 24, 328-344. https://doi.org/10.1002/pen.760240507

Ziccardi, L.M., Edgington, A., Hentz, K., Kulacki, K.J., Kane Driscoll, S. 2016. Microplastics as vectors for bioaccumulation of hydrophobic organic chemicals in the marine environment: A state-of-the-science review. Environ. Toxicol. Chem. 35, 1667-1676. https://doi.org/10.1002/etc.3461

Graphical abstract



Highlights

- Sea-urchin pluteus and copepod nauplius larvae actively ingest microplastic particles.
- Microplastics did not increase accumulation of organic chemicals in sea-urchin larvae.
- Microplastics did not increase the toxicity or 4-n-NP or 4-MBC to zooplankton.
- PE microplastics do not act as vectors of hydrophobic organics to zooplankton.