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Organochlorine Compounds in Beached Plastics and Marine Organisms

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Here we compare bioaccumulation factors in marine organisms to partition ratios in marine debris for dichlorodiphenyltrichloroethane and polychlorinated biphenyls. Both organochlorines are synthetic persistent organic pollutants emitted into the environment since the beginning of the last century in approximately equal amounts. Their vast use and dispersion have resulted in approximately similar median concentrations of the two organochlorines in some pelagic organisms, namely in the liver and muscle tissue of fish. Molluscs, on the other hand, show higher median uptake of PCBs (median = 2.34 ng/g) than of DDTs (median = 1.70 ng/g), probably reflecting more localized conditions. We found that the bioaccumulation factors can be several orders of magnitude higher than the partition ratios. For instance, the median concentrations of organochlorines in the different matrices of fish, birds, and mammals are between one to four orders of magnitude higher than those found in marine debris, when lipid-normalized; or up to two orders of magnitude when measured as wet-weight. But, in molluscs, bioaccumulation/partition equals unity, which agrees with previous studies using passive samplers. Future research should focus on reducing sources of uncertainty by 1) homogenization of chemical procedures; 2) better assessment of chemical partition equilibrium between water and polymers in environmental conditions; 3) use of (multi) polymer passive samplers better aimed at mimicking uptake of specific living tissues.

Keywords: marine organisms, marine plastic debris, PCB, DDT, passive sampler, OSPAR commission, pellet watch

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1 INTRODUCTION

The performance of passive samplers to predict concentrations of persistent contaminants in organisms other than bivalves was considered for some time as unclear (USEPA, 2012a; ICES, 2013). However, more recent studies have found good agreement between concentrations in passive equilibrium samplers (PES) and those in fish (Cerveny et al., 2016; Rusina et al., 2017). This opens the possibility of finding similar results for other organisms, allowing biota monitoring to be supplemented by PES and improving contaminant modelling and risk assessment at the higher trophic levels.

In the present work, we explore this biota-PES relationship using a synoptic approach using large environmental databases for the northeast Atlantic region, as opposed to the most frequent of local case-study analysis (ICES, 2013). The synoptic approach complements the latter by "averaging" local results and expanding the analysis for a larger set of marine organisms.

For the synoptic analysis, two hydrophobic organochlorine compounds (OCCs) were chosen: dichlorodiphenyltrichloroethane (DDT) and polychlorinated biphenyls (PCB), analysed in marine

organisms and in beached plastic pellets. The latter behave like mobile PES (Ogata et al., 2009; Shi et al., 2020). The study focused on the North-East Atlantic, corresponding to the territorial seas of the 16 Contracting Parties of the OSPAR Convention (from Oslo and Paris Conventions, "OS" for Oslo and "PAR" for Paris) (www.ospar.org), expanding over a total area of 13.5 million km^2.

DDT, which has been marketed since the early 1940s, remains as one of the twelve insecticides currently recommended for disease control exclusively by the World Health Organization (WHO) and allowed under the Stockholm Convention on Persistent Organic Pollutants. Global emissions of DDT in the period of 1950–1990 were estimated to be between 1.2×10^6 and 1.5×10^6 tons (UNEP, 2003; Semeena and Lammel, 2005). Since 2003, there have been three countries reporting DDT production, namely, India, China, and the Democratic People's Republic of Korea. From 2003 to 2007, average DDT production was around 5×10^3 tons per year, diminishing further from 2008 to 2014, to about 3.5×10^3 tons per year (Van Den Berg et al., 2017), after China and DPR Korea phased out their production. Between 1950 and 2020, the total global emissions may amount to 1.24×10^6 to 1.54×10^6 tons. Half-lives are estimated to be 180 days in water, 360 days in soil, 4.44 years in sediment, and 3.1 days in the atmosphere (USEPA, 2012b). Mansouri et al. (2017) report longer environmental half-lives, between 2 and 15 years for p,p -DDT and o,p –DDT. Degradation of DDT in environmental compartments is notoriously slow and is close to zero in seawater. Some researchers assume that DDT removal from the environment is by degradation in soil only, represented as a first-order process (4.05×10^{-9} s^{-1} at 298 K, equivalent to a half-life of about 5.4 years, doubling per 10 K of temperature increase) (Hornsby et al., 1996). Background concentrations of DDT are suggested to be 1.0 ng/g (0.0 log units) wet weight in blue mussels, 200 (2.3 log units) μg/g wet weight in cod liver, and 50 ng/g (1.7 log units) wet weight in herring muscle (EEA, 2003).

Polychlorinated biphenyls (PCBs) are a group of man-made oily liquids or solids, with no smell or taste. They are very stable mixtures that are resistant to extreme temperature and pressure, have low flammability, a high boiling point and electrical insulating properties. Thus, PCBs have many applications in electrical equipment like capacitors and transformers, and in hydraulic fluids, heat transfer fluids, lubricants, and plasticizers. Their production began in the late 1920s and continued until its ban, first by the USA in 1977, and later by the global ban under the Stockholm Convention in 2001, which entered into force in 2004. During this period it is estimated that about 2×10^6 tons of PCBs have been produced, of which about 2×10^5 tons remain in mobile environmental reservoirs (WHO, 2003). Half-lives for PCB 153, used here as a proxy for the sum of PCBs, are the same as DDT, except for the atmosphere, where a half-life of 65.4 days (USEPA, 2012b) allows for faster atmospheric mobility of PCB away from the location of release. The persistence of PCB congeners increases as the degree of chlorination and structural uniformity increase. While PCBs are very resistant to abiotic degradation, biodegradation occurs under both aerobic and anaerobic conditions, mostly in sediments and soils (WHO, 2003). Background concentrations of PCB are 2.0 ng/g (0.30 log units) wet weight in blue mussels, 100 μg/g (2.0 log units) wet weight in the cod liver, and 150 ng/g (2.18 log units) wet weight in herring muscle (EEA, 2003).

In general there have been decreasing trends in environmental concentrations of OCCs, although geographic differences are pronounced (UNEP, 2003). The leaking of PCB-rich fluids from equipment, industrial facilities, and incorrect disposal may be the cause for their slower environmental diminution in some places. For instance, the high atmospheric deposition rates reported in the Baltic Sea (10–15 ng/m^2/day) reflect localized sources. In contrast, lower rates of PCB deposition in European and Mediterranean regions (ranging from 1.2 to 5.6 ng/m^2/day), is probably due to the generalized mechanisms of atmospheric circulation in the area (UNEP, 2003). As for DDT, its use for vector control is still contributing to a localized increase in environmental concentrations.

In its 2019 assessment, the European Environment Agency concluded that (EEA, 2019) concentrations of DDT in recent years have generally been moderate in mussels and fish from the North-East Atlantic Ocean and Baltic Sea, and in mussels from the Mediterranean Sea. In the North-East Atlantic Ocean, localized high concentrations of DDT may pose a risk to marine organisms. A general downward trend was found for the North-East Atlantic Ocean from 2003 to 2014, but no general trends were detected from 2008 to 2017, which indicates that the impact of abatement policies, as a whole, may have stabilized. Concentrations of PCBs in recent years have been lower, except for localised high concentrations found in mussels and fish in the North-East Atlantic Ocean and Baltic Sea, and in mussels in the Mediterranean Sea. A general downward trend was found for the North-East Atlantic Ocean and the Baltic Sea from 2004 to 2014, which only continued in the Baltic.

In coastal waters, the input of chemicals from land via discharges of sewage and industrial effluents and rivers are the main sources of OCCs, whereas atmospheric deposition is the major pollutant source in open seawaters (Scrimshaw and Lester, 1996). Consequently, their concentrations in seawater show decreasing gradients offshore, in the low pg/L range which makes reliable quantification difficult. Being very hydrophobic, these OCCs accumulate in sorbing materials including the organic fraction present in suspended matter, living tissues, and synthetic polymers. Thus, bivalve molluscs are being used as sentinel organisms to reflect water quality, bioavailability and bioaccumulation impacts on edible species (Lohmann and Muir, 2010), though it is impossible to find one single species that could be used across the entire world. The limitations of these "living samplers" led to the development of non-living passive samplers, which are considered as promising surrogates for organisms, mimicking bioaccumulation (Figueiredo et al., 2017). Many different devices exist for sampling organic contaminants in water (Vrana et al., 2005), but those using single-phase polymers have been increasingly used, including silicone, polyoxymethylene, and Polyethylene (PE) (Lohmann, 2012).

Plastic pellets are a nuisance to marine life. Their ingestion can cause physical harm, such as internal injuries and impaired ability to breathe, swallow, digest food properly, or immediate death. Following ingestion, hazardous chemicals can be transferred to fish, inducing hepatic stress (OSPAR Comission, 2018). To better

TABLE 1 | DDT samples in different matrices in organisms and plastic pellets.

	Matrix	Code[a]		Nb. of samples			
				DDTs		PCBs	
			Basis[b]	W	L	W	L
Organisms	Blubber	BB		279		362	
	Blood cells	BC		20		—	
	Blood	BL		16		16	
	Blood serum	BS		20		—	
	Egg(s)	EG			100	—	
	Epidermis (skin)	EP		—		5	
	Fat	FA		77		73	
	Kidney	KI		—		5	
	Liver	LI		1750	1376	342	126
	Muscle	MU		916	3538	402	915
	Roe (fish eggs)	RO		4		—	
	Whole soft-body	SB		120	195	212	39
	Miscellaneous	SI		2		2	
	Tail muscle	TM		3		5	
	Whole organism	WO		47		5	
	Total			**3254**	**5209**	**1429**	**1080**
Pellets	Atlantic Ocean	Atl		86		134	
	Indic Ocean	Ind		29		49	
	Mediterranean Sea	Med		19		47	
	North Sea	North		11		13	
	Pacific Ocean	Pac		112		140	
	Total			**257**		**383**	

[a]*For a complete list of matrix code see https://vocab.ices.dk/under "Matrix."*
[b]*Basis: lipid weight (i.e., fat weight); wet weight (i.e., fresh weight).*

understand the exposure of marine life to chemicals sorbed in plastic debris, beached plastic resin pellets were proposed as easily collectible, low-cost passive samplers for some persistent organic pollutants within the International Pellet Watch project (Ogata et al., 2009). Project participants around the world voluntarily collect plastic resin pellets from beaches and send them to a centralized laboratory in Japan via airmail. The project constitutes the largest repository of such information currently available on a global scale.

In this article, we estimate concentrations in marine organisms and beached plastic pellets using data from publicly available databases. From these, the ratios between concentrations in organisms and those in marine pellets are computed and discussed, including the relative contribution of uncertainty sources. Finally, knowledge gaps are identified for future research.

2 MATERIALS AND METHODS

Data on total DDT (DDTs) and total PCB (PCBs) concentrations in marine organisms were retrieved from the International Council for the Exploration of the Sea (ICES), freely available at the institution's website (https://data.ices.dk/). The exhaustive dataset includes organisms that rely exclusively on the sea to feed and others that may eventually feed inland, such as many coastal birds and polar bears. The dataset contains information regarding date of sampling; location of the sampling point, in decimal geographic coordinates; concentration of the chemical (ng/g); identification of the analysed species (*Genera species*); matrix where the sample was taken (see **Table 1**); the basis of determination (wet weight, lipid weight). We added one more variable: animal group (a code to help identify the type of animal: arthropod, bird, fish, mammal, mollusc).

The concentration of the substance in marine organisms is represented by $M_{s,o,m,b}$, (ng/g), and in marine pellets by P_s (ng/g), with s the substance (DDTs, or PCBs), o the organism, m the matrix (see **Table 1**), and b the indication whether the concentrations are reported as wet weight or lipid-normalized. DDTs includes DDT (p,p′), DDE (p,p′), DDD (p,p′), DDT (o,p′), DDE (o,p′), DDD (o,p′). PCBs in the Pellet Watch database are the sum 13 congeners IUPAC (nos. CB66, 101, 110, 149, 118, 105, 153, 138, 128, 187, 180, 170, 206); while the ICES includes 7 congeners, CB28, 52, 101, 118, 138, 153, 180. The two databases include a different number of congeners which introduces some uncertainty when comparing their results. They are not incompatible for the present study, as is it possible to estimate the intervals for the uncertainty. For instance, the relationship between the standard NOAA list of 18 congeners and a "True" PCB value was investigated (NOAA, 1991, 1989) in sediments and mussels, which concluded that the sum of the 18 congeners amounts to about half of the total PCBs present in the sample. This ratio was confirmed by Lefkovitz et al. (2001) for samples of lobster hepatopancreas. However, in a food web study performed with tree swallows (*Tachycineta bicolor*) in the Hudson River

area, New York state, U.S., the sum of seven congeners accounted rather constantly for all biota between 20 and 34% of the total PCBs. It can be expected, then, that the sum of PCBs provided by Pellet Watch is between 1.5 and 2.5 times higher than that of ICES, or around 0.3 log units.

The dataset for concentrations of DDTs and PCBs in marine pellets was retrieved from the International Pellet Watch (IPW)—Global monitoring of OCCs by using beached plastic resin pellets (http://pelletwatch.org/), based in Japan. IPW has been processing samples from all over the world since 2006 (Ogata et al., 2009). The PCB concentration reflects the sum of the 18 congeners mentioned above. A uniform distribution of the different congeners is assumed.

The Kruskal-Wallis non-parametric method was used for comparison of medians, followed, when necessary, by Dunn's post hoc test, with a significance level of 1%. Calculations were made in the R suite using package FSA. Central tendency statistic is represented by the median and dispersion by the coefficient of interquartile variation (cqv), given by

$$cqv = \frac{(p75 - p25)}{(p75 + p25)} \tag{1}$$

with p25 and p75 the 25th and 75th percentiles of the data distribution, respectively.

The ratios, $R_{s,o,m,b}$, of concentration of the substance in marine organisms, $M_{s,o,m,b}$, (ng/g) to that in the pellets, P_s (ng/g) was computed by

$$R_{s,\,o,\,m,b} = \frac{\mathrm{Log\,M}_{s,\,o,\,m,b}}{\mathrm{Log\,P}_s} \tag{2}$$

Log M and Log P are probability density functions estimated from the original dataset. The posterior density, R. Given that measurements in pellets and marine organisms come from different sampling locations, measured at different times, they cannot be compared directly. Instead, the statistical distributions of M and P were modelled with theoretical statistical distributions followed by repeated random sampling and recomputation of R (Monte Carlo simulations). Oracle's Crystal Ball® suite was used to facilitate the computations.

At equilibrium, the partition constant between P_s (ng/kg) and water concentration (free form), C_w (ng/L) is

$$K_P = \frac{\mathrm{P}_s}{C_w} \tag{3}$$

The value of K_p varies with the properties of each polymer and the affinity of the sorbing substance. Here we use the K_P proposed by (Lohmann, 2012) corrected by an ionic strength factor of plus 50%, for a Setschenow (1889) constant of 0.35 (Xie et al., 1997), to convert from freshwater to ocean water. About 90% of the pellets analysed under the Pellet Watch program were of polyethene (PE) (Ogata et al., 2009), so only the K_{PE} is henceforth used and applied to all pellets. This latter assumption brings in some uncertainty to the analysis. It may be somewhat limited by the fact that long-term sorption coefficients of very hydrophilic substances in the ocean are similar for plastics that constitute the large majority of the found pellets, namely low-density polyethene (LDPE), high-density polyethene (HDPE) and polypropylene (PP) (Rochman et al., 2013; O'Connor et al., 2016; Allen et al., 2018).

Bioconcentration factors (BCF) are calculated by considering contaminant tissue concentrations with respect to environmental water concentrations. BCF (L/kg) are commonly lipid normalized by accounting for the concentration of lipids in the organism:

$$BCF = \frac{(M_{s,o,m}/V_L)}{C_w} \tag{4}$$

with V_L the fraction of lipid in the organism, and C_w is the concentration of the substance in seawater (ng/L).

For chemicals that accumulate to a significant extent via diet, bioaccumulation can be estimated from the bioaccumulation factor (BAF) using food chain multipliers (FCM), which for substances with Log K_{OW} between 6.0 and 7.5 are 13 and 23–24 for trophic level 3 (planktivorous) and level 4 (piscivorous fish), respectively (Boethling and Mackay, 2000; USEPA, 2000):

$$BAF = \frac{BCF \cdot FCM}{C_w} \tag{5}$$

The ratio between the estimated bioaccumulation factor and the partition constant K_{PE},

$$R_s^* = \frac{Log\ BAF_s}{Log\ K_{PE,s}} \tag{6}$$

is a theoretical estimate of $R_{s,o,m,lipid}$ at chemical equilibrium, therefore $R \approx R^*$.

3 RESULTS AND DISCUSSION

3.1 OCCs in Marine Organisms

Data for lipid-normalized DDTs in the ICES database is available for a large number of different organisms and matrices, but statistical analysis showed that they could be aggregated into four statistical populations (Kruskal-Wallis test, K-W χ^2 (3) = 1399.1, $p < 2 \times 10^{-16}$, Dunn post hoc test, $p < 0.01$—see **Supplementary Material** for detailed statistical results), $M_{DDT,\,o,\,m,\,lipid}$. They are (**Figure 1A**; **Table 2**): 1) bird eggs (EG), with the highest concentrations; followed by 2) liver (LI), 3) muscle (MU), and finally 4) the whole soft body (SB). The differences result from different factors: samples coming from different organisms, with different feeding habits and position in the food web, and measurements made in different matrices (**Table 1**). To compare accumulation of DDTs in marine organisms and plastic pellets, the mentioned factors were considered separately.

Median concentrations of DDTs in eggs in the dataset are 4.28 log units (ng/g) (= $10^{4.28}$ ng/g) (**Figure 1A**; **Table 2**). At these high concentrations, eggshell thinning has been observed in birds (due to the presence of o,p' DDT). In avian wildlife, thinning can result from a functional malformation in the shell gland, induced by embryonic exposure to estrogenic substances (Holm et al., 2006). Long term effects of eggshell thinning are a

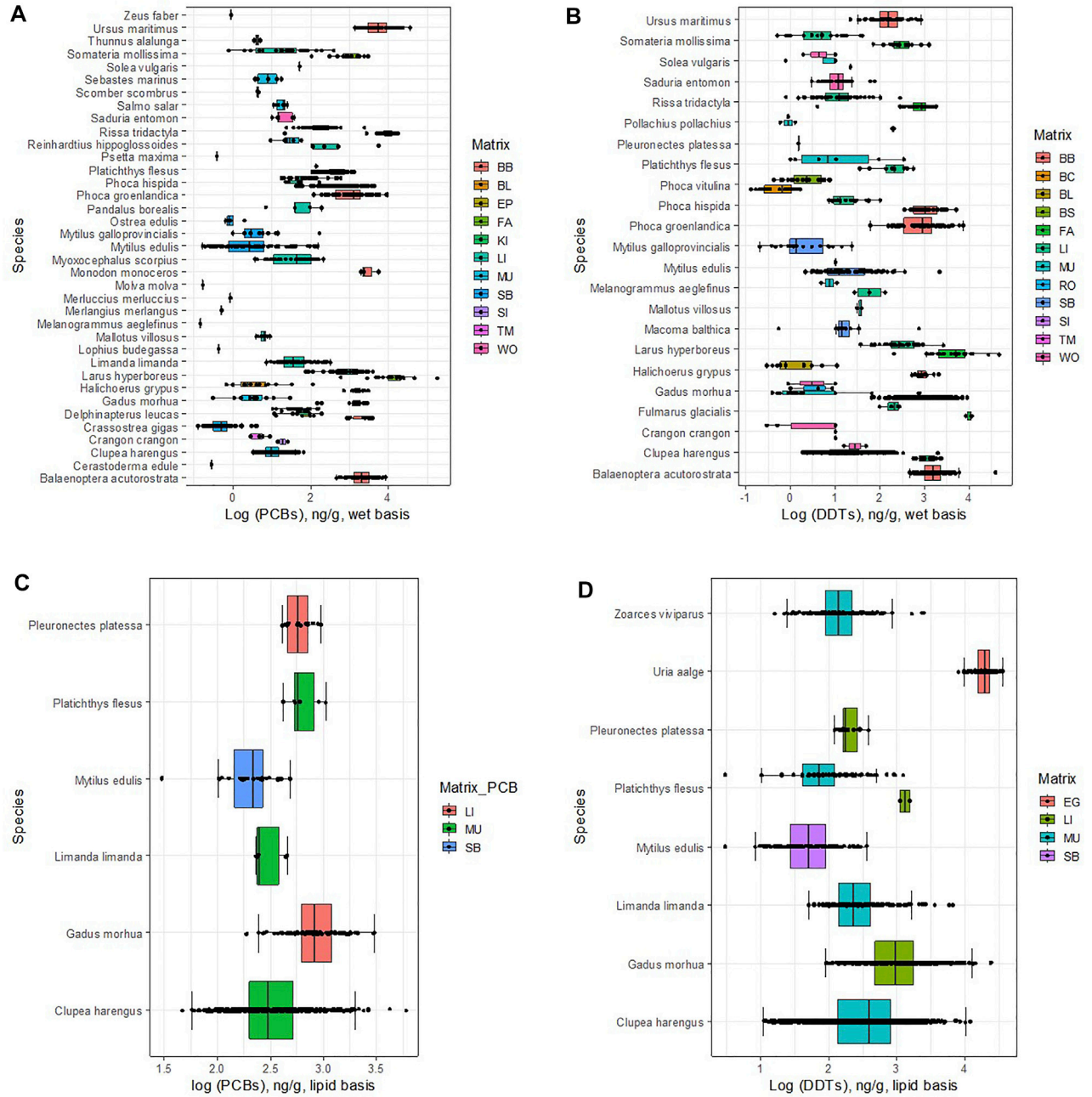

FIGURE 1 | Concentration of DDTs and PCBs in different matrices in marine organisms. Lipid basis: **(A)** $M_{DDT,o,m,lipid}$.; **(B)** $M_{PCB,o,m,lipid}$. Wet basis: **(C)** wet basis, $M_{DDT,o,m,wet}$.; **(D)** $M_{PCB,o,m,wet}$. For a list of matrix, codes see **Table 1**.

drastic decline in the numbers of birds of prey (Mitra et al., 2011). Elevated DDTs concentrations in waterbird eggs have also been found in heavily industrialized marine environments. For instance, in a study in Hong Kong, Wang et al. (2011) found concentrations in the range 3.40–4.21 log units (ng/g), for eggs of the Night Heron (*Nycticorax*) and the Great Egret (*Egretta alba*), respectively. In central Canada, values of 5 log units (ng/g) were reported for Herring gulls (*Larus argentatus*) (Hellou et al., 2013).

Accumulation of DDTs in the fish liver in the OSPAR region (2.95 log units, ng/g), surpasses by more than one order of magnitude those found in remote places of the planet, as reported by Lana et al. (2014) for Antarctic *Trematomus newnesi*, *Notothenia coriiceps*, and *Notothenia rossii,* which were below 1.4 log units (ng/g); or off Pernambuco, north-eastern Brazil, where concentrations of 1.5 log units (ng/g) were found in the liver of king mackerel (*Scomberomorus cavalla*) (Miranda and Yogui, 2016).

TABLE 2 | Concentration of DDTs and PCBs in marine organisms ($M_{s,o,m,b}$).

Basis	Matrix (*m*)	Organism (group) (*o*) (Median; interquartile range; cqv)	Sample size, N
Lipid base	DDTs		
	Eggs (bird) (EG)	Birds (4.28; 0.153 ng/g; 0.02)	100
	Liver (LI)	Fish (2.95; 0.609 ng/g; 0.01)	1376
	Muscle (MU)	Fish (2.47; 0.791 ng/g; 0.16)	3538
	Soft whole body (SB)	Mollusc (1.70; 0.513 ng/; 0.15)	195
	PCBs		
	Liver (LI)	A: Fish (2.90; 0.312 ng/g; 0.05)	126
	Muscle (MU)	A: Fish (2.48; 0.409 ng/g; 0.08)	915
	Soft whole body (SB)	A: Mollusc (2.34; 0.267 ng/g; 0.06)	39
Wet weight base	DDTs		
	Fat and blubber (FA, BB)	Birds and mammals (2.97; 0.609 ng/g; 0.10)	N (bird) = 77; N (mammal) = 279
	Liver (LI)	A: Birds and mammals (1.20; 0.652 ng/g; 0.37 B: Fish (2.93; 0.430 ng/g; 0.07)	N (bird) = 266; N (mammal) = 45; N (fish) = 1436
	Muscle and whole body (MU, WO, SB)	Arthropod, fish, and molluscs (1.18; 0.644 ng/g; 0.27)	N (arthropod) = 35; N (fish) = 928; N (mollusc) = 120
	Blood (BC, BL, BS)	Mammal (0.011; 0.600 ng/g; 4.24)	56
	Tail muscle and fish roe (TM, RO)	Fish (1.00; 0.388 ng/g; 0.18)	9, of which 8 are for roe
	PCBs		
	Fat (FA)	A: Bird (3.97; 0.896 ng/g; 0.09)	73
	Blubber (BB)	A: Mammals (3.18; 0.661 ng/g; 0.10)	362
	Liver (LI)	A: Bird, arthropod, fish, and mammal (2.08; 1.09 ng/g; 0.26)	N ((bird) = 101; N (arthropod) = 5; N (fish) = 226; N (mammal) = 10
	Muscle and blood (MU, BL)	A: Fish (0.982; 0.340 ng/g) B: Mammal (1.56; 0.852 ng/g; 0.22)	N (fish) = 362; N (mammal) = 56
	Soft whole body (SB)	A: Molluscs (0.167; 0.896 ng/g; 2.26)	N (mollusc) = 212

*See **Supplementary Material**, for detailed statistical results.*

Fish muscle samples showed lipid-based DDTs concentrations in the order of 2.47 log units (ng/g), which are similar to those found in other areas with high anthropogenic influence. For instance, Sun et al. (2020) reported values between 1.96 and 2.35 log units of yellowfin tuna (*Thunnus albacares*) and their prey from the South China Sea. In croakers and mullets collected in Guanabara Bay, Rio de Janeiro and Araújo Island, Paraty, Silva et al. (2016) reported (mean) DDTs concentrations in the range 2.55–3.58 log units (ng/g). Concentrations in the OSPAR region are, however, one to two orders of magnitude above those found in more pristine regions of the globe. In Antarctica, DDTs (mean) concentrations did not surpass 1.1 log units (ng/g) in *Trematomus newnesi*, *Notothenia coriiceps*, and *Notothenia rossii* (Lana et al., 2014). In Tanzania (western Indian Ocean) median DDTs concentrations found in the muscle of Mtwara milkfish (*Chanos*) and Pemba mullets (*Mugil cephalus*) were equal to 0.12 and 1.85 log units (ng(g), respectively (Mwakalapa et al., 2008).

In the OSPAR region, median concentrations in molluscs (mainly bivalves) are less elevated than those found in other equally industrialized regions, such as in the continental coast of the United States where median DDTs in mussels ranged between 2.30 and 2.17 log units (ng/g) (Sericano et al., 2014) (values converted from dry weight using wet weight/dry weight ratios from Ricciardi and Bourget (1998) and lipid content of 1.0%, following the recommendations of the European Commission: 5% lipid weight and 26% dry weight content for fish, and 1% lipid weight and 8.3% dry weight content for mussels, for normalizing contaminant concentrations). Yet, OSPAR concentrations are larger than those of more pristine environments, such as Greenland, where reported (mean) DDTs concentrations in mussels are lower than 1.65 log units (ng/g) (UNEP, 2003).

Molluscs include benthic organisms which have contact with the substrate, making them useful as monitors of local pollution. It has been shown, however, that *in situ* biomonitoring shows seasonal patterns and environmentally mediated gaping activity in bivalves (Garcia et al., 2016), though it does not seem to impair their capacity to detect temporal trends related to changes in contaminant levels (Zangrandi et al., 2005). Despite these uncertainties, they have been extensively used, for example, as the pioneers in NOAA's Mussel Watch Program launched in 1985–1986 (Farrington et al., 2016). Other similar programs were later implemented throughout the world (Hamilton, 1989; Ramu and Kajiwara, 2007; Knopf et al., 2020). In more recent years, more emphasis is being put on assessing key biomarkers of biological responses to evaluate the health of organisms, linking observed responses to contaminant exposure, as in the European Marine Strategy Framework Directive (Directive 2008/56/EC, 17 June 2008). At present, the lack of knowledge on assessment criteria of the biomarker responses in certain target species used in integrated monitoring programmes of marine pollution puts limitations to its generalized use (EC, 2014a).

For PCBs in a lipid basis, three statistical populations were found in the data according to the factor with the highest variance (Kruskal-Wallis chi-squared, K-W χ^2 (2) = 179.2, $p < 2 \times 10^{-16}$, Dunn post hoc test, $p < 0.01$—see **Supplementary Material** for detailed statistical results). The statistically different matrices are, except for bird's eggs, the same as those found for DDTs (**Figure 1B**; **Table 2**): 1) liver (LI), with the highest

concentrations; followed 2) muscle (MU), and finally, 3) the whole soft body (SB). The former two were quantified in fish and the latter in molluscs.

The median concentrations of PCBs found in the liver and muscle of fish are similar to those measured for DDTs (**Table 2**), namely 2.90 in the liver, and 2.48 log units (ng/g) in muscle, which compare with 2.95 and 2.47 log units (ng/g), respectively for DDTs. Molluscs, on the other hand, show higher median uptake of PCBs (2.34 log units (ng/g)) than of DDTs (1.70 log units (ng/g)). These results are in agreement with the stabilization of the concentrations of these OCCs in the living tissues we report above. The uptake of similar amounts of the two OCCs by fish have been found elsewhere, namely for *Salvelinus alpinus*, in Bjørnøya Island, Norway (Evenset et al., 2004); for *Xiphias gladius* in the Azores Archipelago (Stefanelli et al., 2004); for *Exocoetus volitans and Sula leucogaster*, in São Pedro and São Paulo Archipelago, Brazil (Dias et al., 2013); for *Larus dominicanus*, in Kerguelen islands, south Indian Ocean (Monod et al., 1992); for *Trematomus newnesi*, *Notothenia coriiceps* and *Notothenia rossii,* in the South Shetland Islands, Antarctica (Lana et al., 2014). DDTs and PCBs metabolites and congeners share similar environmental fate properties, as we discussed in the methods. Considering that the total global mass discharged into the environment may also be similar, then, from a synoptic point of view their environmental concentrations should converge. Of course, this consideration does not apply at a local scale, where particular circumstances gain relevance, as has been demonstrated extensively in the literature (UNEP, 2003). Nor does it apply to molluscs, which due to feeding on phytoplankton or by the filter-feeding of bottom sediments may register a chemical signature in par with that of sediment. The concentrations in the database for PCBs in molluscs are about 0.6 log units above those of DDTs. This may be justified by the fact that in Europe, where DDT has been banned since the eighties, its presence in the sediments is low (though the dataset is still very limited), in the order of 0.05–0.34 ng/g dw (−1.30 to −0.47 log units, ng/g) (Pinto et al., 2016); while the levels of PCBs are about two orders of magnitude higher (10 ng/g dw, or 1.0 log unit) (EC, 2010), reflecting the more continuous input into the environment.

Similar to the lipid basis data, four statistical populations were identified in the data for DDT on a wet basis, $M_{DDT,o,m,wet}$, (Kruskal-Wallis test, K-W χ^2 (11) = 1680.6, $p < 2 \times 10^{-16}$, Dunn post hoc test, $p < 0.01$—see **Supplementary Material** for detailed statistical results). They are: 1) two lipid matrices, fat (FAT) and blubber (BB), with concentrations of DDTs well above the others (**Figure 1C**; **Table 2**); 2) the liver matrix (LI), with intermediate-high concentrations; 3) followed by the muscle and whole-body matrices (MU, WO, and SB); and 4) finally the blood matrices (BC, BL, and BS) with the lowest concentrations. Tail muscle (TM) and fish row (RO) share similar concentrations with the muscle and blood matrices, but the number of samples is still very limited to be statistically relevant. Five statistical populations were identified in the data for PCBs on a wet basis, $M_{PCB,o,m,wet}$ (K-W χ^2 (10) = 1105.9, $p < 2 \times 10^{-16}$, Dunn post hoc test, $p < 0.01$—see **Supplementary Material** for detailed statistical results). They are: 1) two lipid matrices, fat (FAT) and blubber (BB), with concentrations of PCBs well above the others (**Figure 1**; **Table 2**); followed by 2) the liver matrix (LI); 3) the muscle and blood matrices (MU, BL); and 4) finally the soft whole body (SB) with the lowest concentrations. For the remaining matrices, the number of samples is still too limited to be statistically relevant.

Results for the wet-basis for fish liver mirror those of the lipid-basis, so to compare with passive sampling via plastic pellets they seem to be interchangeable (**Table 2**) for both DDTs and PCBs. On the contrary, for fish muscle, the concentrations on a wet basis are about 1.5 log units lower than those obtained for the lipid basis. However, the ratio log DDTs/log PCBs ≈1 is maintained in both the liver and the muscle, again indicating an equal level of exposure/accumulation to both OCCs.

Data on a wet basis for molluscs is undermined by a very high dispersion (cqv = 2.26, which contrasts with cqv <0.35 for the remaining matrices, except for blood). Lipid-normalizing seems to contribute positively to reducing dispersion, which is at odds with the rationale that the whole body biota concentration is linearly correlated with the lipid content of the species (EC, 2014b). The very different number of samples and the period covered by the two sets (212 samples and 25 years against 39 samples and 8 years, for wet and lipid-basis, respectively) may help justify such incongruence.

The fat of birds, unlike their liver, accumulated more PCBs than DDTs by about one order of magnitude (2.97 log units for DDTs and 3.97 log units for PCBs) (**Figure 1D**; **Table 2**). The blubber of mammals, despite being a lipid matrix, seems to accumulate equally in both OCCs (2.97 log units of DDTs and 3.18 log units of PCBs), more in line with the results for the liver. The median concentrations for DDTs are in the range of those found elsewhere (see **Figure 1C**). For example, Lopez et al. (2012) found (mean) concentrations of 2.36 log units (ng/g) in Hawaiian monk seals (*Monachus schauinslandi*) in Hawaiian Islands. In Greenland, Polar bears (*Ursus maritinus*) accumulated 2.51 log units (ng(g). In the same study, the concentration of PCBs (3.70 log units) was, however, higher than that of the most extreme values in the database.

The concentrations of DDTs in blood showed very high variability (cqv = 4.24) and median values of only 0.01 log units (**Figure 1C**; **Table 2**). For PCB the results are better (cqv = 0.22) for a median of 1.56 log units (**Figure 1D**; **Table 2**). The concentrations in the blood are two orders of magnitude lower than in the blubber, results which are coincident with the literature (Gebbink et al., 2008).

The values in the database, as indicated above, reproduce well the published median and range of variability of the studied OCCs in marine organisms, which is a fundamental condition for our subsequent analysis.

3.2 PES as a Proxy for Bioaccumulation

Concentrations of OCCs in marine pellets are statistically similar throughout the world for DDTs, and lower in the Indian Ocean for PCBs (**Table 3**). Therefore, the complete dataset of 263 samples was used in the subsequent analysis for DDTs, while for PCBs data from the Indian Ocean ($N = 39$) were excluded for comparison with data from the OSPAR region, resulting in a total of 344 samples.

TABLE 3 | Concentration of DDTs and PCBs in marine pellets, P_{DDT} and P_{PCB} (Log).

Marine pellets	Location of sampling (median; interquartile range; cqv)	Statistical test (kruskal-wallis), alpha = 0.01*
DDTs	A: Atlantic Ocean, Pacific Ocean, Indic Ocean, North Sea, Mediterranean Sea (0.726 ng/g; 0.917 ng/g; 0.62)	K-W χ^2 4) = 6.35, p= 0.18 N(Atl) = 86; N(Pac) = 114; N (Med) = 19; N(North) = 11; N(Ind) = 33
PCBs	A: Atlantic Ocean, Pacific Ocean, North Sea, Mediterranean Sea (1.66; 1.15 ng/g; 0.35)	K-W (4) = 41.3, $p < 0.01$; followed by Dunn test, significant for Indic OceanN(Atl) = 145; N(Pac) = 138; N(Ind) = 39; N (Med) = 47; N(North) = 14

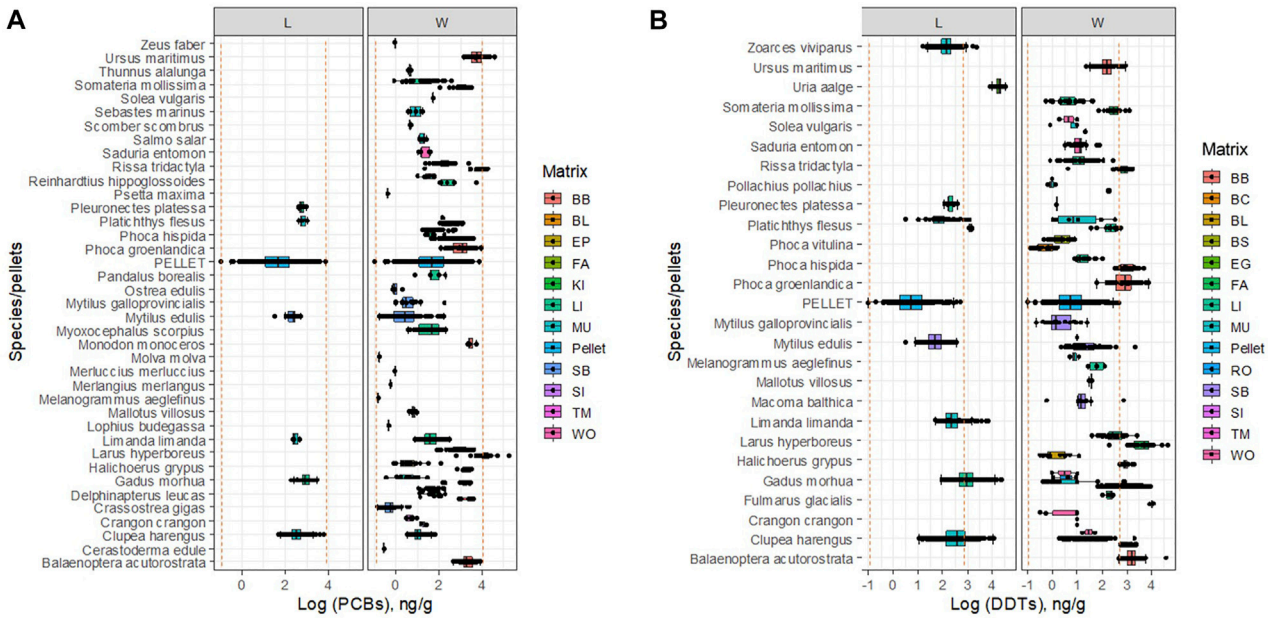

FIGURE 2 | Concentration of PCBs (A) DDTs (B) in marine organisms and marine plastic pellets. L: lipid-normalized; W: wet weight. Vertical lines show a range of concentrations found in.

The concentrations of DDTs in the pellets show high variability, which encompasses in its range most of the concentrations found in non-lipidic matrices in marine organisms when measured as wet weight. On the other hand, concentrations of DDTs in pellets are over two log units lower than those in the lipidic matrices, or in lipid-normalized concentrations in organisms (**Figure 2B**).

The concentrations of PCBs in marine pellets show again high variability, which encompasses in its range most of the concentrations found in both the wet and lipid-basis of non-lipidic matrices in marine organisms (**Figure 2A**).

For computing the R^* ratio (**Eq. 6**), we use bioaccumulation factors for trophic level 4, as available in US EPA EpiSuit, (**Eq. 5**), are Log BAF = 6.365 L/kg for DDT, and Log BAF = 6.888 L/kg for PCB-153 (USEPA, 2012b). The congener 153 was selected here for being one of the seven congeners frequently measured by the International Council for the Exploration of the Sea (Duinker et al., 1988). Partition coefficients, K_{PE}, for many organic substances were compiled by Lohmann (2012) for freshwater. Following the author's recommendation, we applied a compensation factor of plus 50% to correct for the decrease of solubility with the increased ionic strength of ocean water. After correction, for DDT, Log K_{PE} = 5.98 L/kg; and for PCB, Log K_{PE} = 6.98 L/kg. The theoretical Log BAF/Log K_{PE}, R^*, is then equal to 1.06 and 0.99, for DDT and PCB, respectively. Interestingly, these estimates agree with recent research comparing passive sampler uptake to bioaccumulation by organisms of lower trophic levels, under the assumption that the passive sampler polymer and the organism lipid have a similar, proportional affinity for a given hydrophobic organic chemical, or Log M/Log P ≈ 1 (Joyce et al., 2016).

As indicated in the Methods, the concentrations of PCBs in marine pellets may differ from those measured in marine organisms by plus 0.3 log units (ng/g) due to the use of different analytical methods. The correction by this amount would alter the estimates of the ratios by less than +20% (well lower than the interquartile range), i.e., the magnitude of the output uncertainty due to this cause is much smaller than that of the natural variability. Hence, the difference in analytical methods does not impede the use of the estimated ratios.

TABLE 4 | Organism-to-pellet DDTs and *PCBs* ratios, $R_{DDT,o,m,b}$. and $R_{PCB,o,m,b}$.

DDTs			
Basis (b)	**Organism (o)**	**Matrix (m)**	**$R_{s,o,m,b}$ (P25; median; P75; cqv)**
Lipid	Bird	Eggs (EG)	2.75; 4.64; 8.63; 0.52
	Fish	Liver (LI)	1.86; 3.18; 5.98; 0.53
	Fish	Muscle (MU)	1.46; 2.60; 4.92; 0.54
	Mollusc	Soft whole body (SB)	0.99; 1.78; 3.38; 0.55
Wet	Mammal	Blood (BC, BL, BS)	–0.40; 0.05; 0.54; 6.68
	Bird and mammal	Fat and blubber (FA, BB)	1.79; 3.13; 5.91; 0.53
	Bird and mammal	Liver (LI)	0.54; 1.31; 2.77; 0.67
	Fish	Liver (LI)	1.86; 3.15; 5.90; 0.52
	Arthropod, fish, mollusc	Muscle, whole-body (WO, SB)	0.59; 1.23; 2.45; 0.61
PCBs			
Lipid	Fish	Liver (LI)	1.27; 1.66; 2.36; 0.30
	Fish	Muscle (MU)	1.09; 1.44; 2.05; 0.31
	Mollusc	Soft whole body (SB)	0.96; 1.28; 1.83; 0.31
Wet	Mammal	Blubber (BB)	1.35; 1.80; 2,59; 0.31
	Bird	Fat (FA)	1.63; 2.16; 3.10; 0.31
	Arthropod, bird, fish, mammal	Liver (LI)	0.81; 1.18; 1.77; 0.37
	Fish	Muscle (MU) and blood (BL)	0.38; 0.56; 0.85; 0.37
	Mammal	Muscle (MU) and blood (BL)	0.58; 0.89; 1.34; 0.39
	Mollusc	Soft whole body (SB)	–0.13; 0.09; 0.43; 1,92

In order to compute the ratios R (**Eq. 2**), estimates of observed organism-to-pellet posterior statistical distributions, $R_{DDT,o,m,b}$ and $R_{PCB,o,m,b}$ were obtained by sampling 10^5 times from the distributions of Log $M_{DDT,o,m,b}$, Log $M_{PCB,o,m,b}$ and Log P_s (see **Supplementary Material** for details). The percentiles 25, median and 75 were taken from these posterior distributions and the coefficient of quartile variation was computed from the former (**Table 4**).

Median $R_{DDT,o,m,lipid}$, for birds and fish in high lipidic matrices are between one and almost four log units above unity (**Table 4**), which differ substantially from the theoretical ratio $R^* \approx 1$. Uptake in fish muscle and liver deviates in the median about two log units from unity (3.18 and 2.60, respectively). Departures from unity have been reported for organochlorine chemicals in high-trophic level consumers. For instance, no relationship was found between uptake by plastic PE samplers and bioaccumulation in some studies (Ellis et al., 1995; Heltsley et al., 2005), while in others the uptake rates of the two matrices were similar within a factor of 2 (Meadows et al., 1998). The ratio for the soft whole body of molluscs shows again a positive deviation from unity (median = 1.78), but in this case, unity is within the interquartile range of the estimated values (P25 = 0.99; P75 = 3.38), which is concordant with other studies for benthic organisms, including molluscs (Joyce et al., 2016).

Similarly to DDTs, concentrations in marine pellets underestimate uptake of PCBs in liver and muscle of fish, but only by about half log unit ($R_{PCB,o,m,lipid}$ = 1.66 and 1.44, respectively), but the soft whole body of molluscs shows again good agreement with R^* (median = 1.28; P25 = 0.96 and P75 = 1.83) (**Table 4**). These latter results are in contradiction of those obtained by (Ogata et al., 2009), where they found that PCB concentrations in the beached resin pellets from locations around the world underestimated those found in mussels, though their sample size was much smaller.

The ratios for wet basis, $R_{DDT,o,m,wet}$ show that the plastic pellets tend to underestimate uptake of DDTs in some matrices by a factor of two, including the fat of birds and blubber of mammals, and the liver of fish (3.13, and 3.15, respectively. Interestingly, the ratio for the liver of birds and mammals (1.31), and the whole body of arthropods, fish and molluscs (R = 1.23) show good agreement between uptake by organisms and marine pellets, encompassing unity within the interquartile range.

The $R_{PCB,o,m,wet}$ for PCBs in the fat of birds and blubber of mammals are about one log unit above unity, indicating again the underestimation made by marine pellets (1.80, and 2.16, respectively), though smaller than for DDTs. There is a very good match between uptakes for the liver of arthropods, birds, fish and mammals (ratio = 1.18), and the muscle and blood matrices of mammals (0.89). On the other hand, the ratio is lower than unity for muscle and blood matrices of fish (0.56), and the soft whole body of molluscs (0.09).

Several reasons may justify the wide range of R ratios and their deviation from unity. The assumption Log BAF ≈ Log PE is valid when PE and the organism's lipids share similar, and proportional, affinity for a given hydrophobic organic chemical, which may not hold due to several reasons. For instance, bioconcentration for PCBs is expected to increase with an increase in chlorine substitution and a decrease in water solubility (Hawker and Connell, 1988; Jonker and Van Der Heijden, 2007), but due to lower uptake rates, the inverse may happen (Porte and Albaigés, 1994; Bremle et al., 1995; WHO, 2003). The elimination of PCBs from aquatic organisms is both species- and congener-specific: congeners containing two vicinal hydrogen atoms at the meta and para positions in at least one aromatic ring are easily metabolized (Pruell et al., 1993); the PCB congeners 110, 138, 149, 153, and 187 are most recalcitrant in

mussels; congeners 138, 153, 170, and 180 in crabs; 138, 153, 170, 180, and 187 those most recalcitrant in mullet; and 84, 110, 118, and 138 those in tuna (Porte and Albaigés, 1994). So, the PCB congeners with the highest tendency to bioaccumulate are the moderately chlorinated (penta-, hexa-, and hepta-chlorobiphenyls)—which were those synthesized in higher proportions in Aroclor formulations, likely being the most prevalent in the environment. More highly chlorinated OCCs are more tightly bound with soils and sediments, being less bioavailable. Bioaccumulation of OCCs is affected by the water zone in which the organisms reside and feed. For instance, the OCCs levels may be many times higher in the surface strata than in deeper water, and in the sediments, resulting in bioaccumulation levels in fish and bottom-feeding species several times higher in these zones (Södergren et al., 1990). Also, concentrations of persistent organic pollutants in the open ocean are often lower than those observed in coastal areas (Iwata et al., 1993; Schulz-Bull et al., 1998) although they may represent an important part of the global inventory due to a larger oceanic volume (UNECE, 2010).

Another reason for the lower concentrations found in marine pellets may be due to the time of immersion which is insufficient to attain chemical equilibrium and/or for the OCCs to diffuse homogeneously into the plastic pellet. Consider that the partition into the plastic is a first-order equilibrium process described by $P_{s,t} = P_{s,eq}\ (1\text{-}e^{-kt})$, where $P_{s,\ t}$ is the concentration of the substance (ng/g), in the pellet at time t (month), $P_{s,\ eq}$ is the equilibrium concentration (ng/g), k is the sorption rate constant ($month^{-1}$). The latter is equal to 0.1 for high-density polyethene (HDPE) pellets, and 0.2 for low-density polyethene (LDPE) and polypropylene (PP) pellets (Rochman et al., 2013). The sorption rate seems to hold for quite different concentration ranges. For instance, in a study of sorption of 3,3′,4,4′-tetrachlorobiphenyl (PCB77) in PP, in ocean water with concentrations three orders of magnitude above those of Rochman et al., Zhan et al. (2016) found a k = 0.24. The equilibrium concentrations, $P_{s,eq}$, on the other hand, are highly variable according to published studies (as e.g., those above cited, as well as the fact that concentrations of OCCs in the database show very high variability), indicating that for low concentrations only quasi-equilibrium is attained. The process of sorption of non-polar organics in plastic pellets in the environment is still surrounded by much uncertainty. For instance, weathering of plastics increase surface area due to the formation of rougher surfaces and opening of cracks, which will increase sorption (Mato and Isobe, 2001); but the formation of oxygen-containing groups such as ketone or esters, while increasing the affinity for polar organics, decrease that for non-polar (Fotopoulou and Karapanagioti, 2012). The two effects seem to counterbalance each other (Endo et al., 2005). Degradation rates of the surface of pellets in the ocean are very slow for PE and PP, of around 1–3 μm/year (Chamas et al., 2020). The long-term accumulation of organics found in natural conditions may be due to the slow diffusion toward the interior of the material. If that is the case, the concentration would decrease centripetally, eventually becoming zero after some depth from the surface. In that case, the concentrations measured in whole marine pellets would reflect the averaged concentration, including the volume where organics have accumulated as well as the volume of plastic where concentration is zero.

Marine pellets, as well as other synthetic passive samplers, cannot reproduce the processes of food-web biomagnification, which alone can produce up to five log units increase in the lipid-normalized concentration of a bioaccumulative substance (Gobas, 2008). Hence, the ratios here estimated may be useful at screening-level assessments, when detailed data is still missing and high degrees of uncertainty are acceptable (USEPA, 2019).

Beached polyethene marine pellets uptake less OCCs than some organic tissues. The median concentrations of OCCs in the different matrices of fish, birds and mammals are between one to four orders of magnitude higher than those found in marine pellets when lipid-normalized; or up to two orders of magnitude when measured as wet weight. On the other hand, DDTs and PCBs lipid-normalized concentrations and variability in beached marine pellets were similar to those found in molluscs, which agrees with previous studies using passive polyethene samplers (Figueiredo et al., 2017).

Though it is recognized that passive sampling provides information that can be used accurately to predict environmental concentrations (Lohmann and Muir, 2010), some scientific challenges still exist. First, chemical equilibrium between water and the plastic sorbent for high molecular weight apolar organics, such as DDTs and PCBs, can take several months to be attained (Lohmann and Muir, 2010). Second, polyethene samplers have been found to emulate the body burden of benthic biota (Vinturella et al., 2004; Boehm et al., 2005; Fernandez and Gschwend, 2015), but in many cases passive sampling-based concentrations resulted in log BAF – log KEP predictive relationships which were within one to two orders of magnitude of measured bioaccumulation (Joyce et al., 2016). Third, biomagnification at each trophic interaction can lead to food web magnification, often resulting in high concentrations in top predators, which cannot be assessed by a single passive sampling system. Fourth, tissue lipids are often the main factor determining the accumulation of hydrophobic organic contaminants in aquatic organisms (USEPA, 2012c), which again cannot be assessed by a single sampling system. Fifth, pellets are exposed to air when beached, while most of the analysed biota are not. Sixth, the lack of certified reference materials, prohibits the use of commonly agreed values for water-polymer partition and polymer diffusion coefficients (Taylor et al., 2019).

4 CONCLUSION

While acknowledging all the uncertainties mentioned above beached marine plastic pellets may still be considered as part of a synoptic non-biological early warning system, but for organisms other than molluscs, correction of concentrations should be made, namely by using the ratios estimated here.

Future research should focus on reducing the many sources of uncertainty listed in this article, in particular by 1) homogenization of chemical procedures; 2) better assessment of chemical partition

equilibrium between water and polymers in environmental conditions; 3) use of (multi) polymer passive samplers better aimed at mimicking uptake by particular matrices.

DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. This data can be found here: https://data.ices.dk/, http://pelletwatch.org/.

AUTHOR CONTRIBUTIONS

The author confirms being the sole contributor of this work and has approved it for publication.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvs.2021.784317/full#supplementary-material

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