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Occurrence of phthalate esters and preliminary data on microplastics in fish from the Tyrrhenian sea (Italy) and impact on human health

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ABSTRACT

Phthalic acid esters (PAEs) are chemical pollutants widely distributed in the marine environment. They can accumulate in biota, posing a risk to the marine ecosystem and humans. The aim of this study was to measure the content of PAEs in the gills and muscles of three fish species (*Mugil cephalus*, *Diplodus annularis*, and *Mullus barbatus)* caught along the coast of Campania (Italy), as well as to ascertain the dietary exposure to PAEs through the consumption of fish. Secondly, a preliminary insight into microplastics (MPs) pollution in this area was provided through the analysis of *Mugil cephalus* organs. Solid-phase extraction (SPE) and gas chromatography-mass spectrometry (GC-MS) were used for the PAEs analysis, while an Fourier-transform infrared (FTIR) microscope was used to detect MPs after a pre-digestion of the samples. Risk assessment was based on estimated daily intake (EDI) and lifetime cancer risk (LTCR). The results showed higher bioaccumulation of PAEs in *Mullus barbatus* than in the other two species and higher concentration in gills than in muscles. MPs (polyamide, polypropylene, and high-density polyethylene) were detected in half of the gill samples, but no particle was detected in the muscle samples of *Mugil cephalus*. A low carcinogenic and non-carcinogenic risk from the consumption of fish emerged, although a potential risk for the development of cancer was found in the worst-case, especially in toddlers. In conclusion, this study provides insight into PAEs pollution in the Tyrrhenian Sea (Italy), their distribution in fish with different behaviors, and the potential risk to the consumer. Moreover, the data on pollution by MPs in this area could form the basis for future studies.

1. Introduction

Marine pollution is a worldwide burden due to the continuous accumulation of pollutants from anthropogenic sources. Some contaminants can accumulate in the environment over time, becoming a health hazard to living beings. Among these, phthalates or phthalic acid esters (PAEs) represent a critical environmental issue due to their ubiquity. PAEs are characterized by 1,2-benzenedicarboxylic acid and alkyl chains that affect their physicochemical properties, such as water solubility and biodegradation (Kong et al., 2016). They are not chemically bound to the plastic matrix and tend to migrate to the air, water, soil, and food due to particular chemical-physical and biological conditions (temperature, humidity, salinity, solar radiation) (Dhavamani et al., 2022; Cirillo et al., 2013b; Chen et al., 2013; Afshari et al., 2004). PAEs are endocrine disruptors due to their effects on reproduction, metabolism, and growth in humans (Zhang et al., 2021a). Some of them can also have neurotoxic, genotoxic, and carcinogenic effects (Zhang et al., 2021a; Hlisníková et al., 2021; Ventrice et al., 2013; Heudorf et al., 2007). For this reason, PAEs such as di-n-octyl phthalate (DnOP), di-7-methyloctyl phthalate (DiNP), di-2-ethylhexyl phthalate (DEHP), di-butil phthalate (DBP), di-ethyl phthalate (DEP), di-methyl phthalate (DMP) are listed in the Hazardous Substances Data Bank (HSDB). PAEs have been employed in cosmetics, slow-release capsules and dietary supplements, varnishes, medical devices, and pesticides, although their primary use is as plasticizers that add flexibility to plastics (Philippat et al., 2015; Kelley et al., 2012; Heudorf et al., 2007). As a result, the huge amounts of plastic waste in the marine environment, estimated to 12 million tons/year (Boucher et al., 2020), make it likely the primary vector of PAEs in the sea (Ye et al., 2020). For this reason, PAEs can bioaccumulate in marine organisms such as fish due to their

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ingestion/contact directly or indirectly through small fragments or fibers of plastics (0.1 μm–5 mm), namely microplastic (MPs) (Zhang et al., 2021b; Hosseinpour et al., 2021; FAO, 2019). Indeed, Lu et al. (2021) demonstrated the association between the levels of PAEs (i.e., DEP) and MPs occurrence in estuarine fish. This also poses a risk to the human diet, a significant route of exposure to these chemicals (De-la-Torre, G. E., 2020; Shen et al., 2015). According to Wang et al. (2018) and Domenech & Marcos (2021), seafood contributes up to 30.01% of total PAEs and 22.04 × 10^3 p/year of MPs to intake.

Therefore, this study initially investigated the levels of six PAEs in the gills and muscles of stationary and commercial fish species (*Mugil cephalus*, *Diplodus annularis*, *Mullus barbatus*) caught along the Campania coast characterized by intense, productive activity. The aim was to understand the pollution by PAEs and their distribution among species. In addition, the presence of MPs in the gills and muscles of *Mugil cephalus* was studied to obtain the first data on microplastic pollution in this area and its co-occurrence with PAEs. Finally, the potential risk from the ingestion of PAEs by fish consumption was assessed through a deterministic approach.

2. Materials and methods

2.1. Sampling

Three different fish species, such as *Mugil cephalus*, *Diplodus annularis*, and *Mullus barbatus,* were caught by local fishermen. Fifteen adult fish for each species (tot = 45) with median weight are selected for PAEs analysis. Overall, forty-five gills (considering left and right gills as one sample) and forty-five muscles were collected. Instead, 8/15 gills and 8/15 muscle samples belonging to *Mugil cephalus* were used for MPs analysis. Due to the size of the organs, only the samples of *Mugil cephalus* allowed the research for both PAEs and MPs in gills and muscles. For the same reason, only some of *Mugil cephalus* samples were analyzed. The average length was 30–40 cm for *Mugil Cephalus*, 10–20 cm for *Diplodus annularis*, and 10–15 cm for *Mullus barbatus*. Three species were equally collected at each sampling site from July 2020 to February 2021. The sampling sites extended from Domizia Bay (N 41° 8′ 44.081'', E 13° 48′ 8.643'') to the Sarno River mouth (N 40° 41′ 29.594'' E 14° 26′ 31.318'') of the Campania coast (Tyrrhenian Sea). The samples were frozen within a few hours of collection and then dissected to collect gills and muscles.

2.2. Chemical and reagents

DnOP, DiNP, DEHP, DBP, DEP, DMP and deuterated DEHP (DEHP-D_4) standards were purchased from Sigma-Aldrich (Shneldorf, Germany).

Florisil and Bondesil were supplied by VWR International and Agilent Technologies (Palo Alto, CA, USA), respectively.

Solvents for cleaning and other reagents (i.e. n-hexane, dichloromethane, acetone, heptane) were purchased from Merck & Company, Inc. (Kenilwortf NJ, USA).

2.3. Phthalates extraction and purification

Extraction and purification for PAE analysis were performed according to the method described by Tsumura et al. (2001), with minor modifications (Cirillo et al., 2013a). No plastic equipment was used, to avoid external contamination of PAEs and glassware was preliminarily heated in a muffle and washed with acetone and n-hexane before use. For the extraction, 1 g and 0.5 g aliquot of muscle and gills, respectively, previously freeze-dried, was collected in a test tube to which 10 ml of n-hexane-dichloromethane (50:50) were added. The tube was mechanically shaken and placed in an ultrasonic bath for 20 min. Then, the sample was centrifugated at 2500 rpm for 10 min to collect the supernatant in a round-bottom flask (the procedure was performed in duplicate). The supernatant was concentrated using Rotavapor at 45 °C and mixed with 5 ml of n-hexane.

After, the sample was purified on a manually packed chromatographic column. The column was prepared with wadding (preconditioned with acetone and n-hexane and placed in an oven at 100 °C for 1 h), 2.0 g of Florisil, 0.5 g of Bondesil PSA, and 1 g of anhydrous sodium sulfate (Na_2SO_4). Then, the sample was eluted, mixed with 10 ml of acetone and hexane (4:1), and concentrated using Rotavapor. Finally, 2 ml of n-hexane was added to the sample collected in an amber glass vial (2 ml).

2.4. GC-MS analysis

A GC-MS (Agilent 7890A GC system coupled to an Agilent 5975C mass selective detector (MSD) (Agilent Technologies, Santa Clara, CA, USA)) was used for the quantification of PAEs. The parameters were set as follows: initial oven temperature: 60 °C (holding time 0.5 min); injector temperature: 280 °C. After the injection, the ramp rate was programmed from 60 °C to 240 °C at 10 °C/min and until 300 (holding for 18 min) at 15 °C/min. The transfer line of the GC-MS interface was held at 280 °C. Samples (1 μL) were injected in splitless mode into the capillary gas chromatography column. The carrier gas was high-purity helium (99.999%) at a 1.0 ml/min flow rate.

2.5. Sample pretreatment for microplastics analysis

The dry sample was ground, filtered with sequentially decreasing sieves and then digested. 1g of sample was mixed with 20 ml of 30% H_2O_2 and placed in an oven at 30 °C for 24 h. Supplementary H_2O_2 was added when evaporated. Then, NaCl (1.2 g/cm^3) was added, allowing a densiometric separation. The supernatant was collected and mixed with 100 ml Milli-Q water. The digested sample was filtered through a silicon (Si) filter (diameter:10 mm^2, pore size: 5 μm) and placed in a vacuum filtration system (glass funnel with a tube on the bottom of the stem).

2.6. Microplastic identification and quality control/assurance

The identification was carried out through FTIR Nicolet™ iN10 infrared microscope (Thermo Fisher Scientific Madison, WI, USA). A transmittance and reflection mode in each analysis was set to characterize the polymers. The particle size detection was set at 0–5000 μm, whereas the spectral range was set at 4000–675 cm^{-1} with a collection time of 3 s and 16 co-scans. The spectral resolution was 8 $cm^{-1,}$ and the aperture size was from 50 × 50 μm to 150 × 150 μm. The spectra were compared with the library, and matches ≥70% were accepted (Guilhermino et al., 2021; Rasta et al., 2021; Akoueson et al., 2020) based on the analysis performed through Omnic™ Picta™ software.

Potential environmental contamination was avoided following some precautions, according to Prata et al. (2021): use of a laminar flow hood during each step of analysis; filtration of the liquids (water and H_2O_2) before use (0.22 μm filter); use of no plastic equipment and clothes; heating of glass and steel in muffle at 450 °C; covering filtration equipment with Petri dish during the process; application of the blank test. No MPs were detected due to indoor airborne contamination.

2.7. Risk assessment

The potential risk due to PAEs through fish intake was evaluated based on Tolerable Daily Intake (TDI) and Life-Time Cancer Risk (LTCR). At first, the Estimated Daily Intake (EDI, ng/kg_{bw}/day) was calculated (Eq. (1)) and compared with the TDI of each PAEs, if any (Table 1), proposed by EFSA (2019) and WHO (2003).

$$EDI_{PAE} = \frac{IR \times C_{PAE}}{BW} \quad (1)$$

Table 1
Tolerable daily intake (TDI) and slope factor (SF) for each phthalic acid ester (PAEs).

PAE	TDI (μg/kg$_{bw}$/day)	SF (mg/kg$_{bw}$/day)$^{-1}$
DBP	10 (EFSA, 2019)	/
DEHP	50 (EFSA, 2019)	0.014 USEPA (2001)
DiNP	150 (EFSA, 2019)	/
DEP	500 (WHO, 2003)	/
DMP	/	/
DnOP	/	/

- C_{PAE}: Concentrations of each PAE detected in fish muscles (ng/g)
- IR: Intake Rate of fish (g/day) for toddlers (median: 50.00, 95th percentile: 94.63), adolescents (median: 56.50, 95th percentile: 177.73), and adult population (median: 57.50, 95th percentile: 167.2) (EFSA, 2011)
- BW: Body Weight (kg$_{bw}$) for toddlers (11.3), adolescents (52.6), adults (69.7) (Leclercq et al., 2009)

To assess the carcinogenic risk of PAEs such as DEHP, the LTCR (dimensionless) was used. The cancer risk was evaluated based on equation (Eq. (2)) proposed by USEPA

$$LTCR = \frac{EDI \times EF \times TE}{AT} \times SF \quad (2)$$

- EF: Exposure Frequency to the contaminant (350 day/year)
- TE: Total Exposure (70 year)
- AT: Average Lifetime time for non-carcinogenic risk (TE *x* 365 day/year)
- SF: Slope Factor (ng/kg$_{bw}$/day)$^{-1}$ related to each PAE (Table 1).

USEPA considers an LTCR $>1 \times 10^{-4}$ as an unacceptable risk of developing cancer over a human lifetime, whereas values between 1×10^{-6} and 1×10^{-4} are considered an acceptable range for risk (USEPA, 2001). Instead, Health Canada and Alberta Environment and Parks (AEP) propose a threshold of 1×10^{-5} for the risk of developing cancer (Health Canada, 2010).

2.8. Statistical analysis

Data analysis and graph processing were performed using R Software version 3.6.0 and the following package: ggplot2 (R Core Team, 2019; Wickham, 2016). Data were tested for the homogeneity of the variances (Bartlett's test) and normality (Shapiro-Wilk's test). Finally, a one-way analysis of variance with post hoc Tukey's test was performed to ascertain any statistically significant difference.

3. Results and discussion

3.1. Occurrence of phthalic acid esters in different fish species

The concentrations detected in gills and muscles from the three species are listed in Table S1. The levels of PAEs in both organs, except DEHP, showed significantly different means at the 95% confidence level between *Mullus barbatus* and the other two species ($p < 0.05$).

Gills of *Mullus Barbatus* reported the highest median values (median, min-max) for almost all PAEs: DBP (296, 97–680 ng/g), DEP (222, 87–411 ng/g), DiNP (344, 76–6600 ng/g), DMP (625, 181–1273 ng/g), DnOP (198, 80–6523 ng/g).

Differently, the concentrations of contaminants in muscles in *Mullus barbatus* and the other two species showed a low variability among species (Fig. 1).

$\sum$ PAE in gills showed higher levels in *Mullus barbatus* (2009, 1009–15,388 ng/g) than *Diplodus annularis* (1609, 532–5303 ng/g) and *Mugil cephalus* (1040, 591–9388 ng/g).

Likewise, $\sum$ PAE in the muscle of *Mullus barbatus* (914, 497–7259 ng/g) was higher than *Diplodus annularis* (898, 566–1823 ng/g) and *Mugil cephalus* (638, 509–1766 ng/g).

The higher contamination of PAEs in *Mullus barbatus* was likely due to its habitat and their speciation in seawater. Namely, PAEs have higher concentrations near the surface and seafloor due to their release from particular matter and sediments, which resulted in higher contaminant concentrations in *Mullus barbatus* (demersal), than *Diplodus annularis*

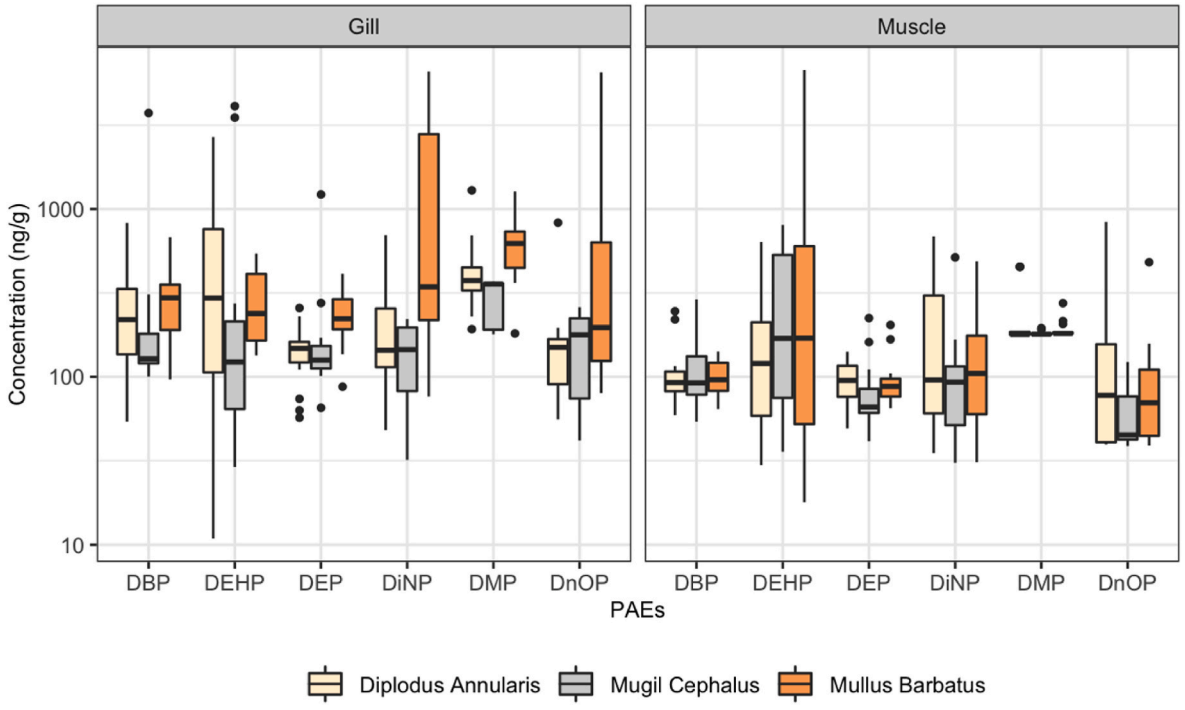

Fig. 1. Concentration (ng/g) on the logarithmic scale of phthalic acid esters (PAEs) detected in gills and muscles belong to three species.

(benthopelagic) and *Mugil cephalus,* (pelagic) (Hidalgo-Serrano et al., 2022; Zhang et al., 2018; Hu et al., 2016). Indeed, these species live on sandy and muddy bottoms, which could lead to greater exposure to these contaminants (Tserpes et al., 2002). In addition, feeding (carnivorous in *Mullus barbatus* and omnivorous-detrital in *Mugil cephalus* and *Diplodus annularis*), as well as tissue fat (higher in *Mullus barbatus*) could influence PAEs accumulation (Grigorakis, 2017; Hu et al., 2016; Pastor et al., 1996).

As regards the different bioaccumulation in the organs, a significantly higher concentration of PAEs was found in the gills than in the muscle ($p < 0.05$). Gills are in close contact with the sea and dissolved contaminants, since they are involved in water filtration, gas exchange, and osmoregulation. On the other hand, the muscle was exposed to pollutants after absorption, detoxification, and distribution processes. These anatomic and physiologic differences could explain the expected trend of bioaccumulation and distribution of PAEs, as also reported by Hu et al. (2016).

3.2. Occurrence of microplastic in fish

A preliminary background about the potential contamination by MPs in addition to PAEs was provided through the analysis of the gills and muscles of *Mugil cephalus.* Some studies reported an increase in (eco) toxicological risk due to the co-occurrence of PAEs and MPs than the single pollutant, although there is still a lack of literature (Liu et al., 2020; Deng et al., 2020).

The analysis revealed the presence of MPs as well as inorganics and plastic additives (e.g., bis(2-hydroxyethyl)dimerate). MPs were detected in 4/8 gill samples ranging from 453 to 3885 μm, while no MPs were detected in muscle samples. Three different polymers were found: fibers of polyamide (nylon), fiber of polypropylene (PP), and fragments of high-density polyethylene (HDPE) (Table 2).

The occurrence of nylon and PP fibers are likely linked to the use and loss of ropes, nets, and fishing gear, as well as the release of textiles from sewage (Rasta et al., 2021; Akhbarizadeh et al., 2018; Welden & Cowie, 2017). Instead, HDPE could be linked to housewares, pipes, packaging, industrial wrappings and film scattered in the sea (Rahim et al., 2013; Achilias et al., 2007).

These preliminary data also provide the potential inability of MPs to migrate in the muscles of *Mugil cephalus,* although further study needs to confirm this trend. Likewise, Su et al. (2019) and Huang et al. (2020) did not detect MPs in the muscles of fish from the East and the South China Sea, respectively. Instead, other studies reported contrary results (Guilhermino et al., 2021; Rasta et al., 2021; Barboza et al., 2020; Akhbarizadeh et al., 2018). Since no standardized methods are available for MPs analysis, there could be inconsistencies in the results, with possible under- or over-estimation of the particles amount (Prata et al., 2021). Several studies have detected MPs in other internal organs, such as fish liver (Avio et al., 2015; Guilhermino et al., 2021) or brain (Atamanalp et al., 2021), whereas toxicological studies highlighted the bioavailability of MPs in the particle uptake pathway according to *in vitro*, murine models (Stock et al., 2019), and some evidence in goldfish liver (Abarghouei et al., 2021): this process seems to occur in a size-dependent manner (<10 μm). Instead, EFSA reported that MPs <150 μm could cross the intestine of mammals, but few data are available on their behavior after absorption (EFSA, 2016). However, it is reported that deep penetration of microparticles into organs is limited to smaller particles (<1.5 μm) due to filtration through the spleen and liver via bile (EFSA, 2016; Yoo et al., 2011). Hence, the absence of MPs in muscles (in the considered size range) could be attributed to the organism's efficient barrier and filtration system. However, for the reasons mentioned above and the limitation of the analytical instrument (< about 10 μm), lower size particles may have reached the organ without detecting them. Overall, MPs in fish should be of concern beyond its presence in muscle because several culinary preparations involve cooking whole fish, potentially releasing contaminants in the cooking broths, and consuming parts other than muscle. Furthermore, as previously mentioned, MPs could absorb and release PAEs as well as other pollutants such as heavy metals, persistent organic pollutants (POPs), and pathogens that may distribute throughout the body (Arienzo et al., 2021; Bowley et al., 2021; Yin et al., 2021; Verla et al., 2019).

Table 2
Type and amount of microplastics (MPs) detected in gills and muscles of fish samples.

Sample	Gill		Muscle	
	Polymer	Amount	Polymer	Amount
1	Polyamide (Nylon)	1	ND	
2	Polypropylene (PP)	1	ND	
3	High-density polyethylene (HDPE)	1	ND	
4	ND		ND	
5	Polyamide (Nylon)	1	ND	
6	ND		ND	
7	ND		ND	
8	ND		ND	

3.3. Phthalates risk assessment

The final part of the study aimed to assess the potential risks to consumers from exposure to PAEs as a risk assessment model for exposure to MPs in humans is yet to be developed. Two different scenarios were evaluated: best- and worst-case. In the former, the median values (min-max) of EDI referred to toddlers' exposure were 102.14 (57.44–322.48) ng/day/kg_{bw} for DBP; 176.37 (18.20–8119.85) ng/day/kg_{bw} for DEHP; 93.36 (44.04–243.90) ng/day/kg_{bw} for DEP, and 107.18 (31.42–886.91) ng/day/kg_{bw} for DiNP. Lower values were reported for adolescents and adults. In adolescents, the values were 25.23 (14.19–79.67) ng/day/kg_{bw} for DBP; 43.57 (4.49–2006.03) ng/day/kg_{bw} for DEHP; 23.06 (10.88–60.26) ng/day/kg_{bw} for DEP; and 26.48 (7.76–219.11) ng/day/kg_{bw} for DiNP. In adults, the values were 19.04 (10.71–60.12) ng/day/kg_{bw} for DBP, 32.88 (3.39–1513.88) ng/day/kg_{bw} for DEHP; 17.40 (8.21–45.47) ng/day/kg_{bw} for DEP, and 19.98 (5.85–165.35) ng/day/kg_{bw} for DiNP (Fig. 2). Instead, toddlers reported worst-case values of 193.34 (108.71–610.33) ng/day/kg_{bw} for DBP, 333.78 (34.43–1649.24) ng/day/kg_{bw} for DEHP, 176.68 (83.34–461.60) ng/day/kg_{bw} for DEP, 202.85 (59.46–1678.56). In adolescents, the values were 77.99 (43.86–246.25) ng/day/kg_{bw} for DBP; 134.67 (13.89–6200.57) ng/day/kg_{bw} for DEHP; 71.28 (33.62–186.24) ng/day/kg_{bw} for DEP; and 81.84 (23.99–677.27) ng/day/kg_{bw} for DiNP. In adults, the values were 55.37 (31.14–174.83) ng/day/kg_{bw} for DBP, 95.61 (9.86–4402.10) ng/day/kg_{bw} for DEHP; 50.61 (23.87–132.22) ng/day/kg_{bw} for DEP, and 50.10 (17.03–480.83) ng/day/kg_{bw} for DiNP (Fig. 3). The assessment of cumulative risk based on Group Phthalates concentration as DEHP equivalent (GPDEq) (EFSA, 2019) considering DEHP (x1), DBP (x5), and DiNP (x0.3) reported in the best case values equal to 719.21 (314.82–9998.33) ng/day/kg_{bw} in toddlers; 177.68 (77.77–2470.12 ng/day/kg_{bw} in adolescents, and 134.09 (58.69–1864.10) ng/day/kg_{bw} in adults, whereas in the worst case were 1361.18 (595.83–18,922.85) ng/day/kg_{bw} in toddlers; 549.21 (240.40–7635.03 ng/day/kg_{bw} in adolescents, and 389.91 (170.67–5420.50) ng/day/kg_{bw} in adults.

Carcinogenic risk assessment based on LTCR estimation showing in the best-case values of 2.36e-06 (2.44e-07 – 1.09e-04) for toddlers, whereas adolescents and adults reported lower results: 5.74e-07 (range: 5.93e-06 – 2.64e-05) and 4.41e-07 (range: 4.55e-08 – 2.03e-05), respectively (Fig. 4). Instead, in the worst-case, toddlers, adolescents, and adults reported values of 4.48e-06 (4.62e-07 – 2.06e-04), 1.80e-06 (1.86e-07 – 8.32e-05), and 1.28e-06 (1.32e-07 – 5.90e-04), respectively (Fig. 5).

Overall, the median EDI values did not exceed the TDI proposed for the four PAEs, suggesting a non-carcinogenic risk. The cumulative risk as DEHP equivalent also showed a value below the TDI. The

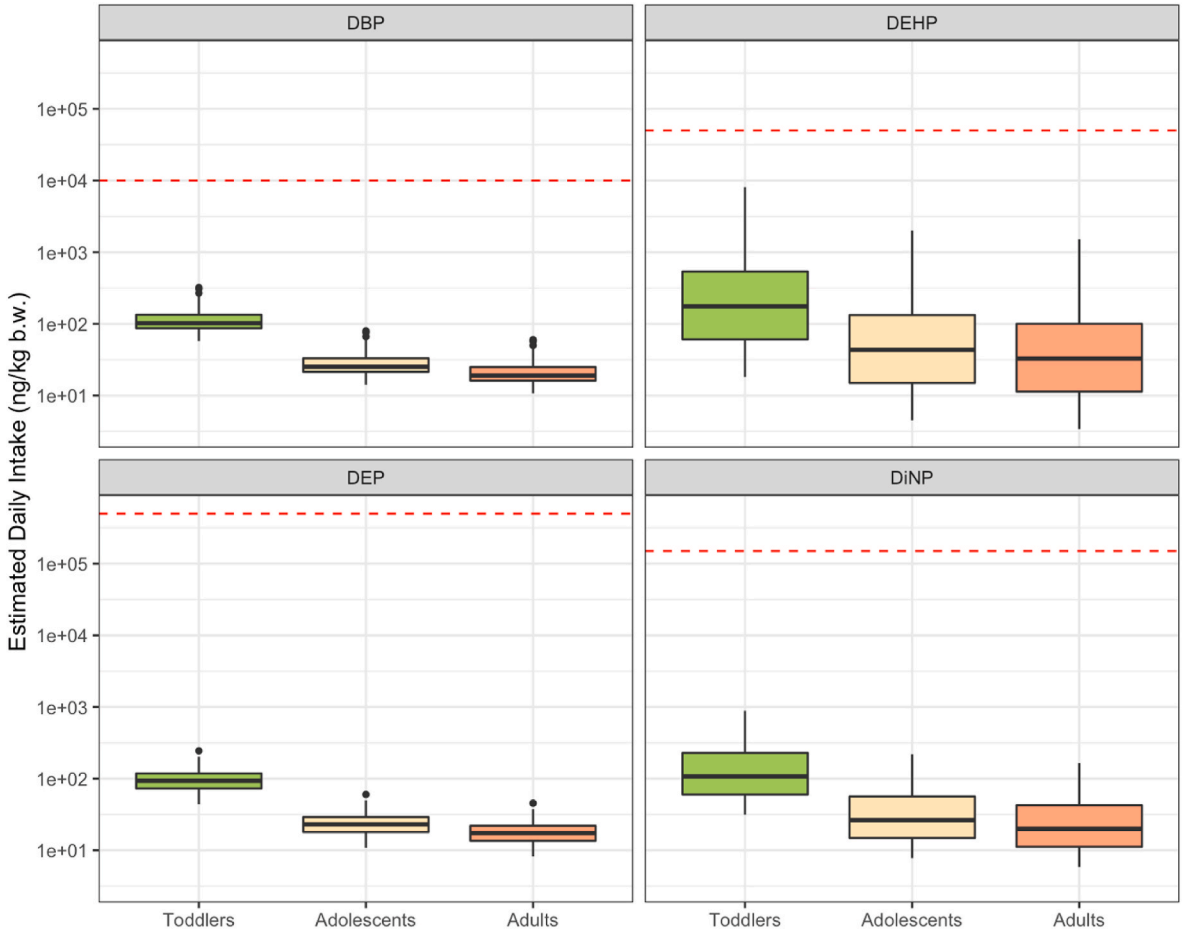

Fig. 2. Estimated Daily Intake (EDI) (best case) on a logarithmic scale of four PAEs detected in fish muscles compared to respective Tolerable Daily Intake (TDI) (red line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

carcinogenic risk was estimated based on DEHP, considering its ability to induce hepatocellular carcinomas and adenomas. Generally, the risk of developing cancer did not occur in any of the three groups, although thresholds (1e-04 or 1e-05) were exceeded for the levels above the third quartile in the worst case. The estimates are based on a conservative approach that assumes 100% of the bio-accessibility and bio-availability of PAEs. However, Koch et al. (2005) demonstrated in a human study that most orally administered DEHP (>70%) was absorbed. On the other hand, fish is only one of the several sources of exposure to PAEs that could significantly increase the risk. Other commonly consumed foods such as cereals, vegetables, meat, dairy products, and beverages may contribute strongly to oral exposure (Wang et al., 2018).

4. Conclusions

Using fish as potential bioindicators, data on PAEs marine pollution in Campania (Italy) and bioaccumulation of these pollutants in the organs of three different species were obtained. The results show that fish behavior and characteristics probably influence the bioaccumulation of PAEs, which is higher in *Mullus barbatus* than in the other two species (*Diplodus annularis* and *Mugil cephalus)*. The gills were the organ with the highest contaminant load due to their anatomical, functional, and physiological characteristics. In addition, three different plastic polymers (nylon, PP, HDPE) were detected in fish gills, whereas no MPs were found in muscles, indicating that they are unlikely to accumulate in these tissues. Based on muscle contamination, the carcinogenic risk from exposure to PAEs by fish consumption was generally low in all age groups, although a potential carcinogenic risk was found in the worst case, especially in toddlers.

In conclusion, safer food choices could be made based on fish characteristics and behavior. The occurrence of PAEs in the Campania marine environment could be of concern and increase the dietary intake of xenobiotics related to fish consumption. Further investigations are needed to provide more in-depth data on MPs pollution and their exposure, as well as the potential correlation with PAEs. In addition, a probabilistic risk assessment for MPs needs to be developed to better characterize the risk of ingestion of these emerging pollutants related to fish consumption.

Credit author statement

Jonathan Squillante: Writing- Original draft preparation, Writing - Review & Editing, Data curation; **Marcello Scivicco:** Methodology, Investigation, Writing- Original draft preparation; **Andrea Ariano:** Writing- Original draft preparation, Writing - Review & Editing, Data curation, Investigation; **Agata Nolasco**: Writing-Original draft preparation, Writing - Review & Editing, Data curation; **Francesco Esposito:** Writing- Original draft preparation, Validation, Data curation, Writing - Review & Editing, Formal analysis, Supervision; **Nunzio Antonio Cacciola:** Writing - Review & Editing, Data curation, Validation; **Lorella Severino**: Conceptualization, Writing - Review & Editing, Resources, Supervision, Project administration; **Teresa Cirillo**: Writing - Review & Editing, Supervision, Resources.

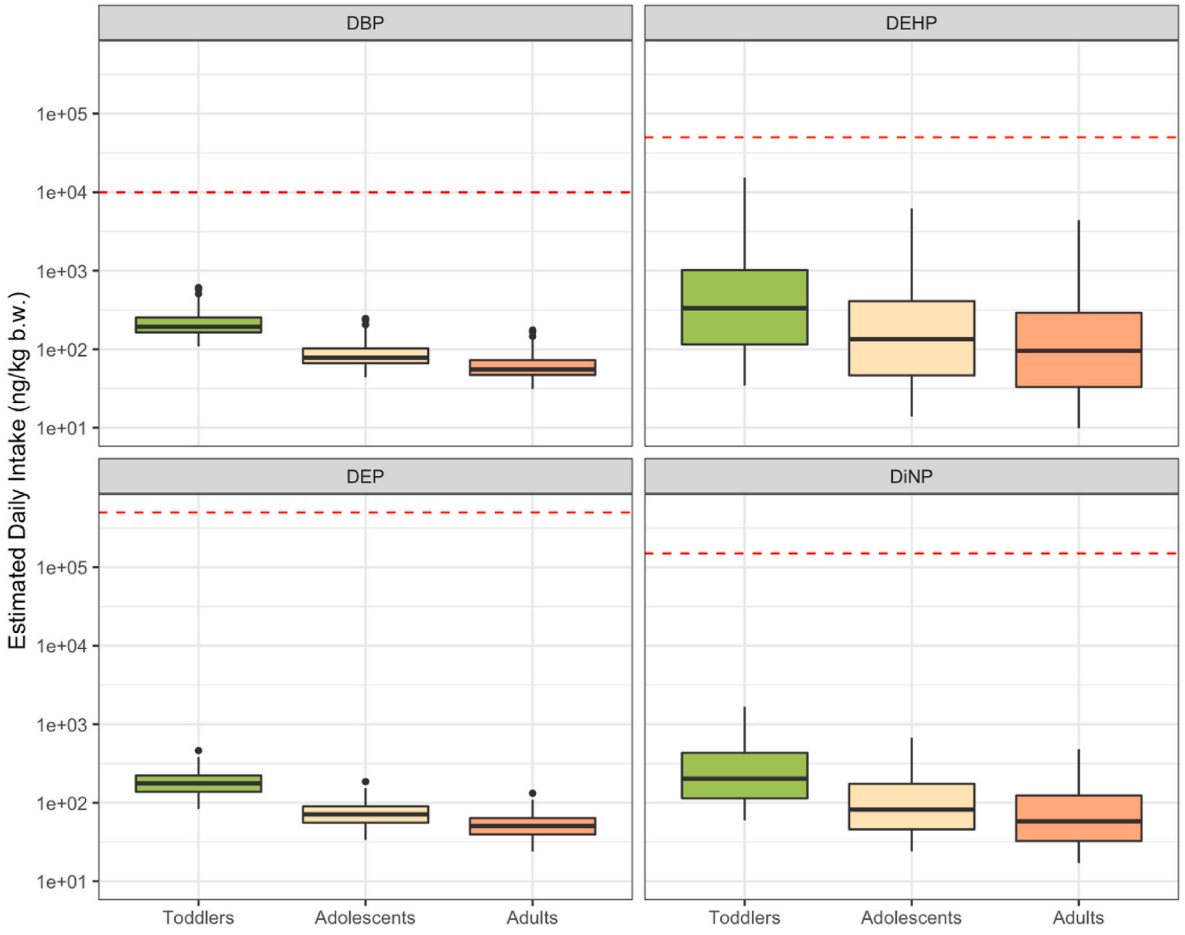

Fig. 3. Estimated Daily Intake (EDI) (worst case) on a logarithmic scale of four PAEs detected in fish muscles compared to respective Tolerable Daily Intake (TDI) (red line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

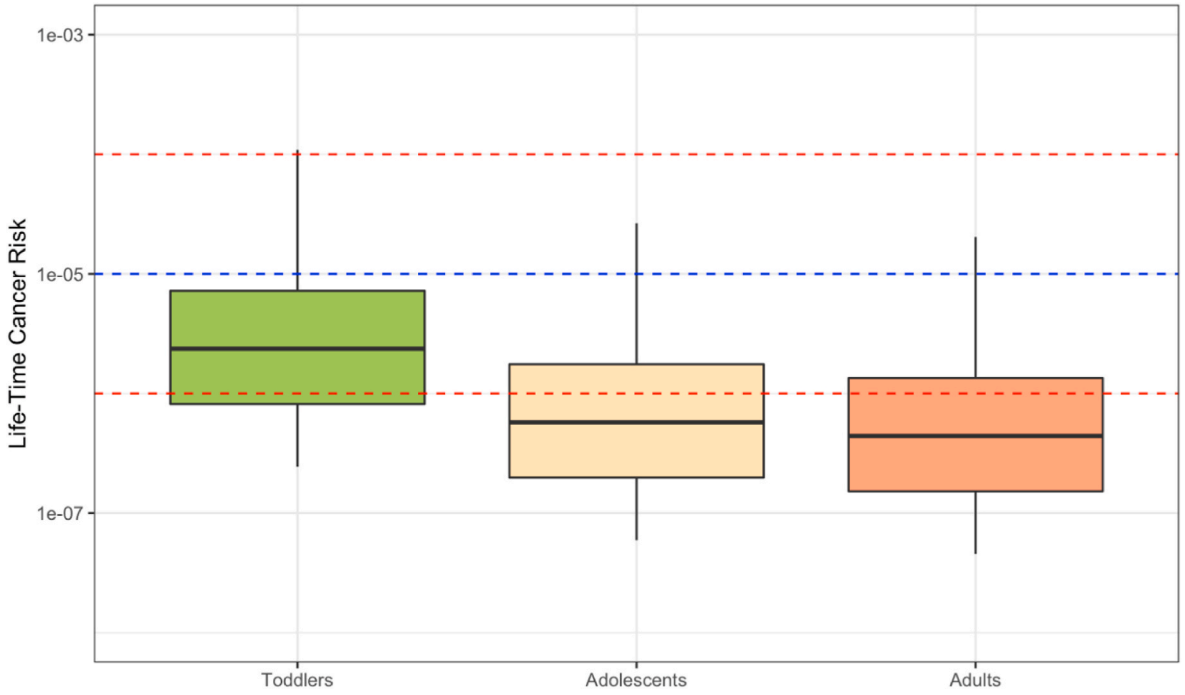

Fig. 4. Life-Time Cancer Risk (LTCR) on a logarithmic scale due to exposure (best case) to PAEs detected in fish muscles compared to threshold values for carcinogenic risk (red and blue line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

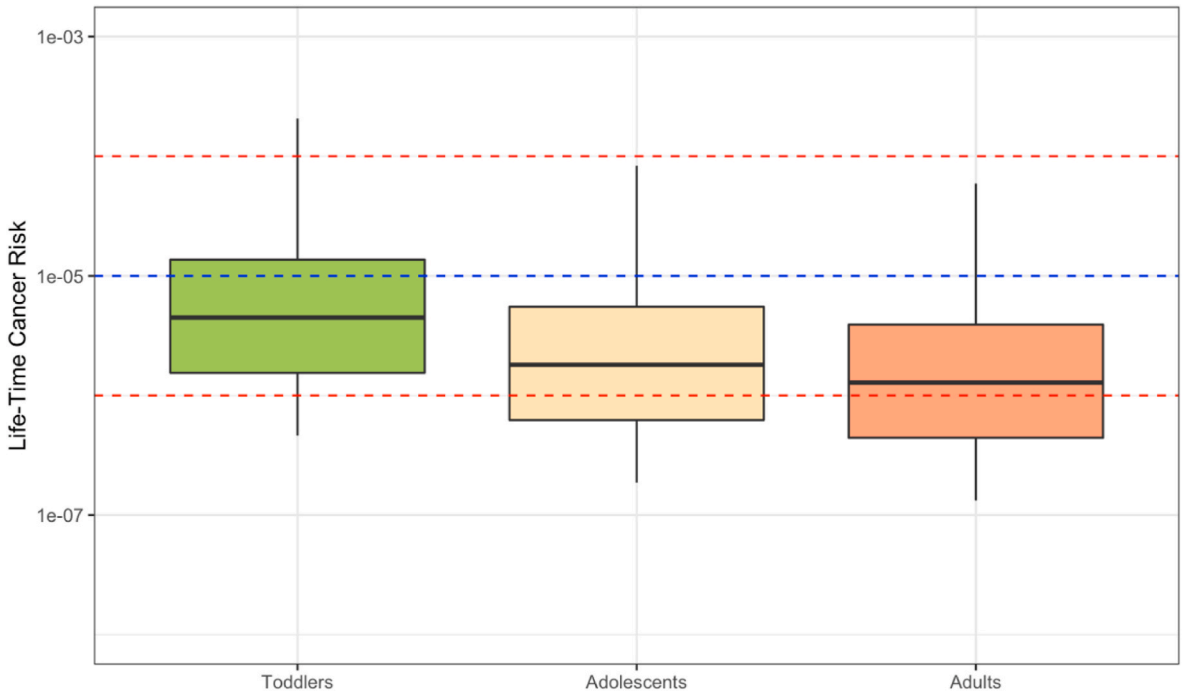

Fig. 5. Life-Time Cancer Risk (LTCR) on a logarithmic scale due to exposure (worst case) to PAEs detected in fish muscles compared to threshold values for carcinogenic risk (red and blue line).

Declaration of competing interest

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

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.120664.

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