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Baseline

Active and passive biomonitoring of trace elements, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls in small Mediterranean harbours

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ABSTRACT

Pollution particularly affects coastal ecosystems due to their proximity to anthropic sources. Among those environments, harbours are subjected to marine traffic but also to accidental and chronic pollution. These areas are thus exposed to complex mixtures of contaminants such as trace elements and organic contaminants which can impact marine species, habitats, and ecosystem services. The monitoring of these compounds is thus a crucial issue for assessment of environmental health. In this context, the aim of the present work was to evaluate the chemical contamination of harbours in Corsica (NW Mediterranean) by measuring the bioaccumulation of trace elements, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls in mussels, limpets, and sea cucumbers. The human health risks associated with seafood consumption were also assessed. Results reveal a relatively low contamination in the Corsican harbours studied compared to larger Mediterranean ports and suggest that the potential health risk for consumers eating seafood is low.

Harbours areas are receiving inputs of contaminants from the coast and are particularly subjected to contaminant accumulation due to reduced water exchange and low tidal currents (Merhaby et al. 2019), therefore representing hotspots for pollution (Lichtfouse et al. 2012; Paladino et al. 2017) and potential sources to the open ocean. These coastal ecosystems in many regions are exposed to complex mixtures of contaminants such as trace elements (TEs), polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs). These compounds can be harmful for marine organisms and persist in the environment, thereby posing environmental and health risks (Wilhelmsson et al. 2013; Castro-Jiménez et al. 2021; Sun et al. 2022; Zaidi et al. 2022).

The monitoring of these contaminants in coastal areas and their impacts on human health and ecosystems is a crucial issue. Native mussels *Mytilus galloprovincialis* have been extensively used for evaluating contamination from TEs (Azizi et al. 2020; Conti and Cecchetti

2003; Esposito et al. 2021; Guendouzi et al. 2018; Santos-Echeandía et al. 2021), PAHs, and PCBs (Benali et al. 2017; Campillo et al. 2019; Fernández et al. 2012; León et al. 2013). Nevertheless, the patchy distribution or absence of this sentinel species in some Mediterranean coastal areas is problematic (Andral et al. 2004). An alternative approach consists in using transplanted mussels which are deployed in cages in situ for coastal pollution studies (Andral et al. 2011; Benedicto et al. 2011; Bodin et al. 2004; Glad et al. 2017; Kucuksezgin et al. 2020; Richir and Gobert 2014). This active biomonitoring also provides improved control of confounding factors and enables to select mussels with homogenous size, age, and physiological state, in contrast to passive biomonitoring which relies on native individuals (Besse et al. 2012; Beyer et al. 2017). A complementary approach is the use of other bioindicator species. Limpets Patella spp. are gastropods widely distributed in Mediterranean rocky coastlines (Poppe and Goto 1991; Storelli and Marcotrigiano 2005) and have been used for monitoring trace metals

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Abbreviations: TE, Trace element; PAH, Polycyclic aromatic hydrocarbon; PCB, Polychlorinated biphenyl; TEPI, Trace element pollution index; EAC, Environmental assessment criteria.

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(Cabral-Oliveira et al. 2015; Conti et al. 2015; Reguera et al. 2018; Lozano-Bilbao et al. 2021; Sánchez-Marín et al. 2022; Zaidi et al. 2022) and organic pollutants (Bartolomé et al. 2011; Delgado et al. 1999; Gianguzza and Orecchio 2006; Peña-Méndez et al. 1996, 2001; Pérez et al. 2019; Viñas et al. 2018). Sea cucumbers *Holothuria* spp. have been identified as potential bioindicators of trace metals (Culha et al. 2016; Hosseini et al. 2022; Marrugo-Negrete et al. 2021; Mohammadizadeh et al. 2016; Parra-Luna et al. 2020; Warnau et al. 2006), however, knowledge on organic contaminant bioaccumulation in these echinoderms is scarce.

In the Mediterranean Sea, Corsica island is often considered to be lightly impacted by contamination pressure due to its low density of population and the scarcity of industrial activities (Andral et al. 2004; Lafabrie et al. 2008). Nevertheless, high concentrations of heavy metals, PAHs, and PCBs have been reported in sediments of the main harbours (i.e., Ajaccio, Bastia, and Bonifacio) (Galgani et al. 2006; Mauffret et al. 2018). To our knowledge, chemical contamination in smaller Corsican harbours remains poorly studied.

The present study investigated chemical contamination in four harbours in the North coast of Corsica by assessing levels of TEs, PAHs, and PCBs in different benthic species. The biomonitoring was conducted using native limpets *Patella* spp., sea cucumbers *H. tubulosa*, and caged mussels *M. galloprovincialis*. Potential human health risks associated with consumption of these species were also evaluated. Although these edible species are not expected to be harvested from harbours, health risk assessment can provide additional information on contamination level for compounds with no environmental thresholds. Moreover, illegal fishing of sea cucumbers has been reported in Corsica and correlates with a strong international market demand, sea cucumbers being mostly exported for oriental consumers (Sadoul et al. 2022). In 2019, a prefectural decree has prohibited sea cucumber harvesting in Corsican territorial waters for 5 years. Assessment of contaminant levels in *H. tubulosa* is thus needed for a food safety perspective before considering potential exploitation of sea cucumbers stocks in Corsica. This paper improves knowledge on chemical contamination in Mediterranean coastal areas which is required to assess their environmental status.

In the Western Mediterranean Sea basin, four French harbours (STARESO, Calvi, Ile Rousse, and Saint-Florent) of the Corsica Island were studied (Fig. 1). The private harbour of the Underwater and Oceanographic Research Station (STARESO; 42°34'49.454"N, 8°43'27.746"E) is located on the Revellata peninsula which is included in the Natura 2000 network. We selected the small STARESO harbour (4



Fig. 1. Sampling locations along the north-western coast of Corsica (France). The black dots represent the studied harbours: STARESO, Calvi, Ile Rousse, and Saint-Florent.

berths) as a reference site characterized by low anthropogenic pressures (Güreşen et al. 2020). The Calvi port ($42^{\circ}33'51.07''N$, $8^{\circ}45'27.939''E$), in the bay of Calvi, is the main gateway for vessels from the French Mediterranean coastline and has a capacity of 500 berths. The port of Ile Rousse ($42^{\circ}38'23.406''N$, $8^{\circ}56'7.778''E$) has a total capacity of 250 berths and hosts a ferry terminal. The port of Saint-Florent ($42^{\circ}40'50.362''N$, $9^{\circ}17'54.373''E$) receives freshwater inputs from Aliso river and is located within the perimeter of the Natura 2000 network and the Marine Nature Park of Cap Corse and Agriate. Among the study sites, Saint-Florent is the port that has the largest reception capacity with 950 berths.

Field work was conducted in January and September 2020. In January, native sea cucumbers (*Holothuria tubulosa*, n = 5–7 per site, length = 154 ± 44 mm) were collected in the four harbours, native limpets (*Patella* sp., n = 6–7 per site, shell length = 34 ± 5 mm) were sampled at all sites except Saint-Florent while native mussels (*Mytilus galloprovincialis*, n = 7, shell length = 78 ± 9 mm) were found only in Saint-Florent harbour. These animals were analysed individually for measurement of contaminant contents. Additional organisms were collected and treated as pool (1 pool for each site and species) for contaminant analysis: 6 mussels (length = 63 ± 9 mm) and 4 sea cucumbers (length = 193 ± 35 mm) were collected at Saint-Florent, and 7 limpets (length = 31 ± 3 mm) were sampled in Ile Rousse.

In June, mussels (M. galloprovincialis, length 57 \pm 4 mm) were obtained from a local mussel farm outside harbour areas, kept on running sea water in STARESO's facilities for 2 weeks, then placed into polypropylene netting bags and transplanted to the 4 sites. The cages were deployed for a thirteen-week period, from mid-June to mid-September, a period assumed sufficiently long to ensure equilibration with environmental conditions (Beyer et al. 2017). In September, limpets (Patella sp., n = 7-8 per site, length = 35 ± 7 mm), sea cucumbers (*H. tubulosa*, n = 4–7 per site, length = 173 \pm 53 mm), and caged mussels (n = 8 per site, length = 64 ± 7 mm) were collected in the sites, except in Calvi where only 3 mussels survived. Animals sampled in September were pooled for contaminant analyses (1 pool for each site and species). After sampling, body walls of sea cucumbers and whole soft tissues of mussels and limpets were dissected, weighted, homogenized in a porcelain mortar and pestle, and stored at -20 °C. Contaminant analyses were conducted on the body wall of sea cucumbers since it is the edible part of these animals (Xing et al. 2021) unlike mussels and limpets which are usually consumed whole.

The content (μ g g⁻¹ dry weight) of 18 TEs (Ag, Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, Sn, V, Zn) in tissues were performed with a Varian Vista-Pro ICP-OES and a Thermofisher Scientific XSeries 2 ICP-MS according to Breitwieser et al. (2017). Details on the methodology can be found in Appendix S1 and Table S2. To investigate overall TE contamination levels among the sites, the trace element pollution index (TEPI) was determined for each species sampled in September 2020. TEPI is the weighted product of mean normalized TE concentrations Cf of the *n* TE analysed (TEPI = $(Cf_1 \times Cf_2 \times ... \times Cf_n)^{1/n}$) (Richir and Gobert 2014). Mean normalization is useful to account for data with varying magnitude such as concentrations of various TE (Moreda-Piñeiro et al. 2001). A higher TEPI value for a given site indicates a higher overall TE contamination.

22 PAHs (naphthalene (N), benzothiophene (BT), biphenyl (B), acenaphthylene (ANY), acenaphthene (ANA), fluorene (F), dibenzothiophene (DBT), phenanthrene (P), anthracene (A), fluoranthene (FA), 2-methylfluoranthene (mFA), pyrene (PY), benzo(*a*)anthracene (BaA), chrysene (CR), benzo(*b*)fluoranthene (BbF), benzo(*k*)fluoranthene (BkF), benzo(*e*)pyrene (BeP), benzo(*a*)pyrene (BaP), perylene (PE), indeno(1,2,3-*cd*)pyrene (IN), dibenzo(*a*,*h*)anthracene (DBA), and benzo (*g*,*h*,*i*)perylene (BPE)) and 14 PCBs (PCB 7, 28, 35, 52, 77, 101, 105, 118, 135, 138, 153, 156, 169, and 180) were analysed (ng analytes g⁻¹ wet weight (ww)) in tissues by stir bar sorptive extraction-thermal desorption-gas chromatography-tandem mass spectrometry (SBSE-GC–MS/ MS) as described by Lacroix et al. (2014). Details on the methodology can be found in Appendix S1 and Tables S3 and S4.

Health risks were estimated for chronic exposure; thus, the dietary exposure was calculated by using mean annual contaminant concentrations (mean of January and September 2020 concentrations) for each site and species (i.e., limpet and sea cucumber). Heath risks related to mussel consumption was estimated separately for native and caged mussels. A medium bound approach was used for estimating contaminant concentrations: results below the limit of detection (LD) were replaced by the numerical values of LD/2 and those below the limit of quantification (LQ) were reported as LQ/2 (Conte et al. 2016; Kiani et al. 2021; Pastorino et al. 2021; Sirot et al. 2012; Veyrand et al. 2013). Ingestion rate (IR) used to estimate dietary exposure in the present study is the mean consumption of molluscs and crustaceans of French adult population (21.47 g day⁻¹), estimated from the INCA3 data (ANSES et al. 2017). We considered a mean human body weight (BW) of 70 kg.

To evaluate potential harmful exposure to TEs or PAHs from seafood consumption, the hazard quotients (HQ) were calculated to assess non-carcinogenic risks for each compound using the following formula (Marengo et al. 2018; Pastorino et al. 2021; Traina et al. 2019):

$$HQ_i = (C_i \times IR \times 10^{-3}) / (BW \times RfD_i)$$

where C_i is the mean annual concentration ($\mu g g^{-1}$ ww) of a compound *i* in the seafood (mussel, limpet, or sea cucumber), 10^{-3} is the unit conversion factor and *RfDi* is the chronic oral reference dose (mg kg⁻¹ day⁻¹) of the compound *i*. The HQ were calculated only for compounds with available *RfD* value (Tables S2, S3). Hazard quotient indicates the ratio between exposure and the reference dose; when HQ is above 1, systemic effects may occur.

The cancer risk due to exposure to PAHs via consumption of seafood was evaluated by estimating the margin of exposure (MOE) for \sum PAH4 (sum of BaA, CR, BbF, and BaP concentrations). The \sum PAH4 was assessed in this study based on the review by the Contaminants in the Food Chain (CONTAM) Panel, relating to occurrence and toxicity of PAHs in food, which concluded that \sum PAH4 is a more suitable indicator of PAHs in food than BaP concentration (EFSA 2008). The MOE was evaluated as an acceptable method of cancer risk assessment (EFSA 2008; Veyrand et al. 2013), as followed:

$$\text{MOE} = (\text{BMDL}_{10} \times \text{BW}) \Big/ \Big(\sum \text{PAH4} \times \text{IR} \Big)$$

where BMDL₁₀ is the benchmark dose lower limit of 3.4×10^5 ng kg⁻¹ day⁻¹ for PAH4 (EFSA 2008), \sum PAH4 (ng g⁻¹ ww) is the mean sum of the concentrations of BaA, C, BbF, and BaP in seafood (mussels, sea cucumbers or limpets). An MOE of 10^4 or higher is considered of low concern from a public health point of view with respect to the carcinogenic effect (EFSA 2008).

Analysis of the 6 indicator PCBs (i.e., iPCBs: PCB 28, 52, 101, 138, 153, and 180) were used for predicting the total PCB content, since the sum of iPCBs represents approximately 50 % of all PCB congeners in food of animal origin (AFSSA 2007). Their selection is also based on their dominant presence in technical mixtures, environment, and animal tissues (EFSA 2010). The average daily dose (ADD; ng kg⁻¹ day⁻¹) represents dietary intake of iPCBs through seafood consumption and is calculated as follows:

$$ADD_{iPCB} = \frac{\sum iPCB \times IR}{BW}$$

where \sum iPCB is the mean annual sum of the iPCB concentrations in seafood. The ADD_{iPCB} values were then compared to the "guidance value" of 10 ng kg⁻¹ day⁻¹ (Arnich et al. 2009; Baars et al. 2001).

The mean TE concentrations measured in limpets, sea cucumbers, and mussels are presented in Table 1 and were compared with levels reported in other Mediterranean geographical areas (Table S5).

Mean Cu concentrations in mussels from Ile Rousse, Saint-Florent, and Calvi (Table 1) were higher than the background levels in North-

ousse, and Saint-l	Florent) in Janua as used for estim	ry and Sel	otember 20: aminant co	20. Native J	mussels w	ere sampl	ed in Janı - limit of	ary 2020	in Saint-F	lorent harbo	ur whereas d hy the m	caged mu imerical v	ssels were	collected	l in Septer	nber 202() in the fo	ur sites. A	medium
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Species	Site	Ag	Al	As	Ba	Cd	Co	ų	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Se	Sn	V	Zn
Limpet	STARESO	0.40	73.30	26.59	0.48	1.58	0.18	4.17	8.85	966.20	3.15	0.66	3.58	2.43	0.08	0.89	0.01	0.56	55.74
	Calvi	0.12	346.75	13.81	1.69	0.40	0.35	7.11	86.23	1617.90	6.23	0.55	4.87	4.75	0.10	1.28	0.22	3.45	176.74
	Ile Rousse	0.11	335.25	37.01	2.73	0.69	0.29	2.00	41.88	1527.78	5.54	0.65	2.00	7.97	0.05	1.16	0.08	1.50	107.64
	Saint-Florent	0.12	224.38	11.37	1.06	0.39	0.50	3.94	25.75	1172.51	13.04	0.81	5.18	1.71	0.02	0.78	0.06	1.28	89.27
Sea cucumber	STARESO	0.01	34.54	13.03	1.18	0.03	0.19	0.36	2.51	23.91	0.56	2.02	2.37	1.39	0.02	4.09	0.20	0.49	24.85
	Calvi	0.02	64.88	6.26	1.32	0.05	0.20	0.49	4.65	118.56	1.25	9.13	2.32	1.41	0.03	3.16	0.20	0.47	29.72
	Ile Rousse	0.02	64.94	13.83	1.03	0.04	0.16	0.40	4.21	140.80	1.76	2.76	1.67	1.64	0.02	3.25	0.24	0.49	34.23
	Saint-Florent	0.00	62.79	15.51	1.53	0.05	0.43	0.85	3.32	151.74	23.25	5.51	3.54	0.92	0.02	4.03	0.09	0.70	27.93
Mussel (native)	Saint-Florent	0.02	363.17	15.89	1.65	0.47	1.15	5.09	28.29	760.59	28.71	1.77	4.96	1.15	0.03	4.83	0.03	1.29	156.41
Mussel (caged)	STARESO	0.01	9.81	12.24	0.22	0.49	0.27	2.37	3.32	87.46	1.57	0.38	1.31	0.39	0.02	2.06	0.01	0.49	98.23
	Calvi	0.04	19.40	5.56	0.27	0.52	0.21	1.60	73.77	134.46	1.11	0.27	0.82	1.13	0.02	0.99	0.16	0.46	182.64
	Ile Rousse	0.01	51.62	9.98	0.59	0.33	0.37	2.91	25.06	229.19	4.92	0.39	1.22	0.98	0.02	1.83	0.05	0.49	121.86
	Saint-Florent	0.02	70.45	9.92	0.45	0.32	0.41	4.75	31.56	309.66	5.68	0.40	1.39	1.34	0.02	1.85	0.06	0.47	134.13

Ile

galloprovincialis sampled in four Corsican harbours (STARESO, Calvi,

Mean trace element concentrations (medium bound values; μg^{-1} dry weight) in limpets Patella sp., sea cucumbers H. tubulosa, and mussels M.

Table 1

western Mediterranean mussels (Marchand et al. 2009; Santos-Echeandía et al. 2021) and the concentrations in mussels caged along Corsican coastline (Mauffret et al. 2018). Similarly, Cu concentrations in limpets and sea cucumbers from Ile Rousse, Saint-Florent, and Calvi exceeded levels reported in previous studies (Campanella et al. 2001; Conti et al. 2015, 2017; Montero et al. 2021; Türkmen et al. 2005; Warnau et al. 2006). Sediments could be a potential source for Cu bioaccumulation in molluscs since high Cu concentrations in sediments from Calvi bay have been reported (Mauffret et al. 2018). In the present study, the high Cu concentrations could be related to shipping since anti-fouling paints and ship scrubbers have been described as substantial anthropogenic sources of Cu in coastal environments (Richir et al. 2021; Warnken et al. 2004). These results are in agreement with port areas being hotspots of Cu contamination (Santos-Echeandía et al. 2021).

The mean concentrations of several TEs (i.e., Ag, As, Co, Cd, Cr, Ni, Pb, Se, Sb and Zn) in mussels from Corsican harbours (Table 1) were in the range of concentrations reported in caged mussels placed in offshore stations close to our study sites (Mauffret et al. 2018) and in a previous study in Corsica (Richir and Gobert 2014). In the present study, the mean concentrations of Cd and Pb in mussels and sea cucumbers were below the levels reported in previous work (González-Wangüemert et al. 2018; Santos-Echeandía et al. 2021; Warnau et al. 2006), thus suggesting a low contamination of these TE in the harbours. This result is in accordance with the Cd concentrations in limpets which were below the lower limit of the Cd baseline range defined for P. caerulea in the Tyrrhenian Sea (Conti et al. 2017). In the present study, Pb levels in limpets were, however, higher than baseline for P. caerulea along Italian coastline (Conti et al. 2017) but were comparable to Pb levels in P. caerulea from Iskenderum harbour area in Turkey (Türkmen et al. 2005). Lead contamination in harbours is mostly archived in sediment, often originating from older sources such as leaded gasoline, battery factories, and coal combustion (Layglon et al. 2020). Moreover, Pb can be remobilized into the water column and transferred to aquatic resources consumed by humans (Kalnejais et al. 2010). Higher Zn levels in limpets and sea cucumbers were observed in the Corsican harbours (except for STARESO; Table 1) compared to previous studies (Table S5). This high Zn bioaccumulation could be related to antifouling paints and aluminiumbased galvanic anodes which have been highlighted as a source of Zn enrichment in harbour sediments (Caplat et al. 2020; Richir et al. 2021). The TEPI was calculated to compare the overall TE contamination

between the study sites (Fig. 2).

This index has been mostly applied to the seagrass *Posidonia oceanica*, the sea urchin *Paracentrotus lividus*, and the mussel *M. galloprovincialis* (El Idrissi et al. 2020; Richir and Gobert 2014; Ternengo et al. 2018). For the first time, the TEPI have been applied to concentrations determined in *Patella sp.* and *H. tubulosa*. When each species was considered separately, the TEPI values were lower in STARESO harbour than in Calvi, Ile Rousse, and Saint-Florent, indicating a higher global TE contamination in these latter sites (Fig. 2).

There was no quantifiable level of PAHs in samples from STARESO, confirming the low PAH contamination pressure on this reference site (Table 2). Conversely, the very potent BaP was quantified in sea cucumbers from Ile Rousse and mussels from Saint-Florent. Moreover, the potent DBA was detected in sea cucumbers and mussels from Saint-Florent. The concentrations of PAH congeners (i.e., N, FA, P, A, PY, BaA, BkF, BaP, and BPE) in mussels (Table 2) did not exceed their respective Environmental Assessment Criteria (EAC) values set by OSPAR (OSPAR 2009), thus suggesting a good environmental status of the study sites regarding PAH contamination. The PAH concentrations in caged mussels from Corsican harbours (Table 2) were in the range of concentrations reported in caged mussels placed in offshore stations close to our study sites (Mauffret et al. 2018). Caged mussels from Saint-Florent exhibited relatively high concentrations of high molecular weight PAHs (Table 2) which are mainly derived from the incomplete combustion of organic matter and traffic exhaust (Yu et al. 2021). This PAH contamination might be partly due to a boat fire which occurred in



Fig. 2. Trace element contamination of Corsican harbours (STARESO, Calvi, Ile Rousse, and Saint-Florent) using trace element pollution index (TEPI) for each bioindicator species, i.e., limpets (A), sea cucumbers (B), and caged mussels (C), sampled in September 2020.

Saint-Florent harbour in September 2020 (i.e., 5 days before sampling).

The \sum PAH4 measured in the present study (Table 2) were in the range of concentrations reported in molluscs representative of French and European diets (Chiesa et al. 2018; Conte et al. 2016; EFSA 2008; Martorell et al. 2010; Veyrand et al. 2013).

In the present study, the sum of the 16 EPA priority PAHs (Andersson and Achten, 2015) measured in limpets was also lower compared to *P. vulgata* sampled in different Sicilian harbours (Gianguzza and Orecchio 2006). Moreover, we observed similar PAH levels in sea cucumbers *H. tubuosa* from Corsican harbours than in *H. polii* sampled in Mediterranean coasts of Spain (León et al. 2021) and Southern Italy (Biandolino et al. 2022).

The mean concentrations of Σ iPCB in mussels from the four harbours in Northern Corsica (Table 3) were in line with the mean levels measured in molluscs and crustaceans consumed by French population (Sirot et al. 2012), mussels sampled at Milan market (Chiesa et al. 2018), and mussels from the River Ebro mouth (Campillo et al. 2019). The Σ iPCB concentrations in mussels from Corsican harbours were similar to levels found in caged mussels placed in areas close to our study sites (Mauffret et al. 2018). Moreover, the concentrations of PCB congeners

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Mean polycyclic a	romatic hydroca	arbon (F	AH) co.	ncentra	tions (m	edium bo	und val	ues; ng £	ξ^{-1} wet	weight)	in limpe	sts, sea cı	ucumbe	rs, and m	ussels san	npled fro	m four	Corsican	harbou	s (STAF	ESO, Cal	vi, Ile Ro	ousse, and
Saint-Florent) in 2 (LD) were replace	020. \sum PAH4 (n 1 by the numeri	ıg g ⁻¹ wı ical valu	et weigh tes of Ll	ht) is the D/2 and	e sum of . 1 those b	BaA, CR, elow the	BbF, and limit of	l BaP cor quantifi	ncentrat cation (ions. A 1 LQ) wei	re report	bound ar ed as LQ	oproach 1/2.	was used	for estim:	ating cor	ıtaminar	it concer	Itrations	: results	below the	e limit of	detection
Species	Site	Гом п	rolecular	r weight	PAHs						High mole	ecular we	ight PAF	łs									∑PAH4
		z	ΒT	в	ANY	ANA	F	DBT	Р	V	FA III	ıFA P	Y B	aA CR	BbF	BkF	BeP	BaP	PE	NI	DBA	BPE	
Limpet	STARESO	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.2 0.	.2 0	.1	.1 0.	2 0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.5
	Calvi	2.5	0.1	0.1	0.1	0.1	0.1	0.2	2.5	0.4	0.9 2.	.3 0	2	.8	7 0.5	0.5	1.4	0.3	0.2	0.2	0.2	0.8	4.3
	Ile Rousse	2.1	0.3	0.2	0.1	0.1	0.2	0.2	2.2	0.3	0.2 2.	.2 0	.1 0	.3 1.	3 0.2	0.2	0.8	0.2	0.2	0.2	0.2	0.5	1.9
	Saint-Florent	5.3	0.1	0.3	0.1	0.1	0.1	0.2	9.7	1.1	0.2 5.	.2 0	.3 0	.5	0 0.2	0.2	0.5	0.2	0.2	0.2	0.2	0.5	2.9
Sea cucumber	STARESO	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.2 0.	.2 0	.1 0	.1 0.	2 0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.5
	Calvi	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.3 0.	.3 0	.1 0	.2	8 0.9	1.0	0.9	0.3	0.3	1.0	0.3	3.1	2.1
	Ile Rousse	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.5 0.	.5	.1 0	.3 0.	5 1.2	1.2	1.1	0.8	0.2	1.1	0.3	4.0	2.7
	Saint-Florent	4.6	0.1	3.4	0.1	0.5	1.3	0.2	1.6	0.2	0.4 0.	.4	.1 0	.1	4 0.2	0.2	0.2	0.2	0.9	0.2	0.3	0.8	0.8
Mussel (native)	Saint-Florent	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.3 0.	.7 0	.2	.4 0.	9 0.5	0.5	0.5	0.3	0.6	0.2	0.4	0.8	2.1
Mussel (caged)	STARESO	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.2 0.	.2 0	.1 0	.1 0.	2 0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.5
	Calvi	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.2 0.	.2 0	.1 0	.1 0.	2 0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.5
	Ile Rousse	0.8	0.1	0.1	0.1	0.1	0.1	0.2	0.8	0.1	0.2 0.	.2 0	.1 0	.3 0.	2 0.5	0.5	0.5	0.2	0.2	0.2	0.2	0.5	1.1
	Saint-Florent	0.8	0.1	0.1	0.1	0.1	0.1	0.2	2.5	2.5	4.1 4.	.3 1	4.4	.1 10.	7 2.9	2.4	1.5	0.5	0.5	0.2	0.5	0.5	18.2

Mean polychlorin	ated biphenyl (P	CB) conce	entrations ((medium b	ound values	s; ng g^{-1} w	et weight) i	n limpets, se	sa cucumbe.	rs, and muss	sels sampled	l from four (Corsican har	bours (STAF	RESO, Calvi,	Ile Rousse,	and Saint-
Florent) in 2020.	\sum iPCB is the me	san sum ce	oncentratio	on (ng g^{-1}	wet weight) of the inc	licator PCBs	; (PCB 28, 5;	2, 101, 138,	, 153, and 1	80) and ADi	D _{iPCB} (ng kg	⁻¹ day ⁻¹) is	s average da.	ily dose of Σ	DiPCB assoc	iated with
seafood consumpt	tion. A medium b.	ound app.	roach was i	used for est	timating cor	ıtaminant c	concentratio	ns: results be	elow the lim	uit of detection	on (LD) wer	e replaced by	y the numeri	ical values o	fLD/2 and tl	iose below	he limit of
(T) nonnonninnh	and the second	· · · · · · · · · · · · · · · · · · ·															
Species	Site	PCB7	PCB28	PCB35	PCB52	PCB77	PCB101	PCB105	PCB118	PCB135	PCB138	PCB153	PCB156	PCB169	PCB180	∑iPCB	ADD _{iPCB}
Limpet	STARESO	0.1	0.1	0.1	0.1	0.2	0.4	0.09	1.0	0.2	5.3	6.6	0.3	0.1	1.7	14.2	4.3
	Calvi	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.1	0.1	0.8	0.2	0.1	0.1	0.1	1.2	0.4
	Ile Rousse	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.2	0.1	0.8	0.2	0.1	0.1	0.1	1.2	0.4
	Saint-Florent	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.3	0.1	0.8	0.2	0.1	0.1	0.1	1.2	0.4
Sea cucumber	STARESO	0.1	0.1	0.1	0.1	0.1	0.3	0.02	0.3	0.2	2.5	3.3	0.2	0.2	0.3	6.5	2.0
	Calvi	0.1	0.1	0.1	0.1	0.1	0.3	0.02	0.3	0.1	0.8	0.5	0.1	0.1	0.2	1.8	0.6
	Ile Rousse	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.2	0.1	0.8	0.5	0.1	0.1	0.1	1.6	0.5
	Saint-Florent	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.1	0.1	0.8	0.2	0.1	0.1	0.1	1.2	0.4
Mussel (native)	Saint-Florent	0.1	0.1	0.1	0.1	0.1	0.1	0.07	0.2	0.1	0.8	0.4	0.2	0.1	0.2	1.7	0.5
Mussel (caged)	STARESO	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.1	0.1	0.8	0.5	0.1	0.1	0.1	1.6	0.5
	Calvi	0.1	0.1	0.1	0.1	0.1	0.3	0.02	0.3	0.1	0.8	0.5	0.1	0.1	0.1	1.7	0.5
	Ile Rousse	0.1	0.1	0.1	0.1	0.1	0.1	0.02	0.1	0.1	0.8	0.2	0.1	0.1	0.1	1.2	0.4
	Saint-Florent	0.1	0.1	0.1	0.1	0.1	0.6	0.02	0.3	0.1	0.8	0.5	0.1	0.1	0.1	2.1	0.6

Table 3

(PCB 28, 52, 101, 118, 138, 153, and 180) in mussels were all below their respective Environmental Assessment Criteria (EAC) values (Beyer et al. 2017; Mauffret et al. 2018), thus demonstrating a low PCB contamination in the study sites. Few studies investigated PCB content in Patella spp. and mainly focused on Atlantic coastal areas (Peña-Méndez et al. 1996; Pérez et al. 2019; Tena and Montelongo 1999; Viñas et al. 2018). Nevertheless, the PCB concentrations in limpets observed in the present study were in line with a previous study conducted along the Mediterranean coast of Israel (Herut et al. 1999) and suggest a low PCB contamination. In the present study, the PCB congeners mainly contributing to Σ iPCB were PCB 138 and PCB 153 (Table 3). These highly chlorinated congeners are generally predominant in marine molluscs (Benali et al. 2017; EFSA 2010; Giandomenico et al. 2016; Herceg-Romanić et al. 2014; Kožul et al. 2009; Perugini et al. 2004; Scarpato et al. 2010) due to their molecular structure and high lipophilicity, which facilitate their accumulation in aquatic food web (Naso et al. 2005) and make them resistant to metabolic degradation by molluscs (Vidal-Liñán et al. 2016).

Contaminant levels in soft tissues of molluscs are influenced by abiotic parameters (e.g., contaminant concentration in the water) (Pérez-López et al. 2003) and biotic factors such as age, size, and reproductive status (Boyden 1977; Cabral-Oliveira et al. 2015; Fattorini et al. 2008; González-Fernández et al. 2016; Pedro et al. 2021). The origin of the organisms can also affect contaminant bioaccumulation since sampled populations may exhibit adaptative responses to a chronic contamination or environmental conditions, and distinct initial physiological status (Lacroix et al. 2015; Mersch et al. 1996; Silva et al. 2018). Relationship between contaminant levels in soft tissues and body size have been investigated on species of the genus Patella, notably for metal contamination (Bebianno et al. 2003; Collado et al. 2006; Cravo et al. 2004; Cubadda et al. 2001; Nakhlé et al. 2006; Ramelow 1985). The results obtained by these studies differed according to the metal and the species studied and no clear pattern was observed in the variations in body levels of metals with body size (Reguera et al. 2018).

Consequently, passive monitoring of contaminants using native organisms has one major drawback: variation of these biotic factors in sampled organisms may hamper accurate interpretation of the results (Besse et al. 2012). Active approaches, based on transplanted organisms, have been developed with the aim of minimizing these cofounding factors. The results of the present study regarding contaminant levels in native limpets and sea cucumbers (passive biomonitoring) and transplanted mussels (active biomonitoring) should therefore be interpreted with caution. Moreover, the *Patella* organisms sampled were not identified to the species level, although according to their distribution and abundance in the intertidal areas sampled, they can be expected to comprise mainly *P. caerulea, P. ulyssiponensis*, and *P. rustica* (Bouzaza and Mezali 2018). Future studies should identify the sampled limpet species by using molecular techniques as proposed by Zaidi et al. (2022), to ensure the relevance of conclusions regarding contaminant levels.

The health risk associated with seafood consumption was evaluated by comparing measured contaminant concentrations with available legal limits set by the European Commission (European Commission 2006). Cadmium and lead concentrations in mussels (Table S6) were compliant with the maximum levels. Moreover, all hazard quotients (HQ) for TEs were lower than 1 (Table S7). These results suggest that the health risks associated with Al, Ba, Cd, Co, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, and Zn exposure for average seafood consumers were insignificant. The present study analysed total As, Cr, and V concentrations, however, refence doses have been established only for inorganic As, CrIII, CrVI, organic Sn, and V pentoxide. There is a lack in literature data on As, Cr, Sn, and V speciation in seafood (Copat et al. 2018), thus HQ for these elements were not determined in the present study.

Regarding regulated PAHs, B(a)P and $\sum PAH4$ (sum of BaA, CR, BbF, and BaP) concentrations in limpets, sea cucumbers, and mussels from Corsican harbours (Table S6) were lower than the European maximum levels (European Commission 2011a). All the HQ values for PAH

congeners were lower than 1 (Table S8), suggesting that seafood consumption would not cause non-carcinogenic effects in humans. Regarding carcinogenic effects, the MOE values for PAH4 ranged from 6.1×10^4 to 2.1×10^6 (Table S8), thus far exceeding 10^4 which is the value recommended by EFSA. Both HQ and MOE approaches suggest that exposure to PAHs through seafood consumption is not a major health problem. The MOE approach, however, disregards many PAH congeners for which toxicological data are lacking. In addition to the 16 USEPA priority PAHs (Andersson and Achten 2015) which are often monitored, we also detected the presence of benzothiophene, 2-methylfluoranthene, benzo(e)pyrene, and perylene in the studied species. Investigations on the toxicity of these congeners are needed to fully assess environmental and health risks related to PAH.

In the present study, the sum concentrations of the six indicator PCBs $(\sum iPCB)$ in marine organisms (Table S6) were below the maximum level established in the European Union (European Commission 2011b). Mean dietary intakes of iPCBs (ADD_{iPCB}) were 0.4–4.3 ng kg⁻¹ day⁻¹ for consumption of limpets, 0.4–2 ng $\mathrm{kg}^{-1}~\mathrm{day}^{-1}$ for sea cucumbers, and $0.4-0.6 \text{ ng kg}^{-1} \text{ day}^{-1}$ for mussels (Table 3). Ingestion of food represents more than 90 % of the iPCB exposure in the general population (EFSA 2005). In France, mean exposure to iPCBs through food ingestion was estimated at 2.78 ng kg⁻¹ day⁻¹ in adults, crustaceans and molluscs contributing to 4 % (mean exposure of 0.1 ng kg⁻¹ day⁻¹) of the total exposure to iPCBs (Sirot et al. 2012). The ADD_{iPCB} in the present study were in line with estimated exposure of French population, except for ingestion of limpets (4.3 ng kg⁻¹ day⁻¹) and sea cucumbers (2 ng kg⁻¹ day⁻¹) from STARESO which represented 43 % and 20 % of the total tolerable daily intake for iPCBs of 10 ng kg⁻¹ day⁻¹ (AFSSA 2007; Baars et al. 2001), respectively.

The contaminant intakes estimated in the present study for TE, PAH, and PCB should be considered as a lower estimate of their total exposure, since their intake through other food items were not considered. Moreover, the ingestion rate used to estimate dietary exposure in the present study was extracted from the French total diet study data (ANSES et al. 2017) and might not be representative of the local population since Corsica was not surveyed in the latter study (Dubuisson et al. 2019). It should be noticed that permissible levels for mercury, dioxinlike-PCBs and polychlorinated dibenzo-p-dioxins and dibenzofurans have been set in fish and seafood in the EU (European Commission 2011b), however, these contaminants were not measured in the present study. Moreover, the present work was partly conducted during the COVID-19 lockdown which restricted travel, fishing, tourism, and industrial activities worldwide, and thus affected contaminant levels in marine environments and ecosystem health (Cecchi 2021; Loh et al. 2021; Patterson Edward et al. 2021; Yang et al. 2022; Yoon et al. 2022). A long-term biomonitoring of Corsican harbours for organic and inorganic contaminants is thus needed to detect an eventual increase in contaminant levels.

In summary, this study demonstrates a relatively low contamination in the Corsican harbours studied compared to other Mediterranean coastal areas, however, results reveal relatively high concentrations of some trace metals (i.e., Cu, Pb, Zn). To our knowledge, this is the first study biomonitoring TE, PAH, and PCB in Corsican harbours. Contaminant levels in mussels, limpets, and sea cucumbers sampled in the harbours were compliant with European regulatory limits. Regarding potential human health risks associated with seafood consumption, results suggested no adverse effects for human health. However, there are substantial data gaps with respect to exposure as well as toxicity of many chemical contaminants detected in seafood in the present study. Further research is necessary to determine sanitary and environmental thresholds of these contaminants to enable ecological assessment.

CRediT authorship contribution statement

Justine Castrec: Conceptualization, Methodology, Investigation, Writing – original draft, Formal analysis, Visualization. Marion Pillet: Conceptualization, Methodology, Investigation, Writing – review & editing. Justine Receveur: Investigation. Quentin Fontaine: Visualization. Stéphane Le Floch: Resources. Carine Churlaud: Investigation, Writing – review & editing. Pierre Lejeune: Project administration. Sylvie Gobert: Writing – review & editing. Hélène Thomas: Conceptualization, Funding acquisition, Supervision, Writing – review & editing, Project administration. Michel Marengo: Conceptualization, Funding acquisition, Supervision, Writing – review & editing, Project administration.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2023.114578.

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