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Potential human health risks from metals (Hg, Cd, and Pb) and polychlorinated biphenyls (PCBs) via seafood consumption: Estimation of target hazard quotients (THQs) and toxic equivalents (TEQs)

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ABSTRACT

Edible marine species (fish, cephalopod molluscs, crustaceans) from the Adriatic Sea were analyzed for content in heavy metals (Hg, Cd and Pb) and polychlorinated biphenyls (PCBs). Health risks to human via dietary intake of seafood were assessed by the target hazard quotients (THQs) and the toxic equivalent factors (TEFs). Mercury maximum concentrations corresponded to fish (0.07–1.56 μg g^{-1} w.w.), followed by cephalopod molluscs (0.10–0.55 μg g^{-1} w.w.), and crustaceans (0.27–0.33 μg g^{-1} w.w.). Cadmium levels in cephalopods (0.18–0.59 μg g^{-1} w.w.) were higher than those in fish (0.01–0.05 μg g^{-1} w.w.) and crustaceans (0.02–0.04 μg g^{-1} w.w.), while for Pb the concentrations were generally low (fish: ND–1.18 μg g^{-1} w.w., cephalopods: ND–0.17 μg g^{-1} w.w., crustaceans: ND–0.03 μg g^{-1} w.w.). For PCBs, concentrations in fish, cephalopods and crustaceans ranged between 141 and 3406 ng g^{-1} l.w., 190 and 542 ng g^{-1} l.w., and 202 and 429 ng g^{-1} l.w., respectively. Cd and Pb THQ values as well as estimates of PCB TEQ exposure indicated the absence of health risks through consumption of the various seafood. In contrast, mercury TEQs values due to consumption of certain fish species (albacore, rosefish and thornback ray) indicated that human health risk might be of concern.

1. Introduction

Fish constitutes an important source of proteins, minerals, vitamins and unsaturated essential fatty acids (PUFAs), especially omega-3 PUFAs. Scientific data indicate that fish consumption reduces the risk of coronary heart disease, decreased mild hypertension and prevents certain cardiac arrhythmias (Kris-Etherton et al., 2002). At the same time, seafood consumption has been reported as an important route of human exposure to a variety of chemical contaminants (Llobet et al., 2003; Usero et al., 2003). Heavy metals, such as mercury, cadmium and lead and polychlorinated biphenyls (PCBs), especially the so-called "dioxin-like" PCBs, deserved special attention among these pollutants, as they represent a group of highly toxic substances accumulating in the tissues of marine organisms and being conveyed through the food chain to human. Reports from literature suggest that these toxicants are responsible of hazardous effects on human health (Luckey and Venugopal, 1977; IPCS, 1993; IARC, 1997; Aschner, 2002). Consequently, potential public health risks from dietary exposure to these pollutants continue to be the subject of must research, regulation and debate. Several methods have been proposed for the assessment of the potential human health risks from these chemical exposure. Current non-cancer risk assessment methods are typically based on the use of the target hazard quotient (THQ), a ratio between the estimated dose of a contaminant and the reference dose below which there will not be any appreciable risk (US EPA, 2000). If such ratio exceeds unity, there may be concern for potential health effects. An other approach for estimating exposure and risks by dietary intake of toxic substances is the use of toxic equivalency factors (TEFs) scheme and TEQ methodology. This approach assigns relative toxicity values to structurally related chemicals in comparison to a reference chemical. Using this latter procedure, human exposure to PCBs via the consumption of seafood has been estimated in several studies (Storelli et al., 2003a,b, 2007a; Judd et al., 2004; Bocio and Domingo, 2005; Darnerud et al., 2006), while few reports on the potential health risk assessment to dietary intake of heavy metals based on target hazard quotient have been found (Han et al., 1998, 2000; Chien et al., 2002; Zheng et al., 2007). In response to this, the presence of heavy metals (Hg, Cd and Pb), as well as "dioxin-like" PCBs and other relevant PCB congeners, in fish and seafood has been evaluated in combination with the health risk associated to their consumption, completing previous studies carried out in the Mediterranean area (Storelli et al., 2003a,b, 2006a,b, 2007a; Storelli and Marcotrigiano, 2004). The differences in the toxic load among the species analyzed are also

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discusses by the examination of some factors that might influence exposure in the marine organisms.

2. Materials and methods

2.1. Collection of samples

All marine organisms were collected in the South-Eastern Mediterranean Italian coast (Adriatic Sea) from May to June 2006. The species chosen are very important commercially and have a significant interest for food use. These species include benthic, demersal and pelagic fish, cephalopod molluscs and crustaceans (Table 1). Pooled edible portion of the similar size specimens (length and weight) was taken and stored below −20 °C pending analysis. The number of individual specimens per pool varied depending on the species (fish: no. 15; albacore: no. 8; cephalopod molluscs: no. 15; crustaceans: red shrimp no. 50, pink shrimp no. 80, shrimp no. 80).

2.2. Extraction procedures

The analytical methods for metals have been previously described (Storelli et al., 2005). Briefly, homogenised subsamples (about 2 g) were digested in a HNO_3–$HClO_4$ mixture conc. for Pb and Cd determination, and in a H_2SO_4–HNO_3 mixture conc. for Hg (Sigma–Aldrich, Germany). The extractive analytical procedure and the instrumental conditions for determine polychlorinated biphenyl (PCBs = 8, 20, 28, 35, 52, 60, 77, 101, 105, 118, 126, 138, 153, 156, 169, 180 and 209) concentrations have been, also, described in detail elsewhere (Storelli et al., 2007a). Briefly, aliquots (1–2 g) of the homogenised samples were ground with anhydrous sodium sulphate in a mortar. The mixture was extracted with petroleum ether according to Erney's procedure (Erney, 1983). The extracts were then concentrated and subsamples were taken in order to determine the tissue fat content by gravimetry. An aliquot of the remaining extract was dissolved in hexane and mixed with H_2SO_4 conc. for the clean up, following the procedure described by Murphy (1972). After centrifugation, the hexane solution was concentrated and transferred on a glass column (i.d. 5 mm) filled with 1 g of florisil (activated at 120 °C for 16 h) for the separation of PCBs from other organochlorine compounds. For the separation of non-*ortho* PCB congeners, 3,3′,4,4′-T_4CB, (IUPAC 77), 3,3′,4,4′,5-P_5CB (IUPAC 126), and 3,3′,4,4′,5,5′-H_6CB (IUPAC 169) from other PCBs the method reported by Tanabe et al. (1987a) was used.

2.3. Instrumental analysis

Quantitative determinations of Pb and Cd were made using an atomic absorption spectrophotometer (Analyst 800 P.E.) equipped with a heated graphite furnace system (THGA-800 P.E.), while Hg was determined by the cold vapour technique after reduction by $SnCl_2$ (FIMS 100 P.E.). For PCBs, analyses were made on a Carlo Erba HR gas chromatograph 8000 Top with automatic injection system and with an electron capture detector ECD-800, Ni^{63} (temperature: 330 °C). For all the analyses a fused-silica capillary column DB-5 Supelco (length = 30 m, inside diameter 0.25 mm and film thickness 0.25 μm), was used. Hydrogen at a flow rate of 1 ml/min was used as gas carrier, nitrogen as make-up gas 60 ml/min. The individual PCB congeners were determined against the corresponding individual standards obtained from ULTRA Scientific, Inc. (chemical purity 99%). The limits of detection (LOD) was 5 pg g^{-1} fish muscle on a wet wt basis for the PCB congeners.

2.4. Quality control and assurance

To ensure the quality of metal analysis a standard reference material (National Research Council of Canada; DORM-2 dogfish muscle) was used. Recovery ranged from 95% to 100% for all investigated elements. For PCBs, the reference material employed was CRM 349. The recovery for each PCB (28, 52, 101, 118, 153, 180, and 138) quantified in the certified material ranged from 91% to 102%. The recoveries for the other PCB congeners, varying between 90% and 110%, were determined adding known amounts of PCB standards (at three levels of concentrations) to samples before extraction (method of additions). Residues in 100% of the samples were confirmed by gas–liquid chromatography–mass spectrometry (Fisons MD 800).

2.5. Target hazard quotient (THQ)

The methodology for estimation of target hazard quotient (THQ) although does not provide a quantitative estimate on the probability of an exposed population experiencing a reverse health effect, but it offers an indication of the risk level due to pollutant exposure. This method was available in US EPA Region III Risk-based Concentration table (US EPA, 2000) and it is described by the following equation:

$$\mathrm{THQ} = \frac{\mathrm{EF} \times \mathrm{ED} \times \mathrm{FIR} \times C}{\mathrm{RFD} \times \mathrm{WAB} \times \mathrm{TA}} \times 10^{-3}$$

where EF is exposure frequency (365 days/year); ED is the exposure duration (70 years), equivalent to the average lifetime; FIR is the food ingestion rate (fish: 36 g/person/day; cephalopod molluscs: 9.80 g/person/day and crustaceans: 5.42 g/person/day) (FAO, 2005); *C* is the metal concentration in seafood (μg g^{-1}); RFD is the oral reference dose (Hg = 5×10^{-4} μg g^{-1}/day, Cd = 1×10^{-3} μg g^{-1}/day, Pb = 0.004 μg g^{-1}/day) (US EPA, 1997, 2000); WAB is the average body weight (60 kg), and TA is the averaging exposure time for non-carcinogens (365 days/year × ED). Total THQ (TTHQ) of heavy metals for individual seafood is the sum of the follow composition: TTHQ (individual seafood) = THQ (toxicant 1) + THQ (toxicant 2) + THQ (toxicant *n*).

Table 1
Overview of sampled species

Scientific name	Common name	Length (cm)	Weight (g)	Number of pool	Habitat
Fish					
Thunnus alalunga	Albacore	72.0 ± 2.7	6015.0 ± 10.9	4	Pelagic
Trachurus trachurus	Horse mackerel	34.2 ± 4.9	338.2 ± 14.7	12	Pelagic
Trachurus mediterraneus	Mediterranean horse mackerel	30.8 ± 3.2	310.5 ± 13.6	10	Pelagic
Sardina pilchardus	Pilchard	17.3 ± 0.5	44.6 ± 0.9	10	Pelagic
Engraulis encrasicholus	European anchovy	13.4 ± 1.3	12.8 ± 0.7	9	Pelagic
Lepidopus caudatus	Frostfish	70.2 ± 2.7	368.3 ± 9.7	8	Pelagic
Merluccius merluccius	Hake	20.2 ± 5.3	65.0 ± 7.9	13	Demersal
Phycis blennoides	Greater forkbeard	37.4 ± 4.7	505.0 ± 19.6	10	Demersal
Pagellus erythrinus	Sea bream	13.1 ± 4.2	36.6 ± 1.1	9	Demersal
Lepidorhombus boscii	Four spotted megrim	18.5 ± 1.7	58.0 ± 2.5	8	Benthic
Lepidorhombus whyffjagonis	Megrim	10.7 ± 1.1	28.8 ± 2.7	8	Benthic
Helicolenus dactylopterus	Rosefish	18.0 ± 5.0	109.0 ± 8.4	11	Benthic
Mullus barbatus	Striped mullet	14.9 ± 3.2	36.0 ± 1.3	13	Benthic
Conger conger	Conger	62.8 ± 4.4	493.0 ± 18.5	8	Benthic
Trigla lucerna	Yellow gurnard	15.3 ± 1.5	32.7 ± 3.9	12	Benthic
Raja miraletus	Brown ray	45.4 ± 3.1	456.0 ± 15.3	8	Benthic
Raja asterias	Starry ray	44.2 ± 3.4	470.0 ± 18.7	8	Benthic
Raja clavata	Thornback ray	60.8 ± 2.8	630.0 ± 20.6	8	Benthic
Cephalopod molluscs					
Loligo vulgaris	European squid	20.8 ± 3.3	336.7 ± 9.6	10	Pelagic
Illex coindetii	Broadtail squid	13.8 ± 8.6	179.2 ± 27.2	13	Pelagic
Eledone cirrhosa	Curled octopus	8.5 ± 2.6	111.7 ± 10.9	10	Benthic
Eledone moschata	Horned octopus	10.7 ± 3.1	207.8 ± 13.8	11	Benthic
Octopus salutii	Spider octopus	9.0 ± 2.9	176.1 ± 21.9	9	Benthic
Crustaceans					
Aristeus antennatus	Red shrimp	3.7 ± 0.3	13.5 ± 0.8	8	Benthic
Parapeneus longirostris	Pink shrimp	2.1 ± 0.2	5.8 ± 0.7	10	Benthic
Plesionika martia	Shrimp	1.8 ± 0.2	5.3 ± 0.6	7	Benthic

2.6. Toxic equivalent factor (TEF) scheme and toxic equivalency (TEQ) methodology

Toxic equivalency factors (TEFs) are being developed to assess the potency of polyhalogenated aromatic hydrocarbons, including polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs). Toxic equivalency factors (TEFs) are essentially weighting factors by which the toxicity of a congener is compared to that of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). In this way, concentrations of individual PCB congeners are multiplied by their TEFs and the results summed to give a toxicity value expressed in TCDD toxic equivalents (TEQs) through the following equation:

$$\text{PCB TEQ} = \sum(\text{PCB}_i \times \text{TEF}_i)$$

The World Health Organization Consultation recommendes the TEF approach, applying the recently established WHO TEFs (Van den Berg et al., 2006), for estimating the weekly intake in human of non-*ortho* (3,3′,4,4′-tetraCB (PCB 77), TEF = 0.0001; 3,3′,4,4′,5-pentaCB (PCB 126), TEF = 0.1; 3,3′,4,4′5,5′-hexaCB (PCB 169), TEF = 0.03) and mono-*ortho* PCBs (2,3,3′,4,4′-pentaCB (PCB 105), TEF = 0.00003; 2,3′,4,4′,5-pentaCB (PCB 118), TEF = 0.00003; 2,3,3′,4,4′,5-hexaCB (PCB 156), TEF = 0.00003) in units of TCDD equivalents for comparison to the Tolerable Weekly Intake (TWI) of 8 pg TEQs/kg b.w., recently established for these substances (Official Journal of the European Communities, 2006). These estimates assumed that non-detected isomer concentrations equal to zero (ND = 0), as well as non-detected isomer concentrations equal to half of the LOD ($\text{ND} = \frac{1}{2}\text{LOD}$) and LOD (ND = LOD).

3. Results and discussion

3.1. Metal levels

The concentrations of different metals (Hg, Cd and Pb) (μg g^{-1} wet weight) in the edible portion of fish, cephalopod molluscs and crustaceans are given in Table 2. The concentrations of mercury varied widely among the different organisms in the order fish > cephalopod molluscs > crustaceans. A comparison of the levels of this metal in fish revealed the highest mean concentrations in albacore (1.56 μg g^{-1}) and different benthic fish (rosefish: 1.25 μg g^{-1}; thornback ray: 0.87 μg g^{-1}; striped mullet: 0.70 μg g^{-1}), while small pelagic fish, as pilchard (0.09 μg g^{-1}) and European anchovy (0.07 μg g^{-1}) presented the lowest mean levels. The other fish showed intermediate concentrations between 0.22 and 0.68 μg g^{-1}. Among cephalopods, octopuses and cuttlefish (curled octopus: 0.51 μg g^{-1}, horned octopus: 0.49 μg g^{-1}, spider octopus: 0.55 μg g^{-1}) presented higher mean levels than squid (European squid: 0.13 μg g^{-1}, broadtail squid: 0.10 μg g^{-1}), while for crustaceans no difference in mercury levels was observed (red shrimp: 0.27 μg g^{-1}; pink shrimp: 0.28 μg g^{-1}, shrimp: 0.33 μg g^{-1}). These interspecific differences are, surely, due to trophic level, foraging method/location and propensity of this metal to undergo biomagnification in the food chain. It is not, thus, surprising that tunas, that are high on the trophic levels, had the highest levels of mercury and that bottom feeding fish had the next highest. Animals live in close association with sediments in which they bury and from where they mainly feed are, in fact, more exposed to eventually sediment-associated contamination than other fish (Storelli et al., 1998, 2003c, 2006a). In this general picture fit well also the findings encountered in cephalopods. Benthic cephalopods (octopuses and cuttlefish) feeding mainly on bottom invertebrates were richer in mercury than pelagic ones (squid). Another relevant point when evaluating the variability in data is the fish size. There is, in fact, a large body of data showing that mercury tissue concentrations increase with age/size of marine fauna (Joiris et al., 1999; Storelli and Marcotrigiano, 2000; Storelli et al., 2007b). So, the largest and potentially oldest fish examined showed high levels of mercury (e.g. horse mackerel: 0.68 μg g^{-1}; Mediterranean horse mackerel: 0.51 μg g^{-1}; frostfish: 0.59 μg g^{-1}). However, independently from the interaction complexity leading to a different toxic load among species, mercury concentrations here detected were in agreement with those reported in seafood from either the same environment (Storelli et al., 2003c, 2006a; Martí-Cid et al., 2007) or other marine areas (Sivaperumal et al., 2007).

Table 2
Range and mean concentrations (μg g^{-1} wet weight) of heavy metals for each species

Species	Hg	Cd	Pb
Fish			
Albacore	0.88–2.34	0.03–0.10	0.30–3.13
	1.56 ± 0.49	0.05 ± 0.03	1.18 ± 0.48
Horse mackerel	0.16–2.41	0.01–0.03	ND–0.06
	0.68 ± 0.70	0.02 ± 0.01	0.03 ± 0.02
Mediterranean horse mackerel	0.09–1.62	0.01–0.03	0.02–0.06
	0.51 ± 0.53	0.02 ± 0.01	0.04 ± 0.01
Pilchard	0.07–0.12	0.02–0.04	0.03–0.09
	0.09 ± 0.04	0.03 ± 0.01	0.06 ± 0.02
European anchovy	0.02–0.21	0.01–0.02	0.09–0.10
	0.07 ± 0.09	0.01 ± 0.01	0.10 ± 0.01
Frostfish	0.16–1.15	0.03–0.15	ND–0.04
	0.59 ± 0.32	0.05 ± 0.02	0.03 ± 0.01
Hake	0.04–0.48	0.04–0.08	0.02–0.07
	0.18 ± 0.12	0.04 ± 0.01	0.04 ± 0.01
Greater forkbeard	0.03–0.51	0.03–0.06	ND–0.08
	0.33 ± 0.20	0.04 ± 0.01	0.04 ± 0.02
Sea bream	0.05–0.70	0.01–0.03	0.05–0.09
	0.24 ± 0.19	0.02 ± 0.01	0.06 ± 0.01
Four spotted megrim	0.14–0.69	0.03–0.07	ND–0.02
	0.35 ± 0.19	0.04 ± 0.01	0.02 ± 0.01
Megrim	0.09–0.41	0.04–0.08	0.01–0.02
	0.29 ± 0.15	0.06 ± 0.02	0.01 ± 0.01
Rosefish	0.24–2.98	0.01–0.08	0.03–0.33
	1.25 ± 0.85	0.03 ± 0.01	0.13 ± 0.09
Striped mullet	0.05–2.76	0.01–0.04	0.04–0.18
	0.70 ± 0.73	0.02 ± 0.01	0.06 ± 0.06
Conger	0.13–1.25	0.01–0.04	0.03–0.13
	0.55 ± 0.39	0.02 ± 0.01	0.07 ± 0.03
Yellow gurnard	0.01–0.53	0.01–0.03	ND–0.04
	0.22 ± 0.17	0.02 ± 0.01	0.02 ± 0.01
Brown ray	0.06–0.78	0.02–0.05	ND–0.07
	0.45 ± 0.55	0.03 ± 0.01	0.03 ± 0.02
Starry ray	0.07–0.89	0.01–0.03	ND–0.06
	0.39 ± 0.36	0.02 ± 0.01	0.02 ± 0.02
Thornback ray	0.50–1.24	0.01–0.04	0.03–0.04
	0.87 ± 0.52	0.02 ± 0.02	0.03 ± 0.01
Cephalopod molluscs			
European squid	0.02–0.49	0.26–0.32	ND–0.06
	0.13 ± 0.14	0.29 ± 0.04	0.04 ± 0.01
Broadtail squid	0.01–0.39	0.06–0.27	0.04–0.21
	0.10 ± 0.10	0.18 ± 0.06	0.12 ± 0.05
Curled octopus	0.20–1.01	0.02–0.60	ND–0.06
	0.51 ± 0.22	0.25 ± 0.21	0.05 ± 0.01
Horned octopus	0.14–1.73	0.10–0.73	0.06–0.30
	0.49 ± 0.45	0.32 ± 0.20	0.17 ± 0.05
Spider octopus	0.13–2.15	0.02–2.18	0.07–0.11
	0.55 ± 0.47	0.59 ± 0.56	0.09 ± 0.02
Crustaceans			
Red shrimp	0.16–0.41	0.03–0.05	ND–0.02
	0.27 ± 0.08	0.04 ± 0.01	0.01 ± 0.01
Pink shrimp	0.09–0.69	0.01–0.03	ND–0.04
	0.28 ± 0.22	0.02 ± 0.01	0.03 ± 0.01
Shrimp	0.11–0.65	0.02–0.05	ND–0.03
	0.33 ± 0.25	0.04 ± 0.07	0.02 ± 0.01

Concerning cadmium, the concentrations in cephalopod molluscs (range: 0.18–0.59 μg g^{-1}) were substantially higher than those encountered in fish (range: 0.01–0.05 μg g^{-1}) and crustaceans (range: 0.02–0.04 μg g^{-1}). The capability of cephalopods to concentrate high quantities of cadmium is well known (Miramand and Guary, 1980; Miramand and Bentley, 1992; Bustamante et al., 1998). Bustamante et al. (2002) suggests that the major part of cadmium is associated with lysosomes and cytosolic proteins, which play a function key in the storage and detoxication of this element. Also the heterogeneity of cadmium concentrations among the different species of cephalopod molluscs (curled octopus: 0.25 μg g^{-1}, horned octopus: 0.32 μg g^{-1}, spider octopus: 0.59 μg g^{-1}, European squid: 0.29 μg g^{-1}, broadtail squid: 0.18 μg g^{-1}) is not an unexpected finding but reflects, not only their great taxonomic diversity, but also ecologic difference (Bustamante et al., 1998; Storelli and

Marcotrigiano, 1999; Craig and Overnell, 2003; Raimundo et al., 2004; Storelli et al., 2006a). Conversely to cephalopods, in fish and crustaceans the concentration ranges of cadmium were quite narrow and similar, confirming that there is no trend in tissue residues of this metal based on taxonomic position or trophic status (Lee et al., 2000; Franca et al., 2005). However, the presented results were within the range of values reported for marine organisms from the Mediterranean Sea (Kljakovic et al., 2002; Marijic and Raspor, 2007).

As can be seen in Table 2, lead content spanned a narrow range (fish: ND–1.18 μg g^{-1}; cephalopod molluscs: ND–0.17 μg g^{-1}; crustaceans: ND–0.03 μg g^{-1}) and was always low, except for albacore (1.18 μg g^{-1}). What here revealed agrees perfectly with data in literature, which report a decreased in lead level in the marine environment (Nicolas et al., 1994; Tian and Ruiz-Pino, 1995). The regulation to limit leaded gasoline consumption applied in many European countries since the 1970s, can be responsible of the decline of its levels in the environment. Serial measurements of lead concentration between 1983 and 1992 illustrate, in fact, that the application of such a regulation in Europe has noticeably influenced residue levels in the Mediterranean Sea (Nicolas et al., 1994).

3.2. PCB levels and accumulation patterns

The concentrations of different PCB congeners (ng g^{-1} l.w.) detected in fish, cephalopod molluscs and crustaceans are presented in Table 3. Among the 17 investigated PCBs, congeners 118, 138, 153, 180 were omnipresent in all the species studied, whereas PCBs 8, 20, 28 and 35 were below the limit of detection in all samples. The remaining PCBs were not detected in many samples. In particular, the Frequency of Occurrence (FO%) of PCB 52, 60, 77, 101, 105, 126, 156 and 169 in fish samples were 27.8%, 44.4%, 83.3%, 72.2%, 50.0%, 5.6%, 33.3% and 11.1%, respectively. Among cephalopod mollusc samples the FO% for PCB 52, 60, 126 and 169 was 20% individually, for 77, 105 and 156 was 80% and for PCB 101 was 40%. In crustaceans PCBs 52, 60, 126 and 156 were absent, PCB 77 and 101 were encountered in all samples, while PCB 105 and 169 were in 33.3% of the examined species. Residue levels of total PCBs varied largely among the different species with values ranging from 141 to 3406 ng g^{-1} in fish, from 190 to 542 ng g^{-1} in cephalopod molluscs and from 202 to 429 ng g^{-1} in crustaceans. Among fish, conger conger (3406 ng g^{-1}), rosefish (2572 ng g^{-1}) and thornback ray (1367 ng g^{-1}) showed the highest concentrations, while pilchard (163 ng g^{-1}) and European anchovy (141 ng g^{-1}) encountered the lowest levels. Consistent amounts of PCBs were also present in albacore (1051 ng g^{-1}) and striped mullet (824 ng g^{-1}), while the other fish showed concentrations between 257 and 655 ng g^{-1}. These results highlight that the bioaccumulation of these pollutants in marine organisms is species-specific. The ecological characteristics of the various organisms, such as feeding preferences, general behaviour and trophic level play, in fact, an important role in accumulation of these pollutants (Pastor et al., 1996; García et al., 2000). With regard to the profiles of polychlorinated congeners, the data revealed that hexa- and pentachlorobiphenyls were predominant, collectively accounting from 70.2% to 90.8% in fish, 86.0–88.7% in cephalopod molluscs and from 93.5% to 95.4% in crustaceans. Heptachlorobiphenyl PCB 180 did not exceed 22.0% in fish, 14.0% in cephalopod molluscs and 5% in crustaceans, while tetrachlorobiphenyls made up the smallest percentages of total PCBs (fish: 0.01–13.0%; cephalopod molluscs: 0.4–3.3%; crustaceans: 0.5–2.8%). In terms of individual congener distributions of PCBs, the predominance of PCB 138 and 153 was detected. On average these compounds collectively accounted for 33.8–67.7%, 58.2–73.1%, 19.9–40.6% of total PCBs concentrations in fish, cephalopod molluscs and crustaceans, respectively. Although individual congener proportions were quite similar among the different marine organisms, in crustacean samples the contribution of congener PCB 101 was remarkable, with percentages ranging from 25.2% to 64.6%. A comparison with literature data showed that isomer composition as well as PCB individual congener pattern were similar to those encountered by the majority of authors on various types of marine organisms from the Mediterranean Sea (García et al., 2000; Bayarri et al., 2001; Storelli et al., 2003a,b, 2007a; Garritano et al., 2006). Also accumulation pattern of highly toxic non-*ortho* chlorine substituted PCBs (77, 126 and 169) was in line with literature data showing that generally in fish samples, PCB 77 is the predominant congener of three non-*ortho* coplanars, followed by PCB 126 and PCB 169 (Tanabe et al., 1987b). Among mono-*ortho* PCBs, it was clearly observed the predominance of congener PCB 118, while pentachlorobiphenyl 105 and hexachlorobiphenyl 156 were found only in some species.

Table 3
Total PCB levels (ng g^{-1} l.w.) and percentage contribution of PCB homologues for each species

Species	% Lipid	∑PCBs	Tetra-	Penta-	Hexa-	Hepta-
Fish						
Albacore	0.3	1051	–	37.1	52.9	10.0
Horse mackerel	20.1	655	2.0	16.0	67.4	14.5
Mediterranean horse mackerel	21.2	456	2.6	15.8	62.9	18.6
Pilchard	3.9	163	7.1	12.9	62.8	17.2
European anchovy	1.6	141	13.5	13.5	56.7	16.3
Frostfish	0.5	585	5.8	14.6	67.7	12.0
Hake	0.6	380	0.5	26.3	61.9	11.3
Greater forkbeard	0.2	319	9.4	37.9	33.8	18.8
Sea bream	0.1	313	1.3	21.4	54.0	23.3
Four spotted megrim	0.3	259	–	15.4	63.4	21.2
Megrim	0.3	642	3.3	22.0	64.2	10.6
Rosefish	0.3	2572	1.5	14.8	67.2	16.5
Striped mullet	1.8	824	6.5	36.0	48.2	9.3
Conger	0.8	3406	1.9	15.1	66.9	16.0
Yellow gurnard	0.5	257	7.8	13.6	67.7	10.9
Brown ray	0.1	499	5.0	33.2	41.4	20.2
Starry ray	0.1	339	5.3	30.3	53.1	11.2
Thornback ray	0.1	1367	2.8	42.2	48.6	6.5
Cephalopod molluscs						
European squid	0.2	487	1.4	19.6	69.1	9.9
Broadtail squid	0.2	542	2.7	22.5	65.5	9.2
Curled octopus	0.3	373	2.4	23.9	62.1	11.5
Horned octopus	0.3	333	3.3	25.8	61.8	9.0
Spider octopus	0.2	190	–	13.2	73.1	13.7
Crustaceans						
Red shrimp	0.7	202	0.5	61.8	33.6	4.0
Pink shrimp	0.2	429	2.8	73.5	21.5	2.3
Shrimp	0.1	397	1.5	52.9	40.6	5.0

3.3. Contaminant intake (Hg, Cd, Pb and dioxin-like PCBs) and health threat from consuming seafood

Table 4 shows the estimated weekly intakes (EWIs) and the target hazard quotients (THQs) for heavy metals caused by the consume of seafood. An important aspect in assessing risk to human health from potentially harmful chemicals in food is the knowledge of the dietary intake of such substances, that must remain within determined safety margins. For Hg, Cd and Pb, the World Health Organization has established as "safe" intake level a Provisional Tolerable Weekly Intake (PTWI) of 5 μg kg^{-1} b.w., 7 μg kg^{-1} b.w. and 25 μg kg^{-1} b.w., respectively (World Health Organization, 2003). In our case, Cd and Pb weekly intakes through the consumption of fish (Cd: 0.04–0.25 μg kg^{-1} b.w., Pb: 0.04–4.96 μg kg^{-1} b.w.), mollusc cephalopods (Cd: 0.21–0.67 μg kg^{-1} b.w., Pb: 0.05–0.19 μg kg^{-1} b.w.) and crustaceans (Cd: 0.01–0.18 μg kg^{-1} b.w., Pb: 0.01 μg kg^{-1} b.w.) were lesser than tolerable weekly intake

Table 4
Estimated weekly intake (EWI) of total mercury, cadmium and lead (μg kg^{-1} b.w.) and target hazard quotients (THQs) for individual metals by consumption of seafood

Species	Hg EWIs	Cd EWIs	Pb EWIs	Hg THQs	Cd THQs	Pb THQs	TTHQ[a]
Fish							
Albacore	6.55	0.21	4.96	1.87	0.03	0.18	2.08
Horse mackerel	2.86	0.08	0.13	0.82	0.01	0.005	0.84
Mediterranean horse mackerel	2.14	0.08	0.17	0.61	0.01	0.01	0.63
Pilchard	0.38	0.13	0.25	0.11	0.02	0.01	0.14
European anchovy	0.29	0.04	0.42	0.08	0.01	0.02	0.11
Frostfish	2.48	0.21	0.13	0.71	0.03	0.005	0.75
Hake	0.76	0.17	0.17	0.22	0.02	0.01	0.25
Greater forkbeard	1.39	0.17	0.17	0.40	0.02	0.01	0.43
Sea bream	1.01	0.08	0.25	0.29	0.01	0.01	0.31
Four spotted megrim	1.47	0.17	0.18	0.42	0.02	0.003	0.44
Megrim	1.22	0.25	0.04	0.35	0.04	0.002	0.39
Rosefish	5.25	0.13	0.55	1.50	0.02	0.02	1.54
Striped mullet	2.94	0.08	0.25	0.84	0.01	0.01	0.86
Conger	2.31	0.08	0.29	0.66	0.01	0.01	0.68
Yellow gurnard	0.92	0.08	0.08	0.26	0.01	0.003	0.27
Brown ray	1.89	0.13	0.13	0.54	0.02	0.005	0.57
Starry ray	1.64	0.08	0.08	0.47	0.01	0.003	0.48
Thornback ray	3.65	0.08	0.13	1.04	0.01	0.005	1.06
Cephalopod molluscs							
European squid	0.15	0.33	0.05	0.02	0.05	0.002	0.07
Broadtail squid	0.11	0.21	0.14	0.03	0.03	0.005	0.07
Curled octopus	0.58	0.29	0.06	0.17	0.04	0.002	0.21
Horned octopus	0.56	0.37	0.19	0.16	0.05	0.01	0.22
Spider octopus	0.63	0.67	0.10	0.18	0.10	0.004	0.28
Crustaceans							
Red shrimp	0.17	0.03	0.01	0.05	0.004	0.0002	0.05
Pink shrimp	0.18	0.01	0.01	0.05	0.002	0.001	0.05
Shrimp	0.90	0.18	0.01	0.06	0.004	0.0004	0.06

[a] Sum of individual metal THQs.

limits. Also for mercury, the calculated weekly intake was below the established limit either for mollusc cephalopods (Hg: 0.11–0.63 μg kg^{-1} b.w.) and crustaceans (0.17–0.90 μg kg^{-1} b.w.), but not for fish. As the figures indicated, the consumption of certain species, such as albacore (6.55 μg kg^{-1} b.w.) and rosefish (5.25 μg kg^{-1} b.w.), gave high levels in comparison to PTWI value, whereas weekly intakes below to safe level were associated with the consumption of the other fish (0.29–3.65 μg kg^{-1} b.w.), thought for some species (e.g. thornback ray: 3.65 μg kg^{-1} b.w.; striped mullet: 2.94 μg kg^{-1} b.w.; frostfish: 2.48 μg kg^{-1} b.w.; horse mackerel: 2.14 μg kg^{-1} b.w.) the estimated exposure was not to underestimate. A similar scenario is encountered with the target hazard quotients (THQs). As shown in Table 4 there were no THQ values for all metals over 1 through the consumption of either cephalopod molluscs (Hg: 0.02–0.18, Cd: 0.03–0.10, Pb: 0.002–0.01) or crustaceans (Hg: 0.05–0.06, Cd: 0.002–0.004, Pb: 0.0002–0.001), indicating health risk was absent. Analogously, the THQs of Cd (0.01–0.04) and Pb (0.002–0.18) from consumption of fish being less than 1, suggested that health risk was insignificant. Conversely, mercury THQs values, ranging from 0.08 to 1.87, were of concern. In particular, the health risk was mainly ascribed to consumption of albacore (1.87) and some benthic fish, such as rosefish (1.50) and thornback ray (1.04), thought also the TEQs values caused by consuming striped mullet (0.84), horse mackerel (0.82) and frostfish (0.71) were rather high being close to 1. Concerning dioxin-like PCBs, Table 5 shows the results of person weekly dietary intakes (μg/person/weekly) from each species and TEQ exposure (pg/person/weekly). In order to be able to estimate the contribution of the non- and mono-ortho PCBs to TEQs, their concentration were reported on a wet weight basis. The intake of dioxin-like PCBs from fish calculated at ND = 0, at ND = $\frac{1}{2}$LOD and ND = LOD was comprised between 0.01 and 4.42 TEQs/kg b.w., 1.38 and 4.73 TEQs/

Table 5
Non- and mono-*ortho* PCB concentrations (ng g^{-1} w.w.) and estimation of weekly TEQs (pg TEQs/b.w./weekly)

Species	Non-*ortho*			Mono-*ortho*			TEQs		
	PCB 77	PCB 126	PCB 169	PCB 105	PCB 118	PCB 156	(ND = 0)	(ND = $\frac{1}{2}$LOD)	(ND = LOD)
Fish									
Albacore	ND	ND	ND	0.59	0.58	ND	0.15	1.51	2.88
Horse mackerel	0.02	ND	ND	1.81	9.65	2.41	1.76	3.13	4.49
Mediterranean horse mackerel	0.01	ND	ND	ND	8.45	2.12	1.34	2.71	4.07
Pilchard	0.02	ND	ND	ND	0.82	ND	0.11	1.48	2.84
European anchovy	0.02	ND	ND	ND	0.21	ND	0.03	1.40	2.77
Frostfish	0.02	ND	ND	ND	0.20	ND	0.03	1.40	2.76
Hake	0.01	ND	ND	ND	0.26	ND	0.04	1.40	2.77
Greater forkbeard	0.02	ND	ND	ND	0.10	ND	0.02	1.39	2.75
Sea bream	ND	ND	ND	ND	0.07	ND	0.01	1.38	2.74
Four spotted megrim	ND	ND	ND	ND	0.12	ND	0.02	1.38	2.75
Megrim	0.02	ND	ND	0.06	0.20	0.20	0.07	1.43	2.80
Rosefish	0.02	ND	ND	0.09	0.71	0.55	0.18	1.54	2.91
Striped mullet	0.02	ND	ND	1.76	2.32	ND	0.52	1.89	3.25
Conger	0.02	0.01	ND	1.22	0.25	0.19	4.42	4.73	8.17
Yellow gurnard	ND	ND	ND	ND	0.18	ND	0.02	1.39	2.76
Brown ray	0.03	ND	ND	0.05	0.12	ND	0.03	1.40	2.76
Starry ray	0.01	ND	0.005	0.05	0.04	0.005	0.64	1.69	2.74
Thornback ray	0.04	ND	0.005	0.03	0.08	ND	0.66	1.71	3.76
Cephalopod molluscs									
European squid	0.005	ND	0.01	0.03	0.10	0.03	0.42	0.70	0.97
Broadtail squid	0.01	ND	ND	0.04	0.13	0.04	0.01	0.38	0.58
Curled octopus	0.03	ND	ND	0.08	0.18	0.02	0.01	0.38	0.75
Horned octopus	0.03	ND	ND	0.06	0.20	0.04	0.01	0.38	0.75
Spider octopus	ND	ND	ND	ND	0.05	ND	0.10	0.37	0.74
Crustaceans									
Red shrimp	0.01	ND	ND	0.08	0.09	ND	0.004	0.20	0.42
Pink shrimp	0.02	ND	0.01	ND	0.08	ND	0.27	0.42	0.58
Shrimp	0.01	ND	ND	ND	0.11	ND	0.003	0.20	0.41

ND = not detected.
LOD = limit of detection (5 pg g^{-1}).

kg b.w., and 2.74 and 8.17 TEQs/kg b.w., respectively. Intake values from molluscs cephalopods at ND = 0, at ND $= \frac{1}{2}$LOD and ND = LOD were between 0.01 and 0.42 TEQs/kg b.w., 0.37 and 0.70 TEQs/kg b.w. and 0.58 and 0.97 TEQs/kg b.w., respectively, while for crustaceans the estimates were between 0.003 and 0.27 TEQs/kg b.w. for ND = 0, 0.20 and 0.42 TEQs/kg b.w. for ND $= \frac{1}{2}$LOD and 0.41 and 0.58 TEQs/kg b.w. for ND = LOD. These estimates, resulting lower than 8 pg TEQs/kg b.w./weekly (Official Journal of the European Communities, 2006), seem to indicate that dietary consumption of these seafood does not implicate an appreciable human health risk.

4. Conclusions

Heavy metals (Hg, Pb and Cd) and polychlorinated biphenyls (PCBs) were determined in fish, cephalopods and crustaceans from Adriatic Sea (Mediterranean Sea). The considerable variation in levels of these contaminants among the different species, highlights the important role of ecological and physiological factors in concentrating pollutants. From the human health point of view, Cd and Pb THQ values (<1) show a situation of no risk for the consumer. In contrast, analysis of mercury data suggests that dietary consumption of certain fish species can vary this neurotoxin intake substantially determining big differences in health risks. Concerning dioxin-like PCBs, the estimated TEQ values seem to indicate that consumption of these fish does not implicate an appreciable human health risk. Nevertheless, it must be remembered either that the limit value of 8 pg TEQs/kg b.w./weekly set by WHO, includes 12 dioxin-like PCBs, PCDDs and PCDFs, or estimated intake does not take into account exposure from other foods. Consequently, intake might be significantly underestimated and might be of concern, above all in the cases where the exposure is closer to the tolerable weekly intake. As a final conclusion, we suggest that more specific recommendations regarding human consumption (kind of species and frequency and size of meals) are done according to the data concerning levels of environmental pollutants in the most consumed fish and seafood species.

Conflict of interest statement

The authors declare that there are no conflicts of interest.

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