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Human exposure to lead, cadmium and mercury through fish and seafood product consumption in Italy: a pilot evaluation

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The presence of selected toxic heavy metals, such as cadmium (Cd), lead (Pb) and mercury (Hg), was investigated in fish and seafood products, namely, blue mussel, carpet shell clam, European squid, veined squid, deep-water rose shrimp, red mullet, European seabass, gilthead seabream, Atlantic cod, European hake, Atlantic bluefin tuna and swordfish so as to assess their human exposure through diet. Metals were detected by quadrupole inductively coupled plasma mass spectrometry (Q-ICP-MS) and hydride generation atomic absorption spectrometry (Hg-AAS). Measurements of Cd, Pb and Hg were performed by means of analytical methods validated in compliance with UNI CEI EN ISO/IEC 17025 [2005. General requirements for the competence of testing and calibration laboratories. Milano (Italy): UNI Ente Nazionale Italiano di Unificazione]. The exposure assessment was undertaken matching the levels of Cd, Pb and total Hg with consumption data related to fish and seafood products selected for this purpose. In order to establish human health implications, the estimated weekly intakes (EWIs) for Cd, Pb and Hg were compared with the standard tolerable weekly intakes (TWI) for Cd and provisional tolerable weekly intakes (PTWIs) for Pb and Hg stipulated by the European Food Safety Authority (EFSA) and the Food and Agriculture Organization/World Health Organization (FAO/WHO) Joint Expert Committee on Food Additives (JECFA). The found metal concentrations were largely below the maximum levels (MLs) established at the European Union level with the exception of Cd. This metal exceeded the MLs in squid, red mullet, European hake and Atlantic cod. Squid and blue mussel showed the highest Pb concentrations which accounted for 60% and 10% of the MLs, respectively. Highest Hg levels were found in predatory fish. The concentrations of Hg in swordfish, Atlantic bluefin tuna and red mullet accounted for 50%, 30% and 30% of the MLs, respectively. The EWIs for Cd, Pb and Hg related to the consumption of fish and seafood products by the median of the Italian total population accounted for 20%, 1.5% and 10% of the standard TWI for Cd as well as PTWIs for Pb and Hg, respectively. Furthermore, the EWIs estimated using consumption data concerning Italian consumers did not exceed the standard TWI and PTWIs, except for Cd at 95th percentile.

Keywords: fish; seafood products; heavy metals; tolerable weekly intake; provisional tolerable weekly intake

Introduction

Fish and seafood products constitute a significant and healthy part of the human diet as they are an important source of easily digestible proteins, vitamins, especially vitamins D, A and B_{12}, and minerals such as iodine and selenium. In addition, fish are particularly rich in unsaturated essential fatty acids (PUFAs), especially omega-3 PUFAs that play an important role in human health (European Food Safety Authority (EFSA) 2005). Several scientific reports indicate that omega-3 PUFAs can beneficially affect the cardiovascular system by reducing the incidence of cardiac events as well as decreasing the progression of atherosclerosis (Kris-Etherton et al. 2002). Moreover, omega-3 PUFAs are found to be essential for the development of the central nervous system during foetal and neonatal period (Clandinin et al. 1989; Martinez 1992; Clandinin 1999). On the other hand, there is a safety concern related to the consumption of fish and seafood products, that is, the presence of a wide variety of chemical contaminants, i.e. heavy metals. Due to the toxicological effects of heavy metals, the European Union established maximum levels (MLs) for cadmium (Cd), lead (Pb) and mercury (Hg) in fish and seafood products (Council of the European Union 2006). MLs for Cd are set from 50 to 300 $\mu g\,kg^{-1}$ in relation to fish species. On the other hand, an ML value of 1000 $\mu g\,kg^{-1}$ is established for bivalve molluscs and cephalopods as well as 500 $\mu g\,kg^{-1}$ for crustaceans. The MLs for Pb are 300, 1500, 1000 and 500 $\mu g\,kg^{-1}$ for fish, bivalve molluscs, cephalopods and crustaceans, respectively. As far as Hg is concerned, an ML of 50 $\mu g\,kg^{-1}$ is established for fish species and seafood products except for some predatory fish. The highest

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ML for predatory fish (1000 $\mu g\,kg^{-1}$) reflects the tendency of this metal to build up in such specimens, largely as methylmercury (methyl-Hg), the chemical form of most concern from the toxicological point of view. Furthermore, tolerable weekly intakes (TWI) and provisional tolerable weekly intakes (PTWIs) are stipulated by the European Food Safety Authority (EFSA) and the Food and Agriculture Organization/ World Health Organization (FAO/WHO) Joint Expert Committee on Food Additives (JECFA) for Cd, Pb and Hg. A TWI of 2.5 $\mu g\,kg^{-1}$ body weight (b.w.) is set for Cd by EFSA in replacement of 7 $\mu g\,kg^{-1}$ b.w. previously established by JECFA (EFSA 2009). Regarding Pb, the JECFA stipulated and reconfirmed in 1986 and 1999, respectively, a PTWI of 25 $\mu g\,kg^{-1}$ b.w. (WHO 1986, 2000). Recently, an update risk assessment on Pb by both the EFSA and JECFA highlighted that 25 $\mu g\,kg^{-1}$ b.w. is not longer considered health protective, particularly in some population groups such as children (EFSA 2010; WHO 2011). As far as Hg is concerned, the JECFA set PTWI values of 5 and 1.6 $\mu g\,kg^{-1}$ b.w. for total-Hg and methyl-Hg, respectively (WHO 2004). The EFSA Panel on Contaminants in the Food Chain focuses on vulnerable groups such as women of childbearing age, pregnant and breastfeeding women, as well as young children. In order to minimise the risk related to methyl-Hg accumulation to toxic level, the EFSA recommends the above-mentioned groups to include a wide range of fish species in their diet as well as to reduce the predatory fish consumption, e.g. swordfish and tuna (EFSA 2004a, 2004b).

The aim of this study was to evaluate the presence of Cd, Pb and Hg in fish and seafood products so as to assess their human exposure through diet. The determination of the above-reported metals was undertaken in the following specimens widely consumed by the Italian population: blue mussel (*Mytilus galloprovincialis*, Lamark 1819), carpet shell clam (*Tapes decussates*, Linnaeus 1758), two squid species typical of Mediterranean sea, namely, European squid (*Loligo vulgaris*, Lamark 1798) and veined squid (*Loligo forbesi*, Steenstrup 1856), deep-water rose shrimp (*Parapenaeus* longirostris, Lucas 1846), red mullet (*Mullus surmuletus*, Linnaeus 1758), European seabass (*Dicentrarchus labrax*, Linnaeus 1758), gilthead seabream (*Sparus orata*, Linnaeus 1758), Atlantic cod (*Gadus morhua*, Linnaeus 1758), European hake (*Merluccius merluccius*, Linnaeus 1758), Atlantic bluefin tuna (*Thunnus thynnus*, Linnaeus, 1758) and swordfish (*Xiphias gladius*, Linnaeus 1758) (see http://www.fishbase.org and http://www.fao.org/fishery/en). The exposure assessment was carried out by matching the levels of Cd, Pb and total Hg with consumption data related to marine species selected for this purpose. The intake values for Cd, Pb and Hg were expressed for the median of the total population, median and 95th percentile of consumers in Italy using consumption data obtained by the National Institute for Food and Nutrition Research (INRAN) (Leclercq et al. 2009). In order to establish human health implications, the estimated weekly intakes (EWIs) for Cd, Pb and Hg were compared with those standard TWI for Cd as well as PTWIs for Pb and Hg established by EFSA and JECFA (WHO 1986, 2000, 2004; EFSA 2009). As far as Hg was concerned, being methyl-Hg the predominant form of Hg in fish and seafood products a PTWI value of 1.6 $\mu g\,kg^{-1}$ b.w. was also considered for this purpose.

Materials and methods

Sampling and pre-treatment

A total of 115 samples of blue mussel, carpet shell clam, deep-water rose shrimp, European squid, veined squid, red mullet, European seabass, gilthead seabream, Atlantic cod, European hake, Atlantic bluefin tuna and swordfish were purchased from different retail outlets in Italy. Information on the investigated species as well as the number of individual samples are listed in Table 2.

In order to reduce the risk of any possible exogenous contamination by the metals under study, the manipulation and pre-treatment of samples were always performed in a Class-100 clean room (Tamco, Rome, Italy). Frozen samples were thawed out at room temperature and then cleaned by rinsing in deionised water. The subsequent dissection of each sample was performed by means of free metal equipment. According to Commission Regulation 2006/1881/EC only edible sample tissues were taken into account for metal determination (Council of the European Union 2006). Specimens were subsequently homogenised by a HMHF Turbo Homogenizer (PBI International, Milano, Italy) and finally mineralised by means of acid-assisted microwave digestion (Milestone Ethos Plus, Sorisole (Bg), Italy). Sampling and preparation were executed in compliance with UNI EN 13804 (2002). As far as microwave-assisted mineralisation was concerned, UNI EN 13805 (2002) was applied for this purpose.

All chemicals used during the analytical procedure were of ultrapure grade (HNO_3, H_2O_2; Carlo Erba-Rodano, Milan, Italy) and all solutions were prepared using deionised Milli-Q water (Millipore, Bedford, MA, USA). Calibrants and internal standard solutions (rhodium) were daily obtained from standard certified solutions with a content of 1 $mg\,ml^{-1}$ of all elements (Carlo Erba-Rodano), followed by dilution with acidified (HNO_3) deionised water as necessary.

Analytical determination

The determination of Cd and Pb was carried out by quadrupole inductively coupled plasma mass spectrometry (Q-ICP-MS) using an Elan 6000 spectrometer (Perkin-Elmer, Norwalk, CT, USA) equipped with an ultrasonic nebuliser (U-5000 AT*, CETAC Technologies, Omaha, Nebraska, USA) and a cyclonic quartz spray chamber. The Q-ICP-MS system was set up as follows: RF power, 1100 W; lens voltage ranging from 7 to 8 V; nebuliser gas flow ranging from 0.97 to 1.01 $l\,min^{-1}$; peristaltic pump speed, 1.0 $ml\,min^{-1}$. The following analytical masses $_{112}Cd$, $_{114}Cd$, $_{207}Pb$ and $_{208}Pb$ were selected for quantification. The internal standardisation by $_{103}Rh$ at a concentration of 2 $\mu g\,l^{-1}$ in the analytical solutions was used to control for drifts of the instrument and matrix effects.

Hg determination was obtained by flow injection-cold vapour atomic-absorption spectrometry (FI-CV-AAS) using an FIAS 400 flow injection mercury system (Perkin-Elmer). $NaBH_4$ 0.2% w/v from Merck (Darmstadt, Germany) and ultrapure HCl 3% v/v from Fluka (Poole, Dorset, UK) were used as the reductant and carrier, respectively. Measurements were performed at a wavelength of 253.7 nm.

Several Certified Reference Materials (CRMs) such as BCR CRM 422 Cod Muscle (IRMM, Belgium), BCR 668 Mussel Tissue (IRMM, Belgium), DORM-3 Fish Protein (NRC, Canada) and DOLT-3 Dogfish Liver (NRC, Canada) were used to check the accuracy of analytical measurements.

Results and discussion

Method validation

Both analytical methods were validated in compliance with UNI CEI EN ISO/IEC 17025 (2005). According to Annex III of Commission Regulation 2004/882/EC (Council of the European Union 2004) on official controls, the following parameters were taken into account for method validation: limit of detection (LOD), limit of quantification (LOQ), precision, trueness and uncertainty. Furthermore, in relation to Commission Regulation 2007/333/EC (Council of the European Union 2007), the validation parameters need to be characterised by performance criteria which are listed hereinafter. In detail, LOD should be lower than one-tenth of the ML set by Commission Regulation 2006/1881/EC (Council of the European Union 2006) with the exception of Pb for which one-fifth of the ML is required in case of an ML value less than 100 $\mu g\,kg^{-1}$. As regards LOQ, a value less than one-fifth of the ML is established for this validation parameter. On the other hand, for an ML of Pb less than 100 $\mu g\,kg^{-1}$, a value less than two-fifths of the ML is required. The precision parameter was obtained by method repeatability considering as reference an HORRATr value less than 2 (Horwitz and Albert 2006) and method trueness was assessed by including in metal analysis different CRMs. As far as uncertainty was concerned, it was estimated by means of a holistic approach and compared with maximum standard measurement uncertainty (*Uf*) reported in the "fitness-for-purpose" approach section of Commission Regulation 2007/333/EC (Council of the European Union 2007).

Methods were periodically tested by involvement to proficiency tests (PTs) organised by European Union Reference Laboratory for Chemical Elements in Food of Animal Origin (EU-RL CEFAO) and the Joint Research Centre–Institute for Reference Materials and Measurements (JRC-IRMM) in compliance with Commission Regulation 2004/882/EC (Council of the European Union 2004). The results obtained by inter-laboratory comparisons always showed satisfactory *z*-scores so as to establish the reliability of analytical procedures (International Organization for Standardization (ISO) 2005; Thompson and Ellison 2006). Table 1 summarises validation parameters, performance criteria as well as PT results.

Cd, Pb and Hg levels in fish and seafood products

Cd, Pb and Hg concentrations are summarised in Table 2. Data grouped by species are expressed as the mean and median for each investigated metal.

In general terms, heavy metal concentrations varied widely among different organisms, indicating a species-specific bioaccumulation process. Metal concentrations ($\mu g\,kg^{-1}$ wet weight) were largely below the MLs established at the European Union level with the exception of Cd. In fact, this metal exceeded the MLs in some samples of squid, red mullet, European hake and Atlantic cod.

Concerning Cd, it reached the highest levels in squid species and blue mussel. Mean concentrations were 728 and 616 $\mu g\,kg^{-1}$ for squid species and blue mussel, respectively. Cd concentrations in squid species and blue mussel amounted to 60% and 70% of the MLs established for these seafood species, respectively. Moreover, carpet shell clam showed a Cd concentration of 159 $\mu g\,kg^{-1}$. The levels of Cd in Atlantic cod and European hake ranged from a negligible concentration lower than the LOQ to 57 $\mu g\,kg^{-1}$, pointing out the high variability related to Cd bioaccumulation in such fish species. Other analysed fish species and shrimp mostly showed low Cd concentrations. The results on Cd bioaccumulation in specimens of bivalve and cephalopod molluscs matched previous findings (Coni et al. 1992; Bustamante, Caurant, et al. 1998; Bustamante, Cherel, et al. 1998; Jureša and Blanuša 2003; Rubio et al. 2006; Storelli 2008). Bustamante et al. (2002) reported the ability of cephalopods to concentrate Cd in the digestive gland which is involved in storage and detoxification of this metal.

As shown in Table 2, the Pb content was generally low. The concentrations of this metal in fish specimens

Table 1. Method validation parameters.

		LOD (µg Kg^{-1})		LOQ (mg Kg^{-1})		Precision		Trueness (%)		Proficiency test (*z*-score)		Uncertainty $U(X)$ $K=2$; 95	
Heavy metals	Reference ML (mg kg^{-1} wet weight)	Value	Performance criteria (2007/333/EC)	Value	Performance criteria (2007/333/EC)	Value	Performance criteria (2007/333/EC)	Value	Performance criteria (2007/333/EC)	Value[a]	Performance criteria (2007/333/EC)	Value	Performance criteria (2007/333/EC)
Cd	0.050 (3.2.1–3.2.5 –3.2.14)	0.01	≤ 5	0.03	≤10	12%	< 15%	90	70–120%	0.27; 0.18; 0.20; –0.20; 0.30	< 2	13%	< *Uf* (0.050 mg kg^{-1}) 37%
Pb	0.020 (3.1.1–3.1.2)	0.20	≤4	0.48	≤8	12%	< 15%	104	70–120%	1.00; 0.25; 0.40; –0.50; 0.50	< 2	13%	< *Uf* (0.020 mg kg^{-1}) 45%)
Hg	0.50 (3.3.1)	0.010	≤50	0.02	≤100	7%	≤10%	108	70–120%	0.80; 0.16; 0.30; 0.10; 0.20	< 2	12%	< *Uf* (0.50 mg kg^{-1}) 37%

Note: [a]Proficiency tests were performed in 2005, 2007, 2009, 2010 and 2011 for each analysed metal.

Table 2. Cd, Pb and Hg levels in fish and seafood products (μg kg^{-1} wet weight).

Species (number of samples)	Cd		Pb		Hg	
	Mean ± SD	Median	Mean ± SD	Median	Mean ± SD	Median
Bivalve molluscs						
Blue mussel (11)	616 ± 272	582	166 ± 58	131	20 ± 16	20
Carpet shell clam (11)	159 ± 84	142	64 ± 25	48	69 ± 39	67
Cephalopod molluscs						
European and veined squid (11)	728 ± 531	691	586 ± 301	587	29 ± 21	29
Crustaceans						
Deep-water rose shrimp (10)	14 ± 9	14	31 ± 19	29	50 ± 28	51
Fish						
Red mullet (10)	53 ± 27	57	52 ± 26	49	311 ± 204	325
European seabass (11)	17 ± 8	21	45 ± 28	43	77 ± 36	76
Gilthead seabream (10)	15 ± 10	14	56 ± 32	60	81 ± 43	82
Atlantic cod (10)	< 0.03	< 0.03	17 ± 11	15	67 ± 31	63
European hake (11)	< 0.03	< 0.03	31 ± 18	22	67 ± 38	64
Atlantic bluefin tuna (10)	46 ± 28	41	61 ± 46	53	316 ± 175	302
Swordfish (10)	59 ± 34	61	66 ± 47	64	529 ± 253	502

were lower than 100 μg kg^{-1}. Pb-negligible levels were also found in carpet shell clam and shrimp samples. On the other hand, squid species and blue mussel showed the highest Pb concentrations, that is, 586 and 166 μg kg^{-1}, respectively. The metal concentrations in squid species and blue mussel accounted for 60% and 10% of the MLs set for these marine specimens, respectively. Previous studies on Pb bioaccumulation in marine fish species highlighted comparable concentrations with respect to those found in this survey (Jureša and Blanuša 2003; Storelli 2008). In contrast, the literature data on Pb bioaccumulation in seafood products such as squid species showed marked differences, that is, lower Pb levels with respect to those checked in our investigation (Falcó et al. 2006; Storelli 2008). As far as bivalve molluscs were concerned, blue mussel showed higher Pb concentrations than carpet shell clam. Jureša and Blanuša (2003) estimated Pb concentrations in bivalve species in good agreement with respect to our data on blue mussel. Falcó et al. (2006) also found levels of Pb in mussel and clam specimens both comparable with blue mussel and carpet shell clam monitored in our laboratory.

The results obtained for Hg confirmed the literature data regarding the presence of the highest Hg levels in predatory fish. It is to be noted that this metal, mainly present in fish and seafood products as methyl-Hg, undergoes a biomagnification process along the food chain (Storelli at al. 2002, 2003; Storelli 2008). Furthermore, other factors such as living habits may affect Hg bioaccumulation in marine organisms, that is, the close association with sediments and, consequently, bottom feeding habits (Storelli at al. 2002, 2005). The Hg concentrations in swordfish, Atlantic bluefin tuna and red mullet accounted for 50%, 30% and 30% of the MLs established for these fish species, respectively. On the other hand, the percentages of Hg measured in the other investigated fish species as well as seafood products were approximately 15% and 8% of the European Union MLs, respectively.

Estimates of Cd, Pb and Hg intake

Table 3 shows the EWIs of Cd, Pb and Hg expressed for the median of the total population, median and 95th percentile of consumers in Italy along with their percentage with respect to standard TWI and PTWIs. In general terms, the EWIs obtained taking into account fish and seafood product consumption by total population and consumers did not exceed the standard TWI and PTWIs recommended by EFSA and JEFCA for Cd, Pb and Hg. In contrast, the EWI found considering the 95th percentile of Italian consumers exceeded 5% of the standard TWI stipulated by EFSA for Cd.

Concerning Cd, the EWI for this metal through fish and seafood product consumption by the total population was 0.51 μg $week^{-1}$ for a subject weighing 65 kg. This value accounted approximately for 20% of the recent TWI established by EFSA. Cd EWIs comparable with that found in this study have been previously reported by other authors (Martorell et al. 2001; Llobet et al. 2003; Rubio et al. 2006; Cirillo et al. 2010). An higher estimated weekly intake (approximately 40% of the standard TWI) was noticed by Muñoz et al. (2005) for fish and shellfish. On the other hand, Falcó et al. (2006) and Karavoltsos et al. (2003) reported lower EWI values, that is, roughly 4% of the EFSA TWI. As shown above in this survey, the highest levels

Table 3. Estimated weekly intakes (EWIs) for Cd, Pb and Hg for the total population.

Cd			Pb			Hg		
Fish and seafood products intake (g day^{-1}) by the median of the total population	EWI (µg $week^{-1}$) b.w.	% TWI	Fish and seafood products intake (g day^{-1})	EWI (µg $week^{-1}$) b.w.	% PTWI	Fish and seafood products intake (g day^{-1})	EWI (µg $week^{-1}$) b.w.	% PTWI
32.2	0.51[a]	21	32.2	0.35[a]	1.4	32.2	0.50[a]	10
Fish and seafood products intake (g day^{-1}) by the median of consumers	EWI (µg $week^{-1}$) b.w.	% TWI	Fish and seafood products intake (g day^{-1})	EWI (µg $week^{-1}$) b.w.	% PTWI	Fish and seafood products intake (g day^{-1})	EWI (µg $week^{-1}$) b.w.	% PTWI
57.2	0.9[a]	36	57.2	0.60[a]	2.5	57.2	0.90[a]	18
Fish and seafood products intake (g day^{-1}) by the 95th percentile of consumers	EWI (µg $week^{-1}$) b.w.	% TWI	Fish and seafood products intake (g day^{-1})	EWI (µg $week^{-1}$) b.w.	% PTWI	Fish and seafood products intake (g day^{-1})	EWI (µg $week^{-1}$) b.w.	% PTWI
165.1	2.6[a]	105	165.1	1.8[a]	7	165.1	2.6[a]	50

Note: [a]Estimated weekly intake for a subjet of 65 kg.

of this metal were found in cephalopod and bivalve molluscs. In order to assess Cd exposure related to cephalopod and bivalve consumption, the EWI was also obtained assuming that these marine organisms were the only contribution to the fish and seafood product category. In this case the estimated weekly intake reached approximately 65% of the 2.5 μg kg^{-1} established by EFSA. Similar results were previously obtained by Storelli et al. (2006). Taking into account each cephalopod species analysed by Storelli and her team, the EWI accounted approximately for 70% of the recent TWI stipulated by EFSA. Moreover, the EWIs were also quantified by considering consumption data related to both the median and 95th percentile of Italian consumers. The found values were 0.9 and 2.6 μg week^{-1} corresponding to 36% and 105% of the EFSA TWI, respectively. This exposure scenario is noteworthy especially considering other great contributors to dietary Cd intake, namely cereals and cereal products, tubercles and vegetables (Martorell et al. 2001; Karavoltsos et al. 2003; Llobet et al. 2003; Rubio et al. 2006; EFSA 2012). In addition, several factors, i.e., nutritional status, multiple pregnancies and diseases which can increase Cd dietary absorption, highlight the existence of vulnerable population groups (EFSA 2009).

As far as Pb was concerned, the estimated weekly intake for this metal through fish and seafood product consumption was 0.35, 0.60 and 1.8 μg week^{-1} for the total population, median and 95th percentile of consumers, respectively. The obtained EWIs account for 1.5%, 2.5% and 7%, respectively, of the standard PTWI stipulated by JEFCA. Previous surveys highlighted comparable results for fish and seafood products (EWIs from 0.5% to 2% of the standard PTWI) (Martorell et al. 2001; Llobet et al. 2003; Falcó et al. 2006; Cirillo et al. 2010). As for Cd, the results showed that the consumption of cephalopod and bivalve molluscs increased the dietary intake of Pb. The exposure assessment related to a possible update of the standard PTWI of this metal, as indicated by EFSA and JECFA, might corroborate the importance of data concerning cephalopod and bivalve consumption. Of further need is the evaluation of Pb levels in cereals and cereal products, vegetables as well as tap water which have been described by EFSA as major contributors to Pb exposure (EFSA 2010).

With respect to Hg, the found EWI for this metal through the consumption of fish and seafood products by the total population was 0.50 μg week^{-1} corresponding to 10% of the standard PTWI set by JECFA for total Hg. The EWIs reached approximately 18% and 50% of the standard PTWI matching the level of total Hg with the consumption data concerning the median and 95th percentile of Italian consumers. As previously reported, being methyl-Hg the predominant form of Hg in fish and seafood products, a standard PTWI value of 1.6 μg kg^{-1} b.w. has been also considered for risk assessment (Storelli at al. 2002, 2003; EFSA 2004a, 2004b). In the case of the total population and median of consumers, the found EWIs were 0.50 and 0.88 μg week^{-1}. These results accounted approximately for 30% and 55% of the standard PTWI established for methyl-Hg by JECFA. In addition to the EWIs obtained considering the total population and median of consumers, a further EWI for methyl-Hg was estimated for the 95th percentile of consumers so as to achieve an exposure assessment related to Italian high consumers of fish and seafood products. In this case the EWI exceeded approximately 50% of the standard PTWI stipulated by JECFA for methyl-Hg. Previous literature revealed a comparable range of EWIs with respect to that found in this study for the total population and median consumers in Italy. In detail, Cirillo et al. (2010) noticed lower results with respect to that found in this study for Italian median consumers (approximately 30% of the standard PTWI for methyl-Hg). A lower percentage with respect to the standard PTWI, that is, 10% for fish and shellfish, was also found by Muñoz et al. (2005) for a population in Santiago, Chile. Martorell et al. (2001) noticed roughly 40% of the JECFA PTWI for methyl-Hg concerning fish and seafood product consumption by male adults in Catalonia, Spain. The percentage of the standard PTWI reported by Llobet et al. (2003) ranged from 33% to 60% in relation to different population groups for fish and shellfish. Falcó et al. (2006) reported comparable values concerning the consumption of fish and seafood products by several groups in the population, that is, from 35% to 65% of the standard PTWI. Fish and seafood products are the major source of Hg intake in humans and, among this food category, predatory and benthic fish showed the highest levels of this metal (mainly present as methyl-Hg) (Storelli at al. 2002, 2003; EFSA 2004a; Storelli 2008). Consequently, the reduction of this fish species consumption should be encouraged in Italian high consumers, such as those from coastal regions as well as vulnerable groups, namely women of childbearing age, pregnant and breastfeeding women, and young children considered by EFSA to be at major health potential risk for methyl-Hg (EFSA 2004a, 2004b).

In conclusion, the results of this survey point out that exposure through the consumption of certain fish and seafood products can be a problem mainly in population groups at high risk of exposure and vulnerability. Further investigations are required especially in relation to continuous updates of standard TWIs on heavy metals by EFSA and FAO/WHO JECFA.

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