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Original Research Article

Human exposure in Italy to lead, cadmium and mercury through fish and seafood product consumption from Eastern Central Atlantic Fishing Area

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ABSTRACT

The presence of cadmium (Cd), lead (Pb) and mercury (Hg) was investigated in fish and seafood products collected from the FAO Major Fishing Area 34, Eastern Central Atlantic. Samples were purchased from different retail outlets in Italy. Samples were selected so as to assess human exposure through diet. Metals were detected by Q-ICP-MS and Hg-AAS. All the metal concentrations detected were largely below the maximum levels (MLs) established by the European Union. The exposure assessment was undertaken by matching the concentration of Cd, Pb and total Hg in fish and other seafood products selected purposed according to Italian consumption data. The estimated weekly intakes (EWIs) for the evaluated elements related to the consumption of fish and other seafood products by the median of the Italian total population accounted for 14%, 2% and 14% of the standard tolerable weekly intake (TWI) for Cd and Hg as well as the provisional tolerable weekly intake (PTWI) for Pb, respectively.

Q2 1. Introduction

Q3 Over the last few decades, there has been growing interest in determining heavy metal levels in the marine environment and attention was drawn to the measurement of contamination levels in public food supplies, particularly fish (Tarik et al., 1993; Rose Q4 et al., 1999; Yousuf and El-Shahawi, 1999). With the exception of occupational exposure, fish are acknowledged to be the single largest source of Hg for man.

In some instances, fish catches were banned for human consumption because total Hg content of some species exceeded the maximum limits recommended by the FAO-WHO (Voegborlo et al., 1999). The likelihood of Hg toxicity from fish consumption has been identified in other parts of the world (Inkship and Piotrowski, 1985; Rose et al., 1999).

Heavy metals can be bioaccumulated and biomagnified via the food chain and finally assimilated by human consumers resulting

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in health hazards (Abdel-Baki et al., 2011). As a consequence, fish are often used as indicators of heavy metals contamination in the aquatic ecosystem because they occupy high trophic levels and are an important food source (Blasco et al., 1998; Agah et al., 2010). The bioaccumulation of heavy metals in living organisms describes the processes and pathways of pollutants from one trophic level to another. Various species of fish are mostly used as bio-indicators of heavy metals contamination (Drastichová et al., 2004).

Concerning heavy metals, due to the related toxicological effects, the European Union established several maximum levels (MLs) for Cd, Pb and Hg in fish and other seafood products (Council of the European Union, 2006). MLs for Cd are set from 50 to 250 μg kg^{-1} in relation to fish species. On the other hand, an ML value of 1000 μg kg^{-1} is established for bivalve molluscs and cephalopods as well as 500 μg kg^{-1} for crustaceans. The MLs for Pb are 300, 1500, 1000 and 500 μg kg^{-1} for fish, bivalve molluscs, cephalopods and crustaceans, respectively. As far as Hg is concerned, an ML of 500 μg kg^{-1} is established for fish species and other seafood products except for some predatory fish. The highest ML for predatory fish (1000 μg kg^{-1}) reflects the tendency of this metal to build up in such specimens, largely as methyl-mercury (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 μg kg^{-1} body weight (b.w.) is set for Cd by EFSA in replacement of 7 μg 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 μg kg^{-1} 1 b.w. (WHO, 1986, 2000). Recently, an update risk assessment on Pb by both the EFSA and JECFA highlighted that 25 μg kg^{-1} b.w. is no longer considered health protective, particularly in some population groups such as children (EFSA, 2010; WHO, 2011a,b). In line with JECFA, the EFSA set a provisional tolerable weekly intakes for methyl-Hg of 1.3 μg kg^{-1} body weight (b.w.) and of 4 μg kg^{-1} b.w. for inorganic Hg (WHO, 2011a,b; EFSA, 2012a). 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,b, 2012a).

The aim of this study was to evaluate the presence of Cd, Pb and Hg in fish and other seafood products so as to assess dietary exposure to these toxic elements. The determination of the above-reported metals was undertaken in fish available for consumption by the Italian population and were purchased from different retail outlets in Italy, selecting samples collected from the FAO Major Fishing Area 34 (Eastern Central Atlantic, see http://www.fishbase.org and http://www.fao.org/fishery/en). As for many developing nations, fish trade represents a significant source of foreign currency earnings, in addition to the sector's important role as a generator of income, source of employment, and provider of food security and nutrition the fishing area 34 has been selected. 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 other seafood products, a TWI value of 1.3 mg kg^{-1} b.w. was also considered for this purpose.

2. Methods

2.1. Sampling and sample preparation

Four hundred and fifty samples were purchased from different retail outlets in Italy, selecting samples collected from the FAO Major Fishing Area 34. FAO Major Fishing Areas for Statistical Purposes are arbitrary areas, the boundaries of which were determined in consultation with international fishery agencies on various considerations, including (i) the boundary of natural regions and the natural divisions of oceans and seas; (ii) the boundaries of adjacent statistical fisheries bodies already established in inter-governmental conventions and treaties; (iii) existing national practices; (iv) national boundaries; (v) the longitude and latitude grid system; (vi) the distribution of the aquatic fauna; and (vii) the distribution of the resources and the environmental conditions within an area (FAO, 2002). Selected pooled (ten specimens per sample) samples were: deep-water rose shrimp (*parapenaeus longirostris*, Lucas 1846, 90 specimens), tilapia heudelotii (*sarotherodon melanotheron heudelotii, Duméril*, 1861, 90 specimens), gilthead seabream (*sparus orata*, Linnaeus 1758, 90 specimens), European seabass (*dicentrarchus labrax*, Linnaeus 1758, 90 specimens), swordfish (*xiphias gladius*, Linnaeus 1758, 30 specimens), European squid (*loligo vulgaris*, Lamark 1798, 30 specimens), silver scabbardfish (*lepidopus caudatus*, Euphrasen 1788, 30 specimens). After collection, specimens were sealed in decontaminated polyethylene bags, frozen at −20 °C and stored at the same temperature until delivery to the ISS. Frozen samples were thawed out at room temperature and then were cleaned by rinsing in deionised water. The subsequent dissection of each sample was performed by means of common free metal equipment (scalpels, scissors and stainless steel microspoon, VWR International PBI S.r.l. Via San Giusto, 85-20153 Milano, Italy). According to Commission Regulation EC No. 1881/2006, only edible sample tissues were taken into account for metal determination. Specimens were subsequently homogenised by a HMHF Turbo Homogeniser (PBI International,) and finally mineralised by means of acid assisted microwave digestion (Milestone Ethos Plus S.r.l., Via Fatebenefratelli, 5, Sorisole Bergamo, Italy). Sampling, preparation as well as acid-assisted microwave digestion of samples were performed in compliance with EN 13804 (2013) and EN 13805 (2002), respectively. In order to reduce the risk of any possible exogenous contamination by the metals under test manipulation and sample preparation were always performed in a Class-100 clean room (T.Am.Cco S.r.l., Rome, Via Lugnano in Teverina 20 00100 Rome, Italy). All of the chemicals used during the analytical procedure were of ultrapure grade (HNO_3 65%, H_2O_2 30%, CARLO ERBA Reagents S.r.l. via Raffaele Merendi 22, 20010 Cornaredo Milan, Italy) and all solutions were prepared using deionised Milli Q water (Resistivity = 18 MΩ, Merck Millipore S.p.a., Via XI Febbraio 99, 20090 Vimodrone Milano, Italy). 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.

2.2. Analytical determinations

The determination of Cd and Pb have been carried out by Quadrupole Inductively Coupled Plasma Mass Spectrometry (Q-ICP-MS) using an Elan 6000 spectrometer (Perkin Elmer Italia

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