



# Evaluation of mercury, cadmium and lead levels in fish and fishery products imported by air in North Italy from extra-European Union Countries



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## ABSTRACT

The levels of cadmium (Cd), mercury (Hg) and lead (Pb) were evaluated in 251 samples of fish and fishery products randomly collected between January 2010 and December 2013 from products imported through the airport of Milan Malpensa from extra-European Union (extra-EU) Countries and inspected at the Border Inspection Post by official public veterinarians. In this study, products were classified in four categories: predatory fish (larger predatory fishes like tunafish and swordfish, and spiny dogfish), non-predatory fish (other species of fishes of lower dimensions and different feeding behavior, mollusc (cephalopods and bivalves) and crustacean). Analyses were performed by accredited laboratory. All the concentrations were below the levels expressed in Regulation (EC) No 1881/2006 except for two samples of swordfish that exceeded in Hg content. Predatory fish had a significantly higher content of Hg (median value 375  $\mu\text{g kg}^{-1}$ ) while mollusc had a significantly higher content of Cd (median value 66.5  $\mu\text{g kg}^{-1}$ ) compared to the other groups of seafood included in this study. No relationship was found between the individual concentration of metals.

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## 1. Introduction

Fish and seafood consumption increased in the recent years (El Mosehly, Othman, Abd El-Azem, & El-Metwally, 2014) and plays a key-role in human diet all over the world; they are one of the best sources of high-quality protein and of other nutrients such as selenium, antioxidants and essential fatty acids of the  $\omega 3$  series (Aydin & Faruk, 2013; Duran, Tuzen, & Soyak, 2014). Meanwhile, in the last few decades the worldwide sensibility for human health related to environmental contamination is increasing considerably (Conti et al., 2012); many chemical compounds generated by human activities are released into the environment, first of all in water and soil, accumulating and causing even severe damages in living

organisms (Conti & Cecchetti, 2001). These compounds could be transferred by the food chain to human beings (Duran, Tuzen, & Soyak, 2014) representing a potentially harmful element, in particular for some population groups such as children and pregnant women, and causing even severe health disorders (Barrento et al., 2008); the severity of adverse health effects is related to the type of heavy metal and its chemical form, and is time- and dose-dependent (Tchounwou, Yedjou, Patlolla, & Sutton, 2012). For this reason, analyses of contaminants content in fishery products is one of the most important activities to take into consideration when controlling their safety. With this purpose, in European Union the European Commission issued Regulation (EC) No 1881/2006, setting the limits for heavy metals and other contaminants in various food categories: for what concerns fish and fishery products, tolerable limits for Hg, Cd, Pb, dioxins, polychlorinated dibenzofurans (PCBs) and polycyclic aromatic hydrocarbons (PAHs) are defined.

In our study we considered Hg, Cd and Pb, that are three of the

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most frequently studied elements and the most important form of pollution of the aquatic environment because of their toxicity and accumulation by marine organisms (Okyere, Voegborlo, & Agorku, 2015); heavy metals accumulation in aquatic organisms is strictly related with species, trophic level in food chain (Storelli & Marcotrigiano, 2004) and environmental characteristics. Hg is emitted both from natural sources (volcanic activities, water's evaporation) (Bernardi, Tirloni, & Cattaneo, 2015) and human sources (gold mining, institutional boilers etc.) (Mozaffarian & Rimm, 2006). Methyl-Hg (MeHg) is the predominant form of organic Hg present in seafood (Seixas, Bustamante, & Pierce, 2005); it accumulates in organisms to a greater extent than other forms (Jinadasa, Ahmad, Edirisinghe, & Wicramasinghe, 2014), and it is the only Hg form that can be readily absorbed and actively transported into tissues (Bernardi et al., 2015; Mozaffarian & Rimm, 2006); furthermore, fish appears to be the primary source of MeHg poisoning in humans (Rice, Walker, Wu, Gillette, & Blough, 2014). Mercury exposure can cause kidney damage and tubular necrosis, glomerulonephritis, arrhythmias, hypertension, DNA damage (Yabanli & Alparslan, 2015), cardiomyopathies, angina, inflammatory bowel disease, digestive disturbances and destruction of intestinal flora, destruction of polymorphonuclear leukocytes (Rice et al., 2014); Hg is also associated with increase oxidative damage, disruption of the pituitary, thyroid, adrenal glands and pancreas (Rice et al., 2014). Food is the primary source of Cd exposure in the general population, and tobacco represents an important additional source for smoking people (Thomas, Elinder, Wolk, & Akesson, 2014). Cd exposure may result primarily in kidney damage as chronic kidney disease, testicular tumors, renal dysfunction, growth inhibition, cancer, hypertension and arteriosclerosis (Yabanli & Alparslan, 2015), reproductive and neurological disorders (Obaidat, Massadeh, Al-Athamneh, & Jaradat, 2015). Pb is a highly toxic and non-essential metal; it accumulates particularly in hepatic and renal parenchyma where it causes renal failure and liver damage (Luckey & Venugopal, 1977); Pb can also cause physical and mental growth retardation in infants, learning deficiency in children, anemia (Yabanli & Alparslan, 2015), health disorders such as sleeplessness, tiredness, hear and weight loss (Zhu et al., 2015).

In the present study the level of contamination of Cd, Hg and Pb was assessed in fishery products imported from extra-EU Countries and destined to the EU market, caught in various FAO fishing areas of the world and inspected by official public veterinarians at the Border Inspection Post (BIP) of Milan Malpensa airport, which is one of the main cargo airports in Europe and the busiest in Italy in terms of freight volume. Data obtained from the fishery products arriving in Milan Malpensa by air are a representative sample of the contamination of fishery products that daily arrive in Europe from the main exporters of the world. We think our work is good especially because we considered a wide range of fish and fishery products and a wide range of FAO fishing areas from which they have been caught.

## 2. Material and methods

### 2.1. Samples

Analyses were performed on samples taken from fishery products originating from extra-EU Countries and reaching EU market through the airport of Milan Malpensa; products were generally destined to the Northern Italian market. Samples were collected during the period included between January 2010 and December 2013; they represent approximately 3% of the total volume of fishery products arriving in Milan Malpensa and, accordingly to the Guidelines of Italian Ministry of Health, were oriented to the

species that majorly accumulate heavy metals: tunafish, swordfish, sharks and cephalopods.

All the products were submitted to the official border veterinary inspection before their entrance in the EU market, in this case the Italian market. For this study fish products were divided into four categories:

- Predatory fish (n = 94): fishes of big dimension and predators in marine ecosystem, which included 26 swordfish (*Xiphias gladius*), 61 yellowfin tuna (*Thunnus albacares*) and 7 spiny dogfish (*Squalus acanthias*) originating from Sri Lanka, Maldives, USA (spiny dogfish) and a few from India, Philippines, Chile, Tunisia, Indonesia and Seychelles;
- Non-predatory fish (n = 66): fishes of lower dimension than predatory fish and that have a lower trophic level in the marine food chain, which included *Lates niloticus*, *Lophius* spp., *Synaptura* spp., *Zeus faber*, *Ephinepelus* spp., *Cynoglossus* spp., *Seriola dumerili*, *Scorpaena scrofa*, *Sebastes marinus*, *Leucoraja ocellata*, *Mullus* spp., *Brotula barbata*, *Xyrichtis novacula*, *Dissistichus eleginoides* from many extra-EU Countries of Africa and, in two cases, from USA and Chile;
- Mollusc (n = 36), which included 28 cephalopods (*Loligo* spp., *Octopus vulgaris*, *Sepia officinalis*, *Sepiolo*) and 8 bivalves (*Placopecten magellanicus*, *Argopecten purpuratus*, *Pecten yessoensis*), originating from USA, Thailandia, Morocco, Egypt, Tunisia;
- Crustacean (n = 55), which included 49 lobsters (43 *Homarus americanus* and 6 *Palinurus* spp.) and 6 other species of crustaceans, like *Penaeus keraturus*, *Pseudopenaeus prayensis*, *Scyllarides* spp., *Palaromis granulosa*, originating from USA, Canada, South Africa.

During the considered period, 220 determinations of Cd, 228 determinations of Hg and 207 determinations of Pb were carried out; in most cases, the same sampling was carried out to analyze on the same specimen two or three heavy metals simultaneously. As shown in Table 1, were analyzed for predatory fish 67 samples for Cd, 94 for Hg and 67 for Pb; for non-predatory fish 65 for Cd, 64 for Hg and 64 for Pb, for mollusc 35 for Cd, 24 for Hg and 28 for Pb while for crustacean 53 for Cd, 46 for Hg and 48 for Pb. The choice of which species subject or not subject to analyses was made basing on scientific knowledge, accordingly to the predisposition of each category to accumulate each heavy metals; therefore, predatory fish were analyzed especially for Hg, mollusc for Cd while crustacean and non-predatory fish were analyzed equally for all three heavy metals considered. Each sampling has been conducted by an official public veterinarian working in the airport. For each consignment, according to EU legislation, the Guidelines for BIP recommend to perform, after systematic documental and identity controls, material and laboratory control in precise percentages, generally oriented to the risk species of the specific kind of analysis. The sampling procedure was made accordingly to Regulation (EC) No 333/2007; the number of elementary samples that constitute each aliquot varied accordingly to the lot's weight: 3, 5 or 10 elementary samples were used if the weight of the lot was <50 kg, 50 kg < weight < 500 kg or >500 kg, respectively. The elementary samples were grouped in 3 aliquots; each aliquot must have a minimum weight of 1 kg. Elementary samples were collected in different ways according to the matrix sample: for what concerns predatory and non-predatory fish, each elementary sample consisted in a piece of variable weight (from 100 g to 400 g, approximately), each taken from 3, 5 or 10 different individuals in accordance with the lot's weight; for cephalopods and crustaceans, each elementary sample was a whole individual or a group of more than one individuals, in order to reach the minimum weight of the aliquot; for bivalves, 3, 5 or 10 samples from different points of the

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