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Mussels and clams from the italian fish market. is there a human exposition risk to metals and arsenic?

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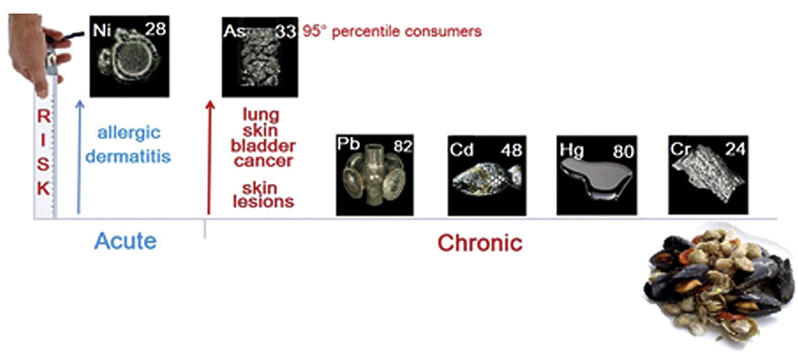
HIGHLIGHTS

- Mussels and clams are filter feeders that can bioaccumulate various chemicals.
- Seafood metal contamination is a recognized public health risk.
- The presence of metals and arsenic in these molluscs from Italian market was checked.
- A very low risk for the consumer health emerged due to the intake of molluscs.
- Allergic dermatitis in Ni-sensitive subjects may be induced by all analysed samples.

GRAPHICAL ABSTRACT



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ABSTRACT

Seafood is associated with many beneficial effects on human health. However, the overall level of contaminants in biota has increased over the last two centuries and seafood is one of the source of oral exposition to contaminants. Therefore, this work aimed to evaluate cadmium, lead, mercury, arsenic, chromium and nickel presence in mussels and clams, from the Italian market, and the associated risk. The samples were from five different FAO areas. Analyses were carried out using inductively-coupled plasms-mass spectrometry. The sample concentrations were below the maximum levels stated by Commission Regulation (EC) 1881/2006, except one mussel sample, which was non-compliant for cadmium (2.13 ± 0.20 mg kg^{-1}). For arsenic, nickel and chromium, maximum levels are not stated by the European Union. In this study, arsenic ranged from 1.29 to 13.35 mg kg^{-1} and nickel ranged from <LOQ–3.98 mg kg^{-1}, except one sample, whose nickel concentration was 21.70 mg kg^{-1}. Chromium was found only in 15 samples, with a maximum concentration of 2.81 ± 0.27 mg kg^{-1}, in one clam sample. Our results indicate that the average Italian consumption of molluscs, does not pose a risk for consumers, except nickel, which can cause allergic dermatitis in nickel-sensitive individuals. However a particular concern is caused by the exposition to As of the 95th percentile consumers: the Hazard Index for skin

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lesions, was >1, and $BMDL_{10}$ for lung bladder and skin cancer in all mussel samples was overcome, in the 100% and 25% of mussel and clam samples, respectively.

1. Introduction

Seafood is an excellent source of nutrients including proteins, vitamins (A, D and B_{12}), minerals and fatty acids, with important human health benefits. Epidemiological and clinical evidence suggests that consumption of high levels of n-3 fatty acids eicosapentaenoic acid and docosahexaenoic acid, from one or two fish-based meals per week, induces protective effects against coronary heart disease, with a 36% decrease in the risk of coronary death and 17% decrease in total mortality. Seafood is also associated with beneficial effects on neurologic development in toddlers and children, such as improved visual acuity, raised mental processing scores and language comprehension (Mozaffarian and Rimm, 2006; EFSA , 2014a). However, over the last two centuries, the level of contaminants, heavy metals and chemical compounds, has increased in the environment and, subsequently, in biota, due to anthropogenic activity, such as industrial activities, mining and agriculture. Some metals, such as lead (Pb) and mercury (Hg) have no biological roles (Sarmiento et al., 2011) and their presence needs to be monitored to prevent and minimise the potential health risks associated with seafood consumption. In many instances, animals near the top of the food chain, such as carnivorous fish species, are most affected by biomagnification of xenobiotics, building up greater and or more dangerous amounts of toxic materials than animals lower down the food chain. Herbivorous species, like mussels and clams, are filter-feeders, bioaccumulating a wide range of environmental pollutants in their tissues and, thus, can be used as suitable bioindicator organisms to monitor trace metal pollution in marine environments (Langston and Spence, 1995). In the European Union (EU), the breeding of bivalves provides the laying of juvenile bivalves in the open sea and their permanence for about 2 years until adult stage (Baylon, 1990). After collection, the animals are microbiologically controlled, before their marketing. If they do not meet the microbiological criteria after collecting, they must stay in a depuration place, provided with periodically replaced clean water, in order to reduce the microbial load (EU, 2004). In this context, it could be supposed that they can release several chemicals, comprising metals.

One fundamental feature of metals is their ability to persist in the environment (Scientific Committee of Problems of the Environment (SCOPE), 1987). Consequently, they can be taken up by marine organisms due to their presence in water and sediments and they can be biomagnified in superior trophic animals including humans (Kim and Lee, 2010). The metal contamination of seafood is a globally recognised public health risk (Lozano et al., 2010; Squadrone et al., 2016). Long-term exposure and/or high concentrations of metals, can cause adverse effects on human health, such as skin diseases, acute and chronic intoxication, nervous system, blood and gastrointestinal dysfunctions, respiratory problems, as well as mutagenic and cancerogenic effects (Martin and Griswold, 2009).

In particular, Pb can cause severe brain and kidney damage, and in pregnant women, may cause miscarriage. Hg can cause nervous system dysfunctions like tremors, irritability, memory problems, changes in vision and hearing. Cadmium (Cd) can cause kidney diseases and respiratory problems. Inorganic arsenic (As) has been linked to cancer of the skin, lungs and bladder. Chromium (Cr) can cause breathing problems, cough, asthma, allergic reactions and chronic exposure could cause liver and kidney cancer, particularly linked to Cr(VI) (Martin and Griswold, 2009). Nickel (Ni) could cause allergic reactions and long-term exposure could result in reproductive diseases. Moreover, it is genotoxic, with cancerogenic, immunotoxic, hepatotoxic, neurotoxic, and nephrotoxic effects only after inhalation (EFSA, 2015; Das et al., 2008).

The EC has set maximum levels (MLs) for Cd (1 mg kg^{-1}) and Pb (1.5 mg kg^{-1}) in bivalve molluscs, and for Hg (0.5 mg kg^{-1}) in seafood (EU, 2006). No MLs have yet been established for As, Ni, and Cr by the EU. In 2009, however, the European Food Safety Agency (EFSA, 2009a) concluded that the provisional tolerable weekly intake (PTWI) of As at 15 μg kg^{-1} body weight, recommended by the Joint Food and Agricultural Organisation/World Health Organisation (FAO/WHO) Expert Committee on Food Additives (JECFA) was no more applicable. Instead, based on epidemiological studies on exposure to As through water and food, the EFSA suggested a range, rather than a single reference point that included the benchmark dose lower confidence limit for a 0.1% increased incidence ($BMDL_{01}$) of skin lesions and cancer of the lung, skin and bladder, between 0.3 and 8 μg kg^{-1} body weight day^{-1} (EFSA, 2009a,b).

Although Cr has no defined MRLs, in 2014, the EFSA suggested a tolerable daily intake (TDI) for Cr(III) of 300 μg kg^{-1} body weight, which was based on the lack of adverse effects and carcinogenicity in mice and rats, and for the inadequate information about reproductive and developmental toxicity. Cr(VI) is classified by the International Agency for Research on Cancer (IARC) as carcinogenic to humans (Group 1). However, it is scarcely present in food, which is mostly a reductive medium and is largely reduced in the gastrointestinal tract. Therefore, the EFSA concluded that the oral exposure to Cr(VI), for the European population is of low concern for health of all age groups but of potential concern only for high consumers of drinking or bottled water, for the oxidising agents present, in the groups “Infants”, “Toddlers” and “Other children” (EFSA , 2014b).

In 2015, the EFSA set a TDI for Ni at 2.8 μg kg^{-1} body weight, obtained from studies on the dose response curve of the incidence of litters with a post-implantation loss in rats. EFSA, also, considered, the possibility of eczematous and allergic reactions elicited by acute oral exposure and reported a $BMDL_{10}$ of 1.1 μg Ni kg^{-1} body weight, with a margin of exposure (MOE) of 10 or higher, accounting for the variability of immune response in nickel-sensitised individuals (EFSA, 2015).

According to the 2016 European Market Observatory for Fisheries and Aquaculture Products (EUMOFA) report, the consumption of seafood products has been constantly increasing in the last few years, with mussels among the major species. In 2014, 1.27 kg mussels per person per year and 0.33 kg clams per person per year, was recorded. In the same year, the EU production of bivalves increased 14%, reaching 609,600 tons, mainly due to an increase in mussels farmed in Spain, the major producer, followed by France and Italy, where bivalve production represents 70% of all fishery national production (EUMOFA, 2016).

In the current study, inductively coupled plasma-mass spectrometry (ICP-MS) was used to quantitate the six mentioned metal contaminants (As, Cd, Cr, Hg, Pb and Ni), in mussels and clams