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Lead, mercury and cadmium levels in edible marine molluscs and echinoderms from the Veneto Region (north-western Adriatic Sea – Italy)

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ABSTRACT

In this study, heavy metal levels detected in molluscs produced in the Veneto Region (north-western Adriatic Sea) are presented and discussed. A descriptive analysis was conducted on data collected in the framework of official monitoring activities in shellfish production areas between 2007 and 2012. Concentrations of lead (Pb), mercury (Hg) and cadmium (Cd) in about 2000 samples of 17 different species of marine bivalve molluscs, echinoderms and gastropods were checked by means of atomic absorption spectrometry. Hg and Pb concentrations were always below the maximum levels set by the European legislation; 2.2% of the analysed samples exceeded the threshold level for Cd, 86.7% of witch belonging to purple dye murex (*Bolinus brandaris*). The highest mean levels of Hg and Cd were found in purple dye murex, with 0.10 mg kg^{-1} and 0.88 mg kg^{-1} respectively, while rayed trough shell (*Mactra stultorum*) and truncate donax (*Donax trunculus*) were the species with the highest mean value of Pb (0.25 mg kg^{-1}). Species of major consumption showed mean values almost ten times lower than the limits set by the European legislation (Manila clam: Pb: 0.20 mg kg^{-1}; Hg: 0.04 mg kg^{-1}; Cd: 0.07 mg kg^{-1}; Mediterranean mussel: Pb: 0.22 mg kg^{-1}; Hg: 0.03 mg kg^{-1}; Cd: 0.13 mg kg^{-1}). A preliminary evaluation of the impact on human health of shellfish according to the detected levels indicates that the species of major consumption are safe for the average consumers, although a potential risk cannot be ruled out for vulnerable groups, such as women of childbearing potential and children, as well as for regular or high-level shellfish consumers, particularly those who frequently eat species such as murex.

1. Introduction

Italy is the third European producer of marine molluscs (~150.000 t), after Spain (~270.000 t) and France (~250.000 t) (FAO, 2011). About 70% of the Italian shellfish production is consumed domestically, while the remaining 30% is exported to European countries, first among them Spain (Sladonja, Bettoso, Zentilin, Tamberlich, & Acquavita, 2011).

The national shellfish production relies mainly on two species, the Manila clam (*Venerupis philippinarum*) and the Mediterranean mussel (*Mytilus galloprovincialis*), which are also the most largely consumed. Italy is the leading European producer of Manila clams and the third worldwide producer of Mediterranean mussels, after China and Spain (Robert et al., 2013).

The Italian mollusc production is mostly concentrated in the north Adriatic regions. In particular, the Veneto Region is a leader in the national shellfish industry, being the first producer of Manila clams and the third producer of mussels in the country (Robert et al., 2013). Other minor species are also produced in this area.

The northern Adriatic Sea is not particularly deep and is fed by three large waterways: the Po, the Adige and the Brenta rivers. They flow across the Po valley, where there are numerous urban settlements, as well as industrial and zootechnical activities. Moreover, the Venice Lagoon is classified as being a sensitive area exposed to anthropogenic pollutants sources, the main being the Industries of Porto Marghera and the discharge of urban waste water sewage and urban runoff from the municipality of Venice (Giusti & Zhang, 2002).

As mentioned above, this area is well known for its shellfish harvesting activity, whose extensive production can be an easy target for chemical contamination. Lead (Pb), cadmium (Cd) and mercury (Hg) are chemical elements of great concern from a public

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health point of view, being environmental pollutants which can occur naturally in the environment and also derive from either industrial (e.g. mining, metallurgical, incineration, pesticide and varnish production activities) or agricultural sources (e.g. pesticide and fertilizers use). These contaminants are highly toxic and may accumulate in seafood, whose consumption can represent an important route of human exposure to these harmful substances (Storelli, 2008).

Shellfish have an excellent ability to filter large amounts of water, and they can accumulate high concentrations of heavy metals (Claisse, 1992; Fisher, 1983; Roméo & Gnassia-Barelli, 1995; Soto, Cajaraville & Marigómez, 1996). The capacity of accumulating metals may vary among molluscs depending on their filtering activity and their position in the water column. Oysters can accumulate concentrations of Cd three times higher than mussels, if placed in the same environment (Claisse, 1992). McConchie and Lawrence (1991) also reported the capacity of bottom dwelling oysters to accumulate a quantity of Cd which can be up to 10 times higher than that of oysters grown in the same site but in baskets placed on the surface of the water. Metal concentration in molluscs can be influenced by many physiological factors like individual size, age or breeding (Claisse, 1992). Regarding the latter, the levels detected in mussels or oysters at the time of sexual maturity remain the same, although the concentration of metals seems to be lower due to the increase of the body size. This pattern is called 'biological dilution' and has been observed in these species for cadmium, copper, lead and zinc (Guéguen et al., 2011).

The current European Food Hygiene Legislation makes the control and monitoring of all classified relaying and production areas of bivalve molluscs, gastropods, echinoderms and tunicates (EU Council, 2004b) mandatory, in order to ensure the compliance of the product with microbiological, biotoxicological and chemical parameters and its suitability for human consumption (EU Commission, 2005; EU Council, 2004a; EU Commission, 2006). In order to properly perform the above mentioned monitoring activities, Italy implemented specific guidelines for the Veterinarian Competent Authority and for Food Business Operators (Presidenza del Consiglio dei Ministri, 2010).

As reported in a survey on the perception of food-related risks in the EU, European citizens seem to be more concerned about the chemical contamination of food rather than bacterial contamination itself or health and nutrition issues (Eurobarometer, 2010). The availability to access data on chemical contamination issues is becoming of increasing significance allowing better health impact assessments of food consumption and helping provide the consumer with more reliable and complete information on the health risks that food contamination may cause.

Little information is available in literature on the heavy metals level of shellfish detected in the framework of official monitoring programmes in Italy.

In this paper, levels of Pb, Hg and Cd found in the period 2007–2012 in edible molluscs and echinoderms from lagoons and marine areas of the Veneto Region in the context of official control activities are presented and discussed together with a preliminary evaluation of the impact on human health of locally produced shellfish.

2. Materials and methods

2.1. Sampling

Sampling was performed in the framework of the classification and monitoring activity on mollusc production areas by the local competent veterinary authority. Molluscs and echinoderms were collected from the lagoons and coastal areas within 12 nautical miles, attributable to the Veneto Region, North eastern Italy (Fig. 1).

Since 2006, lagoons (Venice and the Po River Delta lagoons) and seashore of the sampling area have been divided into production areas defined as "sea", "estuarine" or "lagoon" areas, containing either natural beds of bivalve molluscs or sites used for the cultivation of bivalve molluscs, and from which live bivalve molluscs are taken (EU Council, 2004a) and classified according to EU Regulation 854/04 (EU Council, 2004b). In each area, all the species for which harvesting activities are allowed are sampled at least twice a year to check the level of heavy metals; the period of the year as well as the exact sampling site are established each time by the official veterinarian. Since the overall sampling activity for each species covered all the year, with a slight decrease during the winter months probably due to weather conditions, we can reasonably consider that samples analysed are representative of the average year condition.

The environment from which the sampled species were collected (lagoon or sea) and their type of production site (farm or natural banks) are reported in Tables 2–4. About 2000 samples of 17 different species representative of the Veneto Region production, including bivalves, gastropods and echinoderms, were collected between January 2007 and December 2012 in order to check the levels for Pb, Hg and Cd; more specifically, 2038 samples were analysed for all the three metals and 54 samples of purple dye murex were analysed only for Cd, this being a species particularly at risk for Cd accumulation.

The sample size, reported in brackets, is distributed as follow: Manila clam – *V. philippinarum* (871), Mediterranean mussel – *M. galloprovincialis* (378 for Pb and Hg; 377 for Cd), striped venus – *Chamelea gallina* (147), grooved carpet shell – *Ruditapes decussatus* (109), smooth callista – *Callista chione* (90), common edible cockle – *Cerastoderma edule* (71), minor jackknife – *Ensis minor* (46), variegated scallop – *Mimachlamys varia* (41), Pacific cupped oyster – *Crassostrea gigas* (24), European flat oyster – *Ostrea edulis* (17), rayed trough shell – *Mactra stultorum* (13 for Pb and Hg; 14 for Cd), great Mediterranean scallop – *Pecten jacobaeus* (13), warty venus – *Venus verrucosa* (9) and truncate donax – *Donax trunculus* (8), purple dye murex – *Bolinus brandaris* (109 for Pb and Hg; 163 for Cd), changeable nassa – *Nassarius mutabilis* (51) and purple sea-urchin – *Paracentrotus lividus* (41) (see http://www.sealifebase.org consulted on 06 June 2014).

In general, the most frequently sampled species were Manila clams (42.7%) and Mediterranean mussels (18.5%), followed by striped venus (7.2%), grooved carpet shell (5.3%) and purple dye murex (4.4%). Other minor species accounted for the remaining 21.9% of samples.

The overall yearly distribution of samples was uniform, with a mean of 340 samples *per* year in the case of Pb and Hg (range: 292–375) and 349 samples for Cd (range: 292–396).

Each sample was composed of one aliquot of at least 1.5 kg of adult individuals (commercial size) belonging to the same species, collected directly from the sea or lagoon and placed in a sealed plastic bag. Once collected, samples where transported within the day to the laboratory at refrigerated temperature, accompanied by an identification label and a dedicated sampling form. Once they reached the laboratory, the samples were stored at $-20\ ^{\circ}C$ pending analysis.

2.2. Experimental

2.2.1. Atomic absorption instrumentation

For Pb and Cd determination, a Thermo Electron (Waltham, Massachusetts, USA) M6 mkII Atomic Absorption Spectrometer