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Distribution of heavy metals in internal organs and tissues of Korean molluscan shellfish and potential risk to human health

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Abstract

Molluscan shellfish (gastropods and bivalves) were collected from major fish markets on the Korean coast and analyzed for mercury by direct Hg analyzer and for other metals, such as cadmium, lead, chromium, silver, nickel, copper and zinc, using inductively coupled plasma mass spectrometry. Distribution of heavy metals in muscles, internal organs and whole tissues were determined and a potential risk assessment was conducted to evaluate their hazard for human consumption. Heavy metals were accumulated significantly higher ($P < 0.05$) in internal organs than in muscles for all species. The mean Cd level, which had the highest level of three hazardous metals (Cd, Pb, and Hg) in all internal-organ samples were above the regulatory limit of Korea and the mean level in whole tissue samples of the selected gastropod species, bay scallop and comb pen shell, exceeded the limit (except in a few cases). The sum of the estimated dietary intake of Cd, Pb and Hg for each part of all tested species accounted for 1.59-16.94, 0.02-0.36, and 0.07-0.16% respectively, of the provisional tolerable daily intake adopted by the Joint FAO/WHO Expert Committee on Food Additives. The hazard index for each part of gastropods and bivalves was below 1.0, however, the maximum HI for internal organs of all analysed species was quite high (0.71). These results suggest that consumption of flesh after removing the internal organs of some molluscan shellfish (all gastropod species, bay scallop and comb pen shell) is a suitable way for reducing Cd exposure.

Key words

Distribution, Heavy metal, Korea, Molluscan shellfish, Risk assessment.

Introduction

Fisheries products such as fish, aquatic plants, mollusks and crustaceans are an important food resource worldwide. According to the Food and Agriculture Organization (FAO) of the United Nations (2009), Korea was the seventh-largest consumer of fishery products in the world in 2009, accounting for 56.1 kg per person per year, which is threefold higher than the global average consumption. FAO (2011) also reported that Korea produced 737,855 tons of mollusks in 2011, which is the world's fourth, accounting for almost 3.5% of the global production. The products are exported mainly to the United States, China, Japan and the European Union (EU) (Mok *et al.*, 2014a). Therefore, the safety of Korean seafood is very important to Korea and importing countries.

Heavy metals found naturally in the environment vary widely. Certain metals such as iron, copper, zinc and manganese are essential for the human body since they play important roles in biological systems. However, others such as mercury, lead and cadmium can cause harm to the human body, even in trace amounts (Alina *et al.*, 2012; EOS Ecology, 2012). In general, heavy metals accumulate in marine organisms from aquatic environment, especially in various species of molluscan shellfish (Carrington and Bolger, 2002; Kobal *et al.*, 2004; Mora *et al.*, 2004; Borak and Hosgood, 2007). It is, therefore, obvious that numerous studies have been carried out on metal pollution in the various species of molluscan shellfish (Ayas *et al*., 2007; Sivaperumal *et al*., 2007; Mok *et al*., 2010). It is well known that the accumulation patterns of the contaminants in aquatic organisms depend both on their uptake and elimination rates (Guven *et al*., 1999). Heavy metals are taken up through different organs of the organisms and concentrated at different levels in each organ of the body (Bervoets *et al*., 2001; Bustamante *et al*., 2000; Kumar and Achyuthan, 2007)

Some hazardous metals (Cd, Pb and Hg) accumulated in molluscan shellfish can cause risk to humans *via* consumption of the organisms (Tuzen and Soylak, 2007; Yilmaz *et al*., 2007). To protect public health, the Korean authority has established regulatory limit for three metals in only edible part of the molluscan shellfish (KMFDS, 2013). The internal organs of all gastropods and some bivalves are not officially considered edible by Korean health authorities; however, the internal organs of these species are considered edible in many cultures including Korea. The consequence of heavy metal pollution can be hazardous to humans therefore, it is important to check regularly the amount of heavy metals in edible and "non-edible" tissues of molluscan shellfish to provide information about the hazard levels to consumer.

In the present study, the distribution of heavy metals was determined in muscles, internal organs and whole tissues of molluscan shellfish collected from three fish markets on the Korean coast. Estimated dietary intake (EDI) of heavy metals *via* consumption of each portion of molluscan shellfish was compared with the provisional tolerable daily intake (PTDI) values. In addition, a potential risk assessment associated with these heavy metals in different portions of the molluscan shellfish was carried out using target hazard quotient (THQ) and hazard index (HI).

Materials and Methods

Sample collection and preparation: Three species of gastropods (*Nordotis discus*, *Batillus cornutus* and *Rapana venosa*) and three species of bivalves (*Crassostrea gigas*, *Argopecten irradians* and *Atrina pectinata*) were collected between March and November in 2011 at three major fish markets located on the eastern (Pohang), western (Gunsan) and southern (Yeosu) coasts of Korea (Table 1). These species were selected due to increased popularity among the consumers in Korea. Although the internal organs of the selected species were typically considered non-edible, excluding the oyster, some consumers in Korea occasionally eat the whole tissue of other selected species, including the internal organs. The samples were transported to laboratory under cold condition.

Upon arrival at the laboratory, the collected samples were immediately separated according to species and washed with tap water and deionized water. When possible, at least eight specimens per species were examined. The shells of bivalves and gastropods were shucked. The specimens were dissected and separated into muscle (or adductor) and internal organs (or digestive glands). The separated samples were weighted, homogenized, and then stored frozen below −20 °C prior to analysis. The homogenized tissues were freeze-dried with a vacuum freeze dryer (FDU-2100, EYELA, Tokyo, Japan) and then ground into powder for analysis. About 1.0 g of the powdered sample was placed in 60 ml digestion vessel (Savillex, Eden Prairie, MN, USA), to which 20 ml nitric acid (Merck, Darmstadt, Germany) was added. The vessel was then covered and left overnight at room temperature. The samples were digested using a heating digester (Digi PREP HP, SCP science, Champlain, NY, USA). More nitric acid was added to the completely non-digested samples. The completely digested samples were allowed to cool to room temperature, dissolved in 2% nitric acid, filtered (glass wool) and made up to 100 ml of 2% nitric acid for analyzing heavy metals, except for Hg. Approximately 0.1 g of homogenized sample was used for Hg analysis.

Heavy metal analysis: All the digested samples were analyzed in triplicate for silver, cadnium, chromium, Copper, nickle, lead and zinc, using an inductively coupled plasma mass spectrometry (ICP-MS; ELAN DRC II, PerkinElmer, Waltham, MA, USA). Total Hg in homogenized samples was measured directly in triplicate with a combustion gold amalgamation method using direct mercury analyzer (DMA-80, Milestone, Milano, Italy). The blanks, calibration standards diluting stock standard solutions (Merck, Darmstadt, Germany) and CRMs were also analyzed using the same methods. The concentration of heavy metals are expressed in micrograms per gram of sample wet weight.

The accuracy of heavy metal analysis method was assessed using the SRM-1566b (oyster tissue) certified reference material (CRM) provided by the National Institute of Standards and Technology (Gaithersburg, MD, USA). The quantitative recoveries of heavy metals in the oyster tissue CRM ranged from 91.1 to 107.7%; Cd (96.1%), Hg (91.2%), Pb (103.6%), Ag (91.1%), Ni (107.7%), Cu (97.8%) and Zn (95.6%). The recoveries were within the acceptable values recommended by AOAC International (AOAC, 2002).

Statistical analysis: Statistical evaluation was conducted using analysis of variance with general linear model procedure (SAS version 9.2, SAS Institute, Cary, NC, USA). Duncan's multiple range test was applied to determine the significance of differences between mean concentration of heavy metals in samples.

EDI of heavy metals: EDI of heavy metals via consumption of molluscan shellfish was compared with PTDI values established by the Joint FAO/WHO Expert Committee on Food Additives (JECFA, 1999; 2010a, b) or the US Environmental Protection Agency (US EPA, 2013). EDI for an adult was calculated by the equation: EDI = (HC × ADI) / BW, where HC is the mean heavy metal concentration (micrograms per gram) in each part of the molluscan shellfish, ADI is the average daily intake (grams per day) of the molluscan shellfish shown in Table 1, and BW is the average body weight (62.8 kg) of an adult human in Korea (KCDC, 2011).

The PTDI values for Hg, Pb, and Cd were based on the provisional tolerable weekly intake (PTWI) data (Hg, Pb) and provisional tolerable monthly intake (PTMI) data (Cd) from JECFA (1999; 2010a, b), in which PTWI of inorganic Hg was used for total Hg. PTDI of other metals, such as Cr (assuming that total Cr is Cr[VI]), Ag, Ni (assuming that all Ni is Ni soluble salts), Cu and Zn (assuming that all Zn is Zn and compounds) was based on the oral reference doses (RfDs) established by the US EPA (2013).

Potential risk assessment: A potential risk assessment associated with heavy metals in molluscan shellfish was performed using THQ and HI. THQs were based on the Human Health Risk Assessment approach from the US EPA (2013). The THQ was calculated using the equation THQ = (EF × ED × ADI × HC) / (RfD × BW × ET), where EF is the exposure frequency (350 days per year); ED is the exposure duration (81 years), equivalent to the average lifetime in Korea (Statistics Korea, 2013); ADI is the average daily intake (grams per day) of molluscan shellfish shown in Table 1; HC is heavy metal concentration (micrograms per gram) in each part of molluscan shellfish; RfD is the oral reference dose (micrograms per kilogram per day); BW is the average body weight (62.8 kg) of an adult human in Korea (KCDC, 2011); and ET is the average exposure time for non-carcinogens (ED×365 days per year). The RfDs of Hg, Pb, Cr, Ag, Ni, Cu and Zn used PTDIs shown in Table 3. The RfD of Cd was set at 1.0 as established by the US EPA (2013). HI was calculated by summing the target hazard quotients of individual heavy metals.

Results and Discussion

Heavy metals accumulated significantly higher levels ($P < 0.05$) in internal organs than in muscles of molluscan shellfish. The concentrations of the heavy metals in the whole tissue of molluscan shellfish decreased in the following order : Zn > Cu > Cd > Ni > Pb > Ag > Cr > Hg; however, difference between Ni and Pb and among Pb, Ag, Cr and Hg was not significant (Fig. 1).

Some heavy metals such as Cd, Hg and Pb can harm humans, even in trace amounts (Alina *et al.*, 2012). Korea has established regulatory limits for these three hazardous metals to protect the public that consumes fishery products (KMFDS, 2013). The concentration ratio of three hazardous metals of internal organs to muscles in molluscan shellfish was significantly higher ($P < 0.05$) in gastropods than in bivalves (Table 2). The mean Cd level in all internal organ samples (Fig. 1), which had the highest concentrations of the hazardous metals were above the regulatory limit (2.0 $\mu g g^{-1}$) for the edible portions of molluscan shellfish set by Korea (KMFDS, 2013) and the Codex Alimentarius Commission (2006). Although the mean Cd levels in all muscle samples were within the limit in both Korea and Codex, the mean level (1.761 to 5.173 $\mu g g^{-1}$) in whole tissue samples, with the exception of oyster, were above or a little below the limit. The ratio of internal organ Cd to muscle Cd in molluscan shellfish was 4.1 to 329.5; the highest ratio was found in spiny top shell (Table 2). The internal organs of gastropods contained 98.1 to 99.6% of the total Cd, which was much higher than the percentage in those bivalves (30.3 to 91.0%). In contrast,

Table 1 : Sample lists and average daily intake of molluscan shellfish collected from three fish markets on the Korean coast

Scientific name (*n*)	Common name	Shell length (cm)[1]	Sample tissue		Average daily intake ($\mu g kg^{-1} day^{-1}$)[2]
			Muscle	Internal organ	
Gastropods (31)					0.0115
Nordotis discus (11)	Abalone	8.5±1.2	Muscle	Digestive gland	0.0044
Batillus cornutus (10)	Spiny top shell	8.8±1.7	Muscle	Digestive gland	0.0020
Rapana venosa (10)	Veined rapa whelk	8.9±1.9	Muscle	Digestive gland	0.0038[3]
Bivalves (24)					0.0586
Crassostrea gigas (8)	Oyster	10.0±1.1	Muscle	Digestive gland	0.0198
Argopecten irradians (8)	Bay scallop	9.2±1.2	Adductor	Digestive gland, gill, mantle	0.0027
Atrina pectinata (8)	Comb pen shell	23.9±2.1	Adductor	Digestive gland, gill, mantle	0.0002

[1]Means ± standard deviations of shell length; [2] Based on Korea health statistics 2010 (KCDC, 2011); [3] Average daily intake of the veined rapa whelk include that of the moon snail (*Glossaulax didyma*)

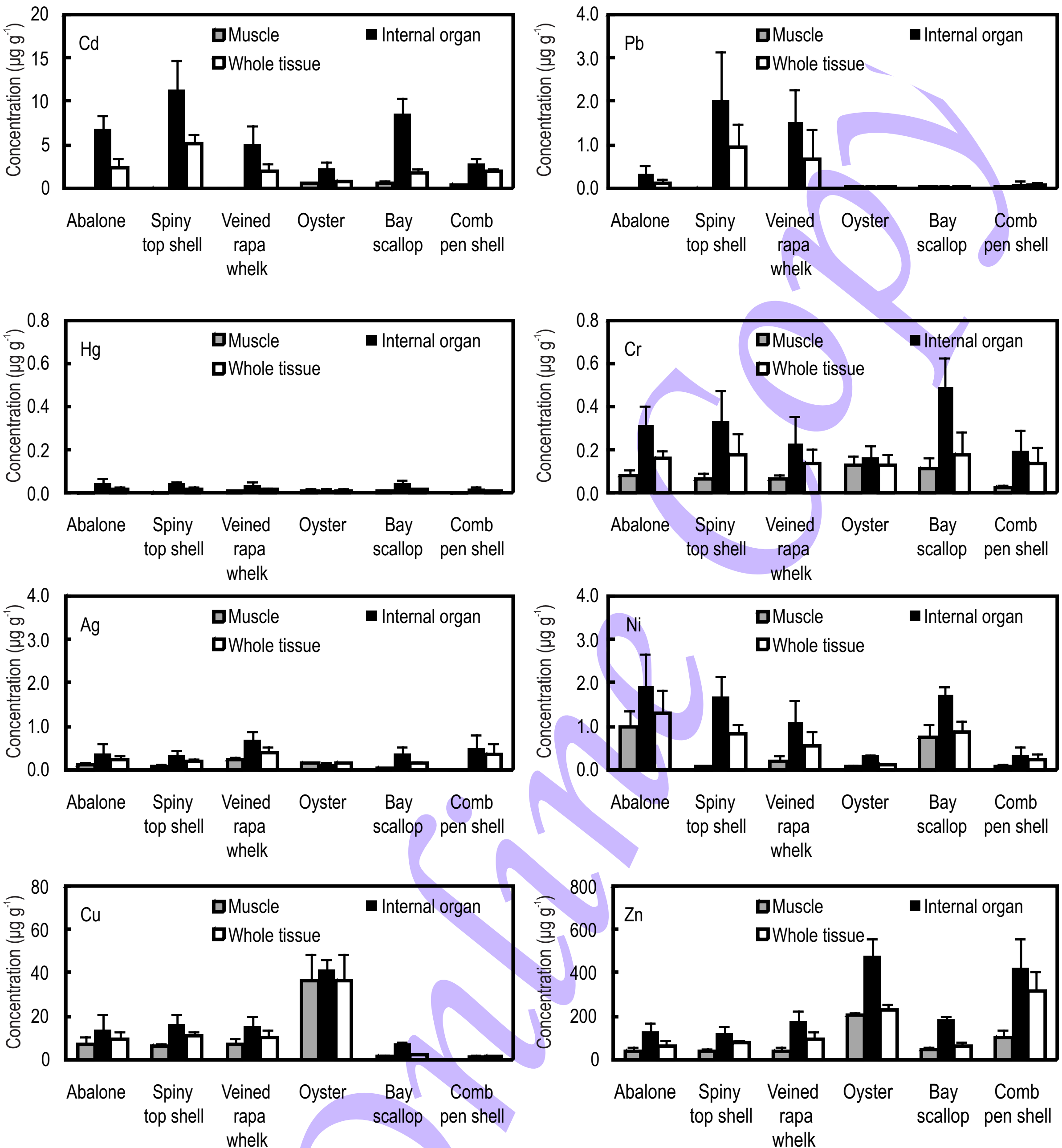

Fig. 1 : Heavy metal concentrations and distributions in muscles, internal organs, and whole tissues of molluscan shellfish collected from three fish markets on the Korean coast. Scale bar represents one standard deviation

Bustamante *et al*. (2000) reported that the digestive gland stored the mostly of the Cd, reaching 98% in some cephalopod species. Therefore, it is clear that ingesting after removing the internal organs of gastropods and some bivalve species, including bay scallop and comb pen shell, can decrease exposure to Cd.

In the present study, the mean Pb concentration in each part of all the tested species (Fig. 1), with the exception of internal organ in spiny top shell, was below the regulatory limit (2.0 $\mu g g^{-1}$) in the edible portions of molluscan shellfish adopt by Korea (KMFDS, 2013), Australia and New Zealand (FSANZ, 2008). The ratios of internal organ Pb to muscle Pb in molluscan shellfish ranged from 2.3 to 686.5; the highest ratio was also observed in spiny top shell (Table 2). Large amount of Pb was present in the

Table 2 : Ratios of heavy metals in internal organs versus muscle in molluscan shellfish from three fish markets on the Korean coast

Common name	Heavy metal ratio (internal organ/muscle)[1)]							
	Cd	Pb	Hg	Cr	Ag	Ni	Cu	Zn
Gastropods								
Abalone	132.3	88.4	12.0	3.8	3.5	2.0	1.9	3.5
Spiny top shell	329.5	686.5	8.3	5.2	3.6	24.6	2.4	3.1
Veined rapa whelk	97.3	335.2	3.9	3.3	3.0	5.6	2.3	4.5
Bivalves								
Oyster	4.1	2.3	1.2	1.3	1.0	3.6	1.2	2.3
Bay scallop	13.4	2.9	3.2	4.2	22.6	2.4	4.7	3.9
Comb pen shell	8.9	4.0	3.9	7.0	50.3	4.5	7.2	4.1

[1)]The ratio was calculated as the heavy metal concentration in a gram of internal organ divided by that in a gram of muscle.

Table 3: Estimated dietary intake (EDI) of heavy metals via the consumption of molluscan shellfish in Korea

Common name		EDI ($\mu g kg^{-1} day^{-1}$)							
		Cd	Pb	Hg	Cr	Ag	Ni	Cu	Zn
Gastropods									
Abalone	Muscle	0.0002	< 0.0001	< 0.0001	0.0004	0.0005	0.0043	0.0328	0.1626
	Internal organ	0.0301	0.0014	0.0002	0.0014	0.0017	0.0085	0.0628	0.5610
	Whole tissue	0.0103	0.0005	0.0001	0.0007	0.0009	0.0058	0.0422	0.2860
Spiny top shell	Muscle	0.0001	< 0.0001	< 0.0001	0.0001	0.0002	0.0001	0.0131	0.0792
	Internal organ	0.0228	0.0041	0.0001	0.0007	0.0006	0.0033	0.0319	0.2490
	Whole tissue	0.0103	0.0019	< 0.0001	0.0004	0.0004	0.0016	0.0217	0.1560
Veined rapa whelk	Muscle	0.0002	< 0.0001	< 0.0001	0.0003	0.0009	0.0007	0.0265	0.1547
	Internal organ	0.0197	0.0059	0.0001	0.0009	0.0026	0.0042	0.0601	0.6970
	Whole tissue	0.0074	0.0026	0.0001	0.0005	0.0015	0.0022	0.0392	0.3582
Bivalves									
Oyster	Muscle	0.0109	0.0005	0.0003	0.0025	0.0028	0.0017	0.7220	4.0510
	Internal organ	0.0443	0.0012	0.0004	0.0033	0.0027	0.0062	0.8337	9.4861
	Whole tissue	0.0140	0.0006	0.0003	0.0026	0.0028	0.0022	0.7311	4.5768
Bay scallop	Muscle	0.0017	< 0.0001	< 0.0001	0.0003	< 0.0001	0.0020	0.0042	0.1289
	Internal organ	0.0232	0.0001	0.0001	0.0013	0.0010	0.0047	0.0198	0.5042
	Whole tissue	0.0048	0.0001	< 0.0001	0.0005	0.0002	0.0024	0.0063	0.1762
Comb pen shell	Muscle	0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001	0.0001	0.0221
	Internal organ	0.0006	< 0.0001	< 0.0001	< 0.0001	0.0001	0.0001	0.0004	0.0914
	Whole tissue	0.0004	< 0.0001	< 0.0001	< 0.0001	0.0001	0.0001	0.0003	0.0675
PTDI ($\mu g kg^{-1} day^{-1}$)[1)]		0.83	3.57	0.57	3.0	5.0	20	40	300

[1)]PTDI, provisional tolerable daily intake.

internal organs of gastropods, which accounted for 95.2 to 99.8% of the total content. In contrast, the internal organs of bivalves contained 21.0 to 86.7% of Pb. As shown in Fig. 1, the mean concentrations of Hg in all the samples were below the regulatory limit (0.5 $\mu g g^{-1}$) in the edible portion of molluscan shellfish established by both Korea (KMFDS, 2013) and the EU (EC, 2005). The Hg ratio of internal organ to muscle in molluscan shellfish was 1.2 to 12.0; the maximum ratio was showed in

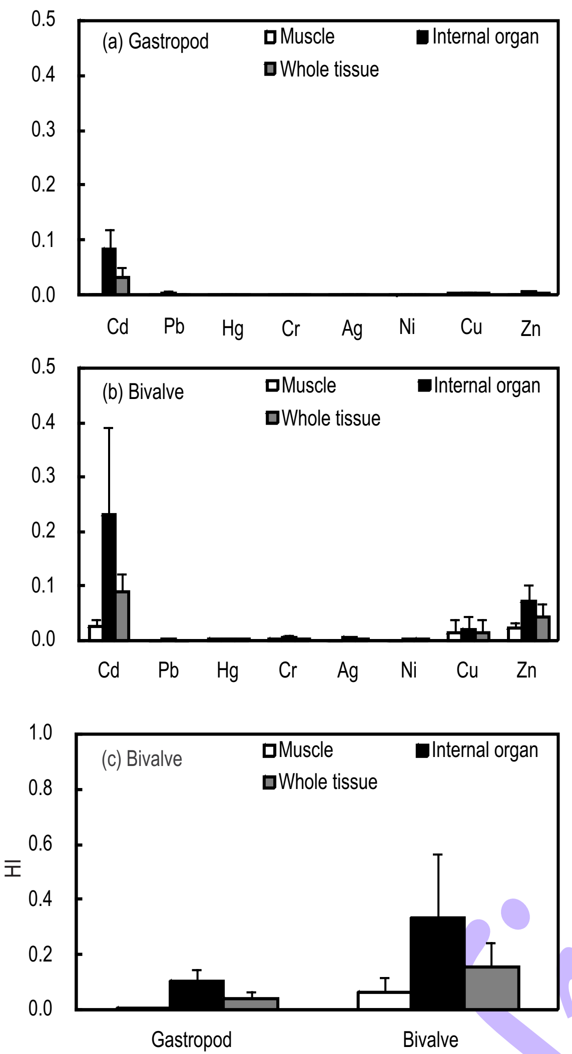

Fig. 2: Target hazard quotients (THQs) for heavy metals ingested from the consumption of molluscan shellfish in Korea. Scale bar represents one standard deviation.

abalone (Table 2). The internal organs of gastropods contained the mostly of Hg, 67.3 to 81.9% of the total content, which was much higher than Hg content in the internal organs of the bivalves (12.5 to 58.0%).

EDIs of heavy metals represent daily intake of heavy metals through consumption of molluscan shellfish for an adult. EDI was compared with PTDI suggested by JECFA (1999; 2010a, b) or US EPA (2013). PTWI for Hg and Pb (JECFA, 1999; 2010a) and PTMI for Cd (JECFA, 2010b) were established by JECFA because of the risks of these metals to human health, even in trace amount. Among the hazardous heavy metals (Cd, Hg, and Pb), the EDI values of Cd for muscles, internal organs, and whole tissues of molluscan shellfish fell in the following range 0.0001-0.0109, 0.0006-0.0443 and 0.0004-0.0140 $\mu g kg^{-1} day^{-1}$, respectively, which account for 0.01 to 1.31%, 0.07 to 5.34%, and 0.05 to 1.69% of PTDI, respectively (Table 3). The sum of Cd EDI for muscles, internal organs and whole tissues of all the tested species was 1.59, 16.94, and 5.68%, respectively, of PTDI. Particularly, the EDIs of Cd were observed relatively highly in the internal organ samples of all gastropod species and two bivalve species (bay scallop and comb pen shell). The highest value of Pb EDI was measured in internal organ of veined rapa whelk, but only 0.17 % of the PTDI value. The sum of the Pb EDIs for muscles, internal organs and whole tissues of all the species was also only 0.02, 0.56 and 0.23%, of PTDI respectively. The dietary intake of Hg was lowest of the hazardous metals in all the analyzed samples; highest EDI value was observed in the internal organs of oyster, with only 0.06% of the PTDI value. EDIs of other metals (Cr, Ag, Ni, Cu, and Zn) were compared to PTDI values based on the RfDs established by the US EPA (2013). Internal organs with the highest level of metals in each species, the highest EDI of Cr, Ag, Ni, Cu, and Zn accounted for 0.11, 0.05, 0.04, 2.08 and 3.16% of PTDI, respectively.

The THQ and HI were used to assess the potential risk of ingestion of heavy metals to an adult via consumption of molluscan shellfish. THQ was estimated by comparing the ingested amount of heavy metal with a standard reference dose (Fig. 2 (a and b)); the HI was the sum of various THQs (Fig. 2c). THQ and HI values proposed by the US EPA are integrated risk indices, and are widely used in the risk assessment of various contaminants in foods (Storelli, 2008). Both THQ and HI are recognized as useful parameters for evaluating the risk of heavy metal ingestion associated with consumption of contaminated foods (Abdallah, 2013; Li *et al*., 2013). The mean THQ of heavy metals was low in each part of gastropod and bivalve species, ranging from < 0.001 to 0.230; the highest THQ was for Cd in internal organ of bivalve. THQ was relatively high for Cd in gastropods and for Cd, Cu and Zn in bivalves, but low for Pb, Hg, Cr, Ag, and Ni, respectively.

HI exceeding 1.0 indicates that the contaminant is toxic and hazardous to human health (Abdallah, 2013; Li, *et al*., 2013; Mok, *et al*., 2014b). Heavy metal HI was higher for bivalve than for gastropod because of relatively large daily intake of bivalves shown in Table 1. The mean HI ranged from 0.005 to 0.101 and 0.064 to 0.335 for each part of gastropod and bivalve, highest was observed for internal organ of bivalves and was substantially below 1.0. In contrast, the maximum HI for internal organs of all the tested molluscan shellfish was quite high (0.71).

In conclusion, the mean level of Cd in all the sample of internal organs of molluscan shellfish were above the regulatory limit of both Korea and Codex. The highest HI for internal organs of all the tested species was quite high (0.71). These results indicate that the internal organs of some molluscan shellfish (all gastropod species, bay scallop, and comb pen shell) are unfit for consumption. Thus, it is recommended that intake of flesh after removal of internal organs, in which Cd is concentrated, is a suitable way for decreasing the exposure to harmful heavy metals.

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