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Concentration of heavy metals in shellfishes and health risk assessment from Korean coastal areas

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Abstract

Shellfish are exoskeleton-bearing aquatic invertebrates that consume various organic and inorganic substances floating in seawater through filter feeding. Heavy metals are known as absorbed and accumulated in seawater. Some of the toxic heavy metals are highly accumulated in seawater, and exposure to them can cause a variety of risks to the human body. Since Koreans like to eat seafood, they are more likely to be exposed to contaminated seafood with heavy metals. In this study, nine types of heavy metals were analyzed on ten different shellfish species in the coastal area of South Korea. The risk assessment was also done on shellfish in which heavy metals were detected. Zinc (Zn) and copper (Cu) were identified at an average of 56.7 mg/kg (6.70 to 466 mg/kg) and 13.2 mg/kg (0.064 to 143 mg/kg), respectively. Lead (Pb) average of 0.208 mg/kg (0.000750 to 1.02 mg/kg), cadmium (Cd) average of 0.454 mg/kg (0.0388 to 1.56 mg/ kg) and mercury (Hg) average of 0.0266 mg/kg (0.00548 to 0.174 mg/kg) were identified. Additionally, arsenic (As), chromium (Cr), nickel (Ni), and silver (Ag) were also identified as average concentrations of 4.02 (0.460 to 15.0 mg/kg), 0.167 (< limit of quantification [LOQ] to 0.820 mg/kg), 0.281 (< LOQ to 1.46 mg/kg), and 0.158 mg/kg (< LOQ to 1.15 mg/kg). The result indicates that the monitoring results of heavy metals in most shellfish satisfied the Korean standard. However, Pb and Cd have exceeded some foreign standards, such as the United States and the EU. The permissible human exposure calculated using the heavy metal intake and detection amount was lower than the Joint FAO/WHO Expert Committee on Food Additives human safety standard, and the risk of heavy metals from shellfish consumption was at an acceptable level.

Keywords: Heavy metals, Shellfish, Health risk assessment, Korea

Introduction

Various types of pollutants in sewage drainage or marine wastewater continuously flow into coastal waters from land. Among them, heavy metals flow into sediments or are adsorbed and accumulated in aquatic organisms because of their physicochemical properties. These characteristics, enable the monitoring of heavy metals in marine organisms to directly determine the pollution level of marine ecosystems and predict the effects on the human body. Thus, several studies have

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been conducted on this in each country. Korea and Japan are countries that consume a lot of seafood. In Korea, the annual intake of seafood per person is 58.4 kg/year, which is relatively higher than that of Norway (53.3 kg/year), Japan (50.2 kg/year), and China (39.5 kg/year) (FAO, 2016). In particular, shellfish are adherent organisms that rarely migrate, they have a wide distribution and long lifespan. Therefore, it is a useful indicator for identifying the origin and actual condition of pollutants through the identification of heavy metals with strong concentrations (Viarengo & Canesi, 1991). In addition, in Korea, heavy metals are highly likely to accumulate in the body because of shellfish consumption; thus, it is essential to evaluate human exposure to heavy metals and risk assessment.

In general, heavy metals are associated with mercury (Hg), cadmium (Cd), lead (Pb), and arsenic (As). They are well known as harmful contaminants. According to the Risk Assessment of Cadmium by MFDS (2016), the exposure of Cd through food was 0.292 μg/kg b.w./day. It was also found that marine products (0.141 μg/kg b.w./day) contributed the most. Joint FAO/WHO Expert Committee on Food Additives (JECFA, 2020) strengthens human exposure safety standards for major heavy metals such as lead, Cd, As, and Hg, and strengthens international standards for heavy metals in food.

In this study, copper (Cu) and zinc (Zn), which are essential heavy metals for the human body, and Hg, Pb, Cd, As, chromium (Cr), silver (Ag), and nickel (Ni), which are non-essential heavy metals, were analyzed in shellfish. Heavy metals are classified into two heavy metals due to their toxicity. Heavy metals were classified into two groups based on their toxicity, essential and non-essential heavy metal. Essential heavy metals are harmless or relatively less harmless at low concentration (Zn, Cu, Iron [Fe], and Cobalt [Co]). Non-essential metals are highly toxic even at low concentration (such as Cd, Hg, As, and Cr) (Kim et al., 2019). Cu and Zn, which are essential heavy metals, have metabolic effects when consumed in an appropriate amount. However, Hg, Pb, and Cd are classified as toxic substances and cause carcinogenesis when accumulated in the human body. In addition, As and Cr cause disease when ingested above the limit value.

Korea is currently conducting heavy metal safety management that focuses on exported shellfish. However, heavy metal safety management of shellfish for domestic use produced in coastal areas of the country is also required.

In this study, nine types of heavy metal monitoring were performed on the different species of shellfish. In addition, a risk assessment was done on shellfish in which heavy metals were detected. This research data is considered to help establish guidelines and reference data for safe seafood consumption.

Materials and Methods

Sample collection

Samples were collected from January to July 2018 by selecting representative species according to regional characteristics in 11 regions, including mussel (26), short neck clam (19), oysters (17), abalone (12), horned turban (11), scallop (6), surf clam (6), ark shell clam (4), comb pen clam (2), and sea squirt (8). The sampling sites are shown in Fig. 1. The collected samples were washed with distilled water according to the sample treatment manual for heavy metal experiment in seafood (MFDS, 2014). After washing, the intestines were removed from the large shellfish and only the muscles and small shellfish were separated and rewashed. The separated samples were frozen at −20℃ until analysis. The frozen samples were used for analyses after freeze-drying using a freeze-dryer (OPR-FDT8612, Operon, Gimpo, Korea). In addition, the moisture content of the wet and dry samples was measured according to the food code (MFDS, 2019).

Standards and reagents

Nitric acid was used as the reagent for sample pretreatment (Suprapur, 65%, Merck, Darmstadt, Germany). Distilled water was deionized using the Milli-Q water purification system (Millipore, Bedford, MA, USA) to obtain 18.2 mΩ. The species analyzed in this study are Hg, lead, Cd, As, Cr, Cu, Ni, Zn, and Ag. Multi-Element Calibration Standard 3 (PerkinElmer, Waltham, MA, USA) was used as the calibration standard, and the certified reference material (CRM) was 1566b (oyster tissue; NIST, Gaithersburg, MD, USA) to verified the recovery (%). CRM did not contain Cr therefore it was prepared by adding the Cr standard (Fisher Chemical, Waltham, MA, USA). The Hg-certified reference standard (Fisher Chemical) was the calibration standard.

Analytical method

Samples for heavy metal analysis excluding Hg, were used for food code (MFDS, 2019) to ensure the reproducibility and accuracy of the experimental results. The microwave method was applied for pretreatment. In other words, 10 mL of nitric acid was added to 0.5 g of a sample, digested by a microwave

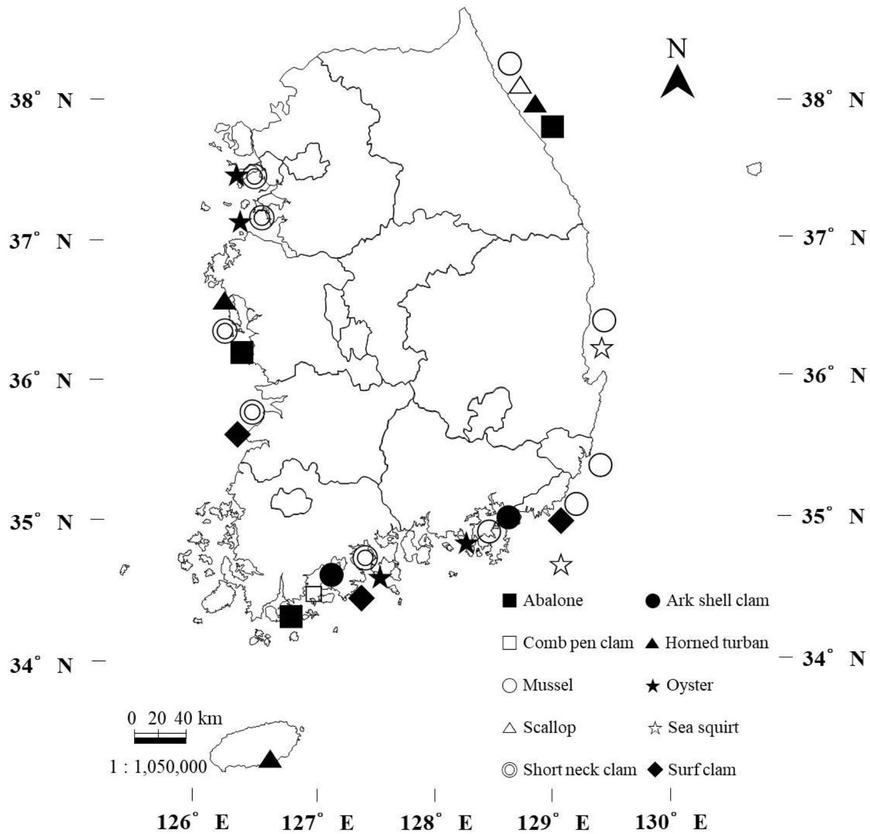

Fig. 1. Sampling sites of shellfishes from the Korean coastal area.

digestion system (START D, Milestone, Sorisole, BG, Italy), transferred to a volumetric flask, and used as a sample solution. An inductively coupled plasma mass spectrometer (inductively coupled plasma-mass spectrometer, 7700, Agilent Technologie, Santa Clara, CA, USA) was used for heavy metal analysis. Then, Hg was analyzed using the amalgam heat vaporization method (Hydra-C, Teledyne Instruments, OH, USA), and the sample weight was 0.02 g. Table 1 presents the conditions of each analytical instrument.

Method validation

The analysis results of this study validated recovery, limit of detection (LOD), limit of quantification (LOQ), and linearity of the standard calibration curve using blank, duplicated, and spiked samples according to Environmental Protection Agency (EPA) Method 3052 (US EPA, 2014).

The recovery (%) was verified using CRM (NIST, 1566b, oyster tissue). However, Cr was not contained in CRM and was confirmed using the spiked sample. Consequently, seven samples were prepared at a concentration near LOQ. In addition, the value obtained by multiplying the standard deviation of the concentration using the test method by 3.14 was determined as LOD, and the value obtained by multiplying by 10 was determined as LOQ (NIER, 2011).

Risk assessment

The risk assessment was performed by calculating the provisional tolerable weekly intake (PTWI) from the heavy metal concentration (mg/kg, ww) of shellfish and shellfish ingestion rate (g/person/day) detected in this study, and comparing it with health based guidance value of JECFA (Table 2).

The PTWI of heavy metals for Korean people through shellfish consumption was calculated using the following formula and expressed as μg per kg of body weight per day (μg/kg b.w./day):

$$\mathrm{PTWI} = \frac{C \times IR \times ED}{ABW}$$

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Table 1. ICP-MS and mercury analyzer operating conditions for the analysis of heavy metals

Instrument	Parameter	Condition
ICP-MS	Plasma flow	16.5 L/min
	Plasma forward power	1.35 kW
	Auxiliary flow	0.6 L/min
	Nebulizer flow	0.78 L/min
	Replicate intergration	3
	Mass range	4–260 amu
	Dwell time	100 μs
Mercury analyzer	Gas flow rate (O_2)	350 mL/min
	Drying	300℃, 20 s
	Catalyst	600℃, 60 s
	Decomposition	800℃, 150 s
	Amalgamator	600℃, 30 s

ICP-MS, inductively coupled plasma-mass spectrometer.

Table 2. Health based guidance value by Joint FAO/WHO Expert Committee on Food Additives

Heavy metals	Health based guidance value	Health based guidance value (PTWI)
Cd	PTMI 25 μg/kg b.w./month	5.83 μg/kg b.w./week
Cu	PMTDI 0.5 mg/kg b.w./day	3,500 μg/kg b.w./week
Zn	PMTDI 0.3–1 mg/kg b.w./day	2,100–7,000 μg/kg b.w./week
Hg	PTWI 4 μg/kg b.w./week	4 μg/kg b.w./week
As	PTWI 15 μg/kg b.w./week	PTWI 15 μg/kg b.w./week

Cd, cadmium; Cu, copper; Zn, zinc; Hg, mercury; As, arsenic; PTMI, provisional tolerable monthly intake; PMTDI, provisional maximum tolerable daily intake; PTWI, provisional tolerance weekly intake.

where C is the heavy metal concentration in shellfishes (mg/g, ww); IR is the shellfish ingestion rate (g/person/day) as used in national food and nutrition statistics (KHIDI, 2016) (Table 3); ED is the exposure duration (seven days); ABW is the average body weight (64.2 kg for adults) as used in Korean exposure factor (NIER, 2020).

Table 3. Ingestion rate (g/day) of shellfishes based on national food and nutrition statistics

Shellfishes	Ingestion rate (g/day)	
	Total person average	Ingestion person average
Abalone	0.4425	18.9439
Ark shell clam	0.0710	34.8498
Comb pen clam	0.0110	7.8624
Horned turban	0.1228	6.4833
Mussel	1.4541	22.5776
Oyster	1.1001	38.6825
Scallop	0.0614	32.4042
Sea squirt	0.2793	72.1370
Short neck clam	1.2516	9.8103
Surf clam	0.0006	0.6415

Results and Discussion

Method validation

In this study, the recovery rate was excluded from CRM (NIST, 1566b, oyster tissue) and Cr, consisting the same matrix as shellfish. The results of the spiked samples, showed that the recovery rate ranged from 83.8% to 102% (Table 4). The LOD and LOQ of the test method were less than 0.001 mg/kg (Table 5).

The coefficient of determination (R^2) of the standard calibration curve obtained by analyzing the standard solution for each step using the US EPA Method 6020B (US EPA, 2014) was 0.99 or higher. Furthermore, to the standard calibration curve, the intermediate concentration was analyzed once for every ten samples immediately after the test curve through the environmental test/inspection QA/QC handbook. The concentration satisfied the range from 90% to 110%.

A blank sample was prepared using the US EPA Method 3052 (US EPA, 1996) and analyzed following the same procedure as the sample. As a result of identifying sample contamination in the experimental procedure, there was no effect on the

Table 4. Recovery (%) of heavy metals

	Pb	Cd	As	Cr	Cu	Ni	Zn	Ag	Hg
Spiked concentration (mg/kg)	0.308	2.48	7.65	2.50	71.6	1.04	1,424	0.666	0.0371
Measured concentration (mg/kg)	0.265	2.32	7.02	2.37	61.2	0.983	1,403	0.558	0.0379
Recovery (%)	85.9	93.6	91.8	94.9	85.5	94.5	98.5	83.8	102

Pb, lead; Cd, cadmium; As, arsenic; Cr, chromium; Cu, copper; Ni, nickel; Zn, zinc; Ag, silver; Hg, mercury.

Table 5. LOD (mg/kg) and LOQ (mg/kg) of heavy metals

	Pb	Cd	As	Cr	Cu	Ni	Zn	Ag	Hg
LOD (mg/kg)	0.0002	0.0002	0.0003	0.0001	0.0002	0.0003	0.0003	0.0003	0.0003
LOQ (mg/kg)	0.0005	0.0008	0.0008	0.0005	0.0006	0.0009	0.0010	0.0010	0.0008

LOD, limits of detection; LOQ, limits of quantitation; Pb, lead; Cd, cadmium; As, arsenic; Cr, chromium; Cu, copper; Ni, nickel; Zn, zinc; Ag, silver; Hg, mercury.

experiment. Moreover, reproducibility (%) was calculated from a duplicate sample analysis of one sample for every 20 samples, and it was 0.25% to 16.8%. Moreover, the accuracy (%) of the experimental process was obtained by analyzing one spiked sample for every 20 samples, and it was 75.9% to 119%.

Concentration of heavy metals in shellfishes

Moisture was measured to calculate the content of heavy metals in the sample by wet weight. The moisture content of shellfish samples was abalone (71.5%–76.7%), ark shell clam (69.4%–74.9%), comb pen clam (69.0%–75.1%), horned turban (72.5%–77.9%), mussel (74.3%–81.9%), oyster (68.7%–78.8%), scallop (75.8%–82.6%), sea squirt (78.6%–82.1%), short neck clam (71.2%–83.0%), and surf clam (74.9%–79.2%).

The results of heavy metal concentration by shellfish are shown in Table 6 and Fig. 2. In the food code, the maximum levels of heavy metal concentrations for shellfish in seafood are 2, 2, and 0.5 mg/kg for Pb, Cd, and Hg, respectively. The heavy metal concentrations in all the shellfish samples analyzed were lower than the maximum levels. Among heavy metals without a maximum level, Cu (maximum of 143 mg/kg) and Zn (maximum of 466 mg/kg) in oysters were relatively high.

In particular, the concentrations of Zn and Cu are detected higher than other heavy metals. This is because these substances accumulate relatively more than other heavy metals in the human body. Biswas et al. (2013) measured heavy metals in oysters and reported that Zn or Cu, along with Fe, may accumulate preferentially in the body compared to other heavy metals.

Comparing the results of this study with the results of other studies (Table 7), Zn and Cu showed higher concentration levels than other species. The concentrations of Zn (6.69 to 466 mg/kg) and Cu (0.0636 to 143 mg/kg) were similar to or higher than those of other studies (Zn: 3.34 to 217 mg/kg; Cu: 0.9 to 137 mg/kg). Furthermore, As (0.460 to 15.0 mg/kg) was higher than the Korean results (0.095 to 3.545 mg/kg). The samples with a high concentration of As are shellfish that mainly inhabit the bottom of the sea, such as horned turnban, ark shell clam, and short neck clam, which are considered to be influenced by the environment. However, it showed a lower concentration range than the foreign results (0.216 to 22.1 mg/kg). Moreover, Cd, Cr, Ni, Pb, and Hg were similar to or lower than the Korean and foreign results. The concentration of heavy metals was compared by the region where shellfishes were collected (Table 8). Cu and Zn, which have higher concentrations than other heavy metals, showed relatively higher concentrations in Jeonnam than in other regions. Also, high concentrations of Cu and Zn were detected in Gyeonggi and Incheon. All samples with high concentrations of Cu and Zn were oyster samples. In addition, heavy metals, except Cu and Zn, were detected in a similar range; thus, there is no significant difference by region.

We compared Korean and foreign maximum levels with the results of this study to evaluate the safety of shellfish produced along the Korean coast. The maximum concentration of Pb detected in shellfish in this study was 1.02 mg/kg, which was lower than that of Korea (2.0 mg/kg), EU (1.5 mg/kg), and Australian (2.0 mg/kg) levels. It was also higher than the Chinese maximum level of 1.0 mg/kg. Cd was detected at a maximum of 1.56 mg/kg, which was lower than the Korean, Chinese, and Australian maximum levels of 2.0 mg/kg; however, it exceeded the EU level of 1.0 mg/kg. Hg was detected at a maximum of 0.174 mg/kg, which was lower than all national and international standards. Although all national standards were satisfied with this result, it may have been detected at a higher concentration than some foreign standards, and continuous monitoring will be required in the future.

Risk assessment by calculating human body exposure

Human exposure was calculated for a total person average for all surveyors in the National Health and Nutrition Examination Survey (KHIDI, 2016) and an ingestion person average for only the ingestion person. The results are presented in Table 9. Since the human body exposure to all surveyors is an average value including the case of not ingesting, the human body exposure to only the ingestion person was further calculated to investigate the effect on the actual ingestion person. The

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Table 6. Heavy metal concentrations (mg/kg, ww) in shellfishes (abalone; ark shell clam; comb pen clam, horned turban; mussel; oyster; scallop; sea squirt; short neck clam; surf clam) collected from Korean coastal areas

Shellfish species	Value	Heavy metal (mg/kg, ww)								
		Pb	Cd	As	Cr	Cu	Ni	Zn	Ag	Hg
Abalone	Ave	0.0280	0.664	4.52	0.0346	2.39	0.665	18.8	0.102	0.0193
	Min	0.00470	0.231	2.22	< LOQ	1.06	0.0882	8.32	0.0296	0.00548
	Max	0.0741	1.36	7.89	0.0955	4.97	1.17	106	0.256	0.0582
Ark shell clam	Ave	0.105	0.304	3.382	0.065	0.671	0.030	14.552	0.053	0.027
	Min	0.078	0.108	1.843	0.035	0.468	0.001	12.708	0	0.015
	Max	0.149	0.441	5.775	0.078	0.806	0.050	16.991	0.153	0.038
Comb pen clam	Ave	0.0184	0.363	1.42	0.0262	0.291	0.00585	102	0.0379	0.0136
	Min	0.00892	0.202	1.14	< LOQ	0.0636	< LOQ	58.1	< LOQ	0.0130
	Max	0.0279	0.524	1.70	0.0524	0.517	0.0117	145	0.0758	0.0142
Horned turban	Ave	0.0103	0.215	7.63	0.0142	4.31	0.0357	21.5	0.150	0.0171
	Min	0.000750	0.0403	5.82	< LOQ	1.40	< LOQ	12.5	0.0972	0.00973
	Max	0.0274	0.749	15.0	0.0392	16.1	0.393	52.6	0.261	0.0325
Mussel	Ave	0.356	0.413	4.03	0.178	2.78	0.194	40.9	0.0247	0.0198
	Min	0.0582	0.152	1.76	< LOQ	0.675	< LOQ	12.8	< LOQ	0.00922
	Max	1.02	0.975	7.30	0.562	21.3	0.795	328	0.229	0.0436
Oyster	Ave	0.353	0.820	3.25	0.257	73.4	0.260	208	0.398	0.0368
	Min	0.0400	0.416	1.84	0.00505	17.0	< LOQ	41.8	0.113	0.0107
	Max	0.696	1.45	5.14	0.764	143	0.752	466	1.15	0.0560
Scallop	Ave	0.0223	1.09	0.880	0.0454	0.621	0.102	20.9	0.0478	0.0291
	Min	0.00485	0.478	0.460	0.00451	0.393	0.0591	14.1	0.00515	0.0183
	Max	0.0343	1.56	1.27	0.0890	0.915	0.131	34.1	0.103	0.0462
Sea squirt	Ave	0.108	0.146	1.78	0.0267	3.18	0.0411	67.3	0.121	0.0211
	Min	0.0711	0.0388	1.50	< LOQ	2.41	< LOQ	45.9	0.0414	0.00933
	Max	0.178	0.246	2.18	0.101	3.90	0.294	80.6	0.199	0.0553
Short neck clam	Ave	0.221	0.257	5.40	0.332	1.45	0.533	10.4	0.294	0.0382
	Min	0.0544	0.0964	2.73	< LOQ	0.721	0.0806	6.69	0.155	0.0176
	Max	0.588	0.510	8.77	0.820	1.96	1.46	13.5	0.920	0.174
Surf clam	Ave	0.293	0.149	1.687	0.308	1.395	0.354	16.769	0.027	0.030
	Min	0.054	0.181	2.322	0.032	1.067	0.356	12.548	0.038	0.022
	Max	0.548	0.257	2.208	0.712	2.090	0.678	29.288	0.009	0.032

Pb, lead; Cd, cadmium; As, arsenic; Cr, chromium; Cu, copper; Ni, nickel; Zn, zinc; Ag, silver; Hg, mercury; LOQ, limits of quantitation.

total amount of human body exposure for all surveyors in the National Health and Nutrition Examination Survey (KHIDI, 2016) is shown for each heavy metal substance. However, when only the ingestion person is targeted, the populations differ, and the total amount of human body exposure is calculated. Table 2 presents the health based guidance value of JECFA. Among the heavy metals investigated, PTWI of JECFA was suggested for Cd, Cu, Zn, Hg, and As. Moreover, there were no standards for other heavy metal species. Therefore, the risk assessment through comparison with PTWI could not be performed. Health based guidance value are expressed as provisional tolerable daily, weekly, or monthly intake, which were converted to weekly (Table 2) in this study. The human exposure to heavy metals calculated in this study was lower than the health based guidance value of JECFA. This means that the exposure of the human body to Cd, Cu, Zn, Hg, and As by ingestion of shellfish in Korean coastal area does not risk.

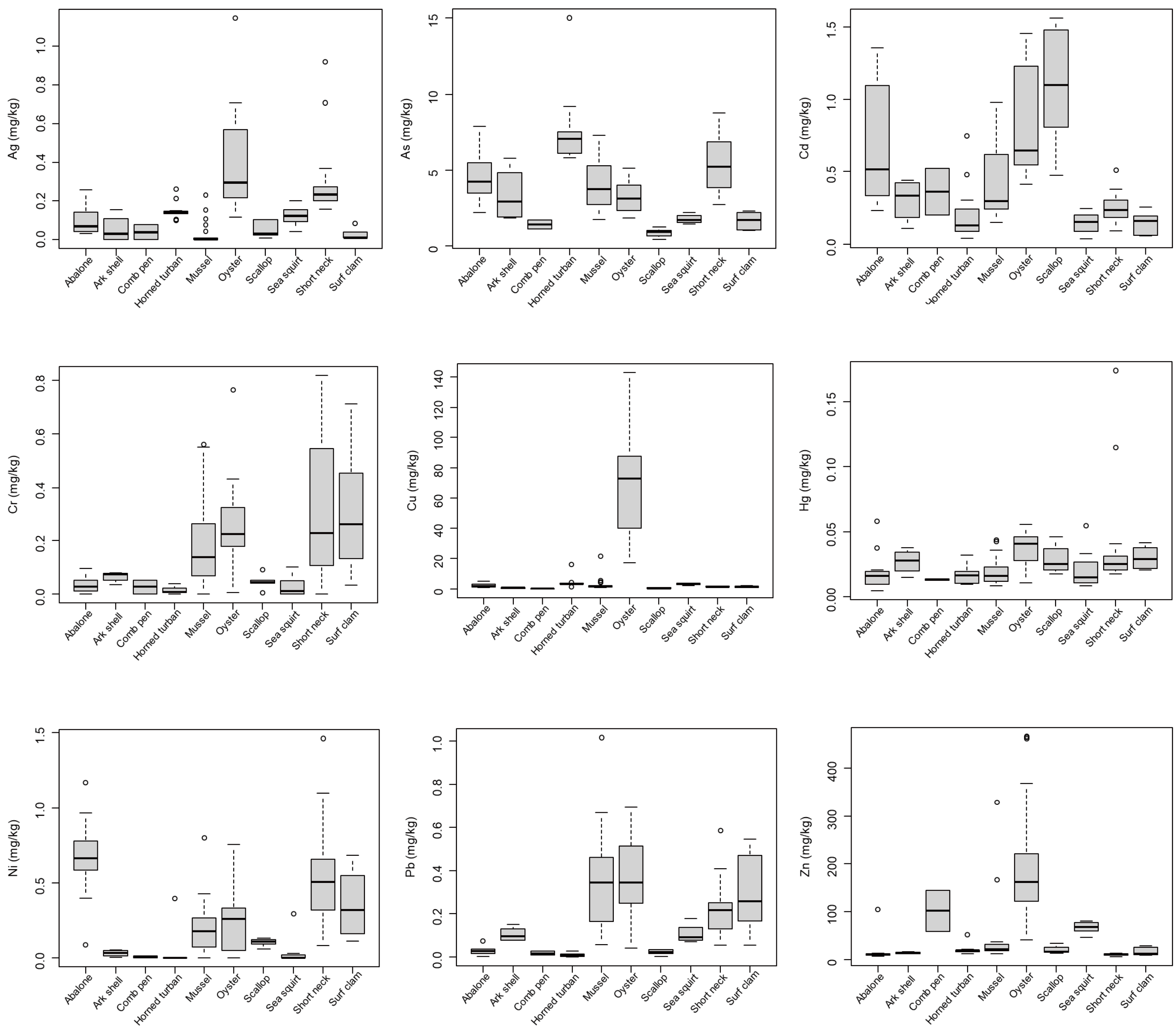

Fig. 2. Heavy metal concentrations (mg/kg, ww) in shellfishes (abalone; ark shell clam; comb pen clam, horned turban; mussel; oyster; scallop; sea squirt; short neck clam; surf clam) collected from Korean coastal area.

Conclusion

In this study, we analyzed heavy metals in the Korean coastal shellfish. We used this result for a safety evaluation. The heavy metals investigated in this study were Hg, Pb, Cd, As, Cr, Cu, Ni, Zn, and Ag.

Based on the analysis results, Zn and Cu were detected at an average of 56.7 mg/kg (6.70 to 466 mg/kg) and 13.2 mg/kg (0.064 to 143 mg/kg), respectively, compared with other heavy metal. Pb (average of 0.208 mg/kg, 0.000750 to 1.02 mg/kg), Cd (average of 0.454 mg/kg, 0.0388 to 1.56 mg/ kg), and Hg (average of 0.0266 mg/kg, 0.00548 to 0.174 mg/kg) were detected. Additionally, As, Cr, Ni, and Ag with average concentrations of 4.02 (0.460 to 15.0 mg/kg), 0.167 (< LOQ to 0.820 mg/kg), 0.281 (< LOQ to 1.46 mg/kg), and 0.158 mg/kg (< LOQ to 1.15 mg/kg) were detected. Region-specific, shellfish, except that Cu and Zn were detected relatively high in oysters. No significant difference was found.

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Table 7. Comparision of heavy metal concentrations (mg/kg, ww) of shellfishes from Korea and foreign countries

Country	Heavy metal (mg/kg, ww)								Reference
	Pb	Cd	As	Cr	Cu	Ni	Zn	Hg	
Korea	0.761–1.708	0.168–0.247	–	0.682–1.645	3.914–6.484	0.785–1.998	40.36–77.98	–	Hwang et al., 2001
Korea	0.763–1.306	0.128–0.298	–	0.648–1.346	8.89–27.48	0.810–1.856	97.9–217.0	–	Hwang et al., 2002
Korea	0.045–0.210	0.198–2.654	–	ND	1.385–9.985	–	–	–	Kim et al., 2002
Korea	Trace–0.984	0.030–0.617	–	Trace–0.849	–	–		0.002–0.020	Kim et al., 2003
Korea	ND–1.38	ND–1.85	–	–	–	–	–	ND–0.19	Ham, 2002
Korea	0.5471–1.1571	0.1748–0.3529	–	0.4280–0.9300	–	–	–	0.0057–0.0203	Ha & Song, 2004
Korea	0.061–0.246	0.010–0.256	0.095–3.545	–	–	–	–	0.002–0.052	KCA, 2011
India	ND	0.1	–	0.9	6.5	0.8	15.7	ND	Sankar et al., 2006
Greece	0.68	0.34	–	0.26	1.4	0.34	40	0.024	Copat et al., 2013
China	0.033–0.243	0.027–0.329	0.216–0.807	–	1.31–9.11	–	3.34–17.90	0.006–0.022	Huang et al., 2007
Malaysia	0.18–0.88	0.04–5.45	0.95–22.10	0.47–3.39	–	0.37–2.23	–	–	Sharif et al., 2016
Egypt	0.2–17	0.04–1.7	–	–	0.9–137	7.8–41	41–192	ND–0.2	Nemr et al., 2016

Pb, lead; Cd, cadmium; As, arsenic; Cr, chromium; Cu, copper; Ni, nickel; Zn, zinc; Ag, silver; Hg, mercury; ND, not detected.

Table 8. Heavy metal concentrations (mg/kg, ww) by sampling regions from Korean coastal areas

Sampling regions	Value	Heavy metal (mg/kg, ww)								
		Pb	Cd	As	Cr	Cu	Ni	Zn	Ag	Hg
Gangwon	Ave	0.120	0.836	3.22	0.0573	2.290	0.182	23.3	0.0489	0.0256
	Min	0.00485	0.475	0.460	< LOQ	0.393	< LOQ	10.7	< LOQ	0.0114
	Max	0.593	1.56	15.0	0.211	16.1	0.670	52.6	0.142	0.0462
Gyeongbuk	Ave	0.262	0.402	3.60	0.109	2.27	0.153	65.4	0.0777	0.0210
	Min	0.0711	0.0388	1.50	< LOQ	0.802	< LOQ	16.2	< LOQ	0.00933
	Max	0.669	0.975	7.30	0.562	3.90	0.795	328	0.199	0.0553
Ulsan	Ave	0.448	0.265	2.87	0.293	4.51	0.201	24.7	0.00470	0.0382
	Min	0.433	0.264	2.70	0.285	4.46	0.185	19.6	0.00440	0.0334
	Max	0.463	0.266	3.05	0.301	4.56	0.216	29.8	0.00499	0.0431
Busan	Ave	0.585	0.262	3.73	0.461	2.92	0.414	26.3	0.0168	0.0235
	Min	0.348	0.195	1.90	0.265	1.82	0.202	18.1	0.00225	0.0127
	Max	1.02	0.316	6.07	0.712	5.51	0.678	33.4	0.0744	0.0361
Gyeongnam	Ave	0.148	0.312	3.54	0.0662	9.77	0.0744	56.0	0.119	0.0136
	Min	0.0582	0.108	1.76	< LOQ	0.468	< LOQ	12.8	< LOQ	0.00922
	Max	0.265	0.645	5.77	0.186	55.6	0.195	185	0.675	0.0250
Jeonnam	Ave	0.247	0.605	4.03	0.256	27.3	0.426	77.4	0.244	0.0276
	Min	0.00845	0.0964	1.14	< LOQ	0.0636	< LOQ	6.69	< LOQ	0.00548
	Max	0.696	1.45	8.77	0.820	143	0.962	466	1.15	0.0560
Jeonbuk	Ave	0.163	0.133	2.66	0.191	1.25	0.302	11.0	0.128	0.0296
	Min	0.0845	0.0586	1.05	0.0903	1.08	0.161	9.97	0.0105	0.0183
	Max	0.262	0.207	4.90	0.343	1.66	0.461	12.4	0.253	0.0420
Chungnam	Ave	0.105	0.265	4.50	0.157	1.99	0.420	11.1	0.231	0.0291
	Min	0.00129	0.127	2.22	< LOQ	1.12	< LOQ	7.69	0.0310	0.0171
	Max	0.387	0.480	7.34	0.806	3.62	1.17	18.2	0.709	0.0582
Gyeonggi	Ave	0.0646	0.419	3.61	0.171	55.2	0.124	236	0.432	0.111
	Min	0.0400	0.292	1.99	< LOQ	0.917	0.0407	10.1	0.155	0.0484
	Max	0.0893	0.546	5.23	0.342	110	0.208	462	0.709	0.174
Incheon	Ave	0.166	0.445	5.58	0.0983	30.5	0.727	126	0.242	0.0603
	Min	0.0515	0.263	1.84	0.0113	0.721	0.0485	12.0	0.186	0.0366
	Max	0.257	0.625	8.16	0.171	118	1.46	464	0.368	0.115
Jeju	Ave	0.00837	0.126	6.92	0.0144	3.29	< LOQ	19.2	0.154	0.0148
	Min	0.00075	0.0403	5.82	0.00130	2.76	< LOQ	15.8	0.0972	0.0097
	Max	0.0172	0.305	9.18	0.0392	4.24	< LOQ	21.1	0.261	0.0284

Pb, lead; Cd, cadmium; As, arsenic; Cr, chromium; Cu, copper; Ni, nickel; Zn, zinc; Ag, silver; Hg, mercury; LOQ, limits of quantitations.

Table 9. PTWI of heavy metals from shellfish consumption by adults living in Korea

	Shellfish species	PTWI (μg/kg b.w./week)								
		Pb	Cd	As	Cr	Cu	Ni	Zn	Ag	Hg
Total person average	Abalone	1.35×10^{-3}	3.20×10^{-2}	2.18×10^{-1}	1.67×10^{-3}	1.16×10^{-1}	3.21×10^{-2}	9.05×10^{-1}	4.92×10^{-3}	9.29×10^{-4}
	Ark shell clam	6.07×10^{-4}	2.13×10^{-3}	3.76×10^{-2}	3.99×10^{-4}	4.48×10^{-3}	9.38×10^{-5}	1.18×10^{-1}	8.16×10^{-4}	1.54×10^{-4}
	Comb pen clam	2.22×10^{-5}	4.37×10^{-4}	1.71×10^{-3}	3.15×10^{-5}	3.50×10^{-4}	7.05×10^{-6}	1.23×10^{-1}	4.56×10^{-5}	1.64×10^{-5}
	Horned turban	1.38×10^{-4}	2.88×10^{-3}	1.02×10^{-1}	1.89×10^{-4}	5.77×10^{-2}	4.78×10^{-4}	2.88×10^{-1}	2.01×10^{-3}	2.29×10^{-4}
	Mussel	5.65×10^{-2}	6.54×10^{-2}	6.39×10^{-1}	2.83×10^{-2}	4.40×10^{-1}	3.07×10^{-2}	6.49	3.92×10^{-3}	3.14×10^{-3}
	Oyster	4.23×10^{-2}	9.84×10^{-2}	3.89×10^{-1}	3.08×10^{-2}	8.80	3.12×10^{-2}	2.50×10^{1}	4.78×10^{-2}	4.42×10^{-3}
	Scallop	1.49×10^{-4}	7.27×10^{-3}	5.89×10^{-3}	3.04×10^{-4}	4.16×10^{-3}	6.80×10^{-4}	1.40×10^{-1}	3.20×10^{-4}	1.95×10^{-4}
	Sea squirt	3.28×10^{-3}	4.43×10^{-3}	5.43×10^{-2}	8.13×10^{-4}	9.69×10^{-2}	1.25×10^{-3}	2.05	3.69×10^{-3}	6.44×10^{-4}
	Short neck clam	3.02×10^{-2}	3.50×10^{-2}	7.37×10^{-1}	4.53×10^{-2}	1.98×10^{-1}	7.28×10^{-2}	1.41	4.01×10^{-2}	5.22×10^{-3}
	Surf clam	1.50×10^{-5}	5.69×10^{-6}	8.05×10^{-5}	1.42×10^{-5}	6.92×10^{-5}	1.20×10^{-5}	7.17×10^{-4}	2.36×10^{-6}	2.21×10^{-6}
	Sum	1.35×10^{-1}	2.48×10^{-1}	2.18	1.08×10^{-1}	9.72	1.69×10^{-1}	3.65×10^{1}	1.04×10^{-1}	1.50×10^{-2}
Ingestion person average	Abalone	5.77×10^{-2}	1.37	9.33	7.14×10^{-2}	4.95	1.37	3.88×10^{1}	2.11×10^{-1}	3.98×10^{-2}
	Ark shell clam	2.98×10^{-1}	1.04	1.84×10^{1}	1.96×10^{-1}	2.20	4.61×10^{-2}	5.81×10^{1}	4.00×10^{-1}	7.55×10^{-1}
	Comb pen clam	1.58×10^{-2}	3.11×10^{-1}	1.22	2.25×10^{-2}	2.49×10^{-1}	5.02×10^{-3}	8.73×10^{1}	3.25×10^{-2}	1.17×10^{-2}
	Horned turban	7.26×10^{-3}	1.52×10^{-1}	5.39	1.00×10^{-2}	3.05	2.53×10^{-2}	1.52×10^{1}	1.06×10^{-1}	1.21×10^{-2}
	Mussel	8.77×10^{-1}	1.02	9.92	4.39×10^{-1}	6.84	4.77×10^{-1}	1.01×10^{2}	6.08×10^{-2}	4.87×10^{-2}
	Oyster	1.49	3.46	1.37×10^{1}	1.08	3.09×10^{2}	1.10	8.78×10^{2}	1.68	1.55×10^{-1}
	Scallop	7.88×10^{-2}	3.84	3.11	1.60×10^{-1}	2.20	3.59×10^{-1}	7.39×10^{1}	1.69×10^{-1}	1.03×10^{-1}
	Sea squirt	8.47×10^{-1}	1.15	1.40×10^{1}	2.10×10^{-1}	2.50×10^{1}	3.24×10^{-1}	5.30×10^{2}	9.53×10^{-1}	1.66×10^{-1}
	Short neck clam	2.36×10^{-1}	2.74×10^{-1}	5.78	3.55×10^{-1}	1.56	5.70×10^{-1}	1.11×10^{1}	3.15×10^{-1}	4.09×10^{-2}
	Surf clam	1.60×10^{-2}	6.08×10^{-3}	8.61×10^{-2}	1.52×10^{-2}	7.40×10^{-2}	1.28×10^{-2}	7.67×10^{-1}	2.52×10^{-3}	2.37×10^{-3}

Pb, lead; Cd, cadmium; As, arsenic; Cr, chromium; Cu, copper; Ni, nickel; Zn, zinc; Ag, silver; Hg, mercury; PTWI, provisional tolerance weekly intake.

Most heavy metal materials were detected with a concentration tendency similar to that of other studies. The heavy metal results of Korean coastal shellfish met all Korean maximum levels. However, Pb and Cd exceed some foreign standards as the maximum detection concentration standard, and continuous monitoring in the future is considered necessary. The human body exposure calculated using the congestion intake and heavy metal detection concentrations was less than the JECFA human safety standard, and the risk of heavy metals due to shellfish intake was at an acceptable level. The results of this survey will be used as basic data for heavy metal pollution in the Korean coastal shellfish. It can also be used as policy data for the safe seafood supply.

Competing interests

No potential conflict of interest relevant to this article was reported.

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Availability of data and materials

Upon reasonable request, the datasets of this study can be available from the corresponding author.

Ethics approval and consent to participate

This article does not require IRB/IACUC approval because there are no human and animal participants.

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