Heavy metals (Cd, Pb, Cu, Zn) concentrations in edible bivalves harvested from Northern Coast of Central Java, Indonesia

Heavy metals (Cd, Pb, Cu, Zn) concentrations in edible bivalves harvested from Northern Coast of Central Java, Indonesia

B Yulianto[1], P S Oetari[2], S Februhardi[3], T W C Putranto[4] and A Soegianto[4, a]

[1] Department of Marine Science, Faculty of Fisheries and Marine Sciences, Diponegoro University, Kampus Undip Tembalang, Semarang, Indonesia
[2] CV. Mitra Adi Pranata, Environmental Consultant, Semarang, Indonesia
[3] Research and Development Board of Central Java, Semarang, Indonesia
[4] Department of Biology, Faculty of Sciences and Technology, Universitas Airlangga, Surabaya, Indonesia

[a] agoes_soegianto@unair.ac.id

Abstract. This study measured the concentration of Cd, Pb, Cu and Zn in bivalves captured from the northern coast of Central Java, Indonesia. We also evaluated the hazard level posed by the metals in relation to the maximum acceptable limit (MAL) for human consumption and to estimate the weekly intake and compare it with the provisional tolerable weekly intake. The highest Cd levels were recorded in *Amusium pleuronectes* (16.32 mg Cd/kg) and the lowest levels of Cd were observed in *Perna viridis* (0.18 mg Cd/kg*). A. granosa* contained the highest level of Pb (9.050 mg Pb/kg) and *P. viridis* contained the lowest level of Pb (1.07mg Pb/kg). The lowest Cu level was observed in *A. granosa* (1.49 mg Cu/kg) and the highest Cu level was noted in *C. gigas* (86.21 mg Cu/kg). The levels of Zn in all species were relatively higher compared to other metals, ranged from 33.56 to 846.81 mg Zn/kg. The Cd level in bivalves was below the MAL of WHO and FAO (2 mg/kg), except in A. pleuronectes from Tegal city. The level of Pb in bivalves was exceeded the MAL value of the various existing authorities. Cu level in *C. gigas* (59.2 mg Cu/kg) exceeded the MAL of Indonesian authority and WHO. Zn levels in *C. gigas*, *A. pleuronectes* and *A. granosa* exceeded the MAL of various authorities. In order to avoid the impact of metals on people health, then safely weight of bivalves were allowed to be consumed were 0.10 kg/week for *A. granosa*, 0.10 kg/week for *C. gigas*, 0.16 kg/week for *M. lyrata*, 0.36 kg.week for *P. viridis* and 0.03 kg/week for *A. pleuronectes*.

1. Introduction

The north coast of Central Java, Indonesia, is one of the most populated regions in Java island, even in Indonesia. A number of human activities such as industries, settlements, and harbors exist along the coastal line of this area. Consequently, the coastal waters of this area potentially receive various pollutants including heavy metals from industrial and domestic wastes. The presence of heavy metals can pose a threat to aquatic organisms. Some heavy metals (i.e. Cu, Zn, and Mo) are essential for living organisms, because they are required in some activities for the body metabolism; while some others (i.e. Cd, Pb, Hg, As) are non-essential metals, which have no important role for biological needs of the organism body [1].

However, both metals are very dangerous for aquatic life if their concentration exceeds the threshold levels for organism [2]. Various studies concerning heavy metals have been carried out along the northern coast of Java island, for examples: the levels of Cd, Cu, Pb, Zn, and Ni in sediment of the

Jakarta Bay (western part of Java island) [3], the concentrations of heavy metals in fish from North Coast of Central Java [4-5], fractionation of Pb, Cd, Cu and Zn in sediment and its bioavailability to aquatic biota in Jakarta Bay [6], content of Hg and Cd in water, sediment and blood mussels (*Anadara granosa*) [4], and Hg concentration in edible fish harvested from Gresik Indonesia [7]. The results of these studies showed that the levels of metals tended to increase both in media (water and sediment) and aquatic animals along the north coast of Java. Among aquatic animals bivalve is considered as a sentinel or indicator species for trace metals [2]. In present study, some bivalve species were chosen due to their strongly capacity to bio-accumulate the metals that can reflect the metal concentrations in coastal environment. Considering the bivalves to be one kind of food containing high protein and widely consumed by the people, abundant availability and low prices, making bivalves to be very important fishery product in Indonesia. Therefore, it is important to conduct monitoring and research of heavy metals (Cd, Cu, Pb, Zn) in various species of bivalves harvested from the north coast of Central Java, in order to protect the community from the potential hazard of heavy metals when consuming the bivalves.

2. Materials and methods

2.1. Sampling location

Samples of bivalves were collected from 12 regencies or cities along the northern coast of Central Java. Sampling locations were determined to be at the embouchure (mouth of river) and their surrounding coastal waters. This location is considered as the entrance of all discharges of pollutants (including heavy metals) from the mainland towards the coastal areas and the sea. The 12 sampling locations were specified as Pati Regency (1), Jepara Regency (2), Demak Regency (3), Semarang City (4), Kendal Regency (5), Batang Regency (6), Pekalongan Regency (7), Pekalongan City (8), Pemalang Regency (9), Tegal Regency (10), Tegal City (11), and Brebes Regency (12). Two sampling stations were established at each location. Locations of the study and sampling stations are presented in Figure 1.

2.2. Bivalves samples

Five species of bivalves, blood mussel (*Anadara granosa* Linnaeus 1758), green mussel (*Perna viridis* Linnaeus 1758), oyster (*Crassostrea gigas* Thunberg 1793), Asiatic hard clam (*Meretrix lyrata* Sowerby, 1851), and Asian moon scallop (*Amusium pleuronectes*, Linnaeus 1758) were collected from 12 locations in northern coast of Central Java, Indonesia during June-July 2009. The species selected was based on general abundance found in each location, such as their size and their potential to be consumed by local people. The bivalves were caught by using dredges operated from a boat and also captured directly by hand. All of the samples were kept in cool box at a temperature maintained of about 4ºC prior to transporting to the laboratory. The information of sample location and number of animal collected is shown in Figure 1.

2.3. Metals analyses

Analysis of metals (Cd, Pb, Cu and Zn) was performed only of the soft tissues of each bivalve species. Sample of soft tissues washed, dried and pulverized with a mortar, then sieved until pass 100 mesh size and homogenized. Homogenized tissue samples, weighed 0.5 g in a teflon bomb digester, moistened with splashes aquatrides, then add 1 ml of concentrated HNO3. After that, teflon bomb digester sealed then put in the furnace and heated at a temperature of 150º C for 4 hours. The samples, then, was poured into a beaker glass and heated on an electric heater with the addition aquatrides repeatedly. The result, after being cold, put in volumetric flask 10 ml and samples ready for analysis of heavy metals by Atomic Absorption Spectrophotometer (AAS, Shimadzu AA-6200). Before measuring the concentration of heavy metals using AAS, the first, checked the optimum conditions of analysis of each element by a calibration curve of Cd, Pb, Cu and Zn. Standard solution of each heavy metal needed to be made to make a calibration curve of the heavy metals to be measured. Standard solution prepared from a stock solution of each metal, were made with a concentration of 1000 mg/L. Range of standard solutions of each metal is Pb from 0.5 to 10 mg/L, Cd from 0.1 to 20 mg/L, Cu from 0.1 to 100 mg/L, and Zn from

0.1 to 1000 mg/L. A calibration curve was obtained by creating a curve between the concentrations against absorption of each metal element. Once all parameters are met, then the measurement of heavy metal content in the samples can be performed. All samples' concentrations of metals were quoted as mg/kg wet weight. The accuracy of the Cd determination was verified using dogfish muscle reference material (DORM-2) provided by the National Research Council of Canada (Ottawa).

2.4. Data analyses

All the data obtained, such as the concentration of heavy metals in water, sediment, and bivalves were analyzed descriptively. This analysis was selected considering the species of bivalves found among the study sites is different. Also, the difference in the sampling period, that is not due to treatment considerations, but rather the inability of sampling performed on all study sites at the same time. Therefore, the data analysis of heavy metals content of various species of bivalves are interpreted by comparing it to threshold standards issued by various authorities (Indonesia and International), both state and the competent institutions on the environment.

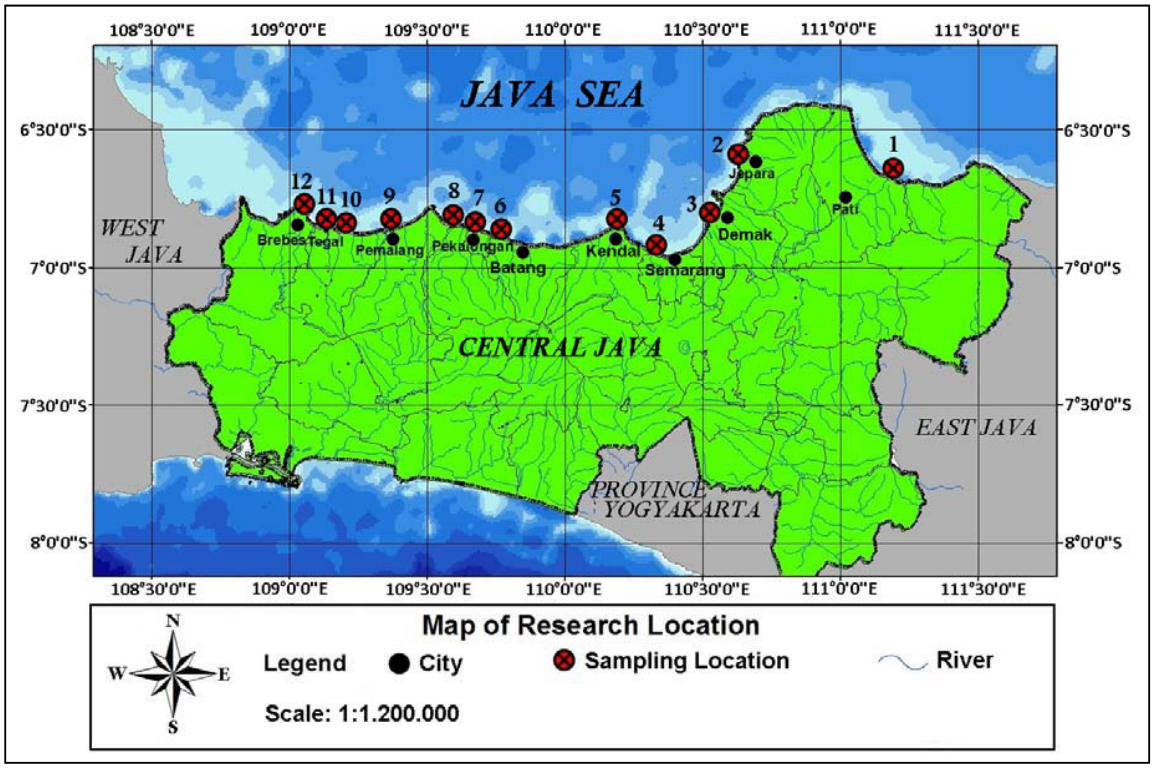

Figure 1. Twelve sampling locations at northern coast of Central Java, Indonesia

3. Results and discussion

Five species of bivalves, i.e: *Anadara granosa, Perna viridis, Crassostrea gigas, Meretrix lyrata*, and *Amusium pleuronectes* were collected from northern cost of Central Java (Table 1). The number of species of bivalves was varied among sampling locations. The difference in species captured in such location was due to the typical endemic of bivalves species which live in respectively location. The variations of heavy metal concentrations (Cd, Pb, Cu and Zn) in bivalves captured from northern coast of Central Java in June and July 2009 were shown in Table 2. Since bivalves captured from all the stations consisted from different species, they were grouped based on each species. *Anadara granosa*

dominated the species captured from most locations of sampling. They were found at 5 locations (10 stations) dispersed on five regencies/cities, i.e.: Juwana (Regency of Pati), Morodemak (Regency of Demak), Banjirkanal (Semarang City), Sambong (Regency of Batang), and Pemali (Regency of Brebes) (Table 2).

The present study indicated that heavy metal levels in tissues demonstrated a high variation among all bivalve species, metals, and locations. The concentrations of the investigated metals in the soft tissues of *A. granosa* decreased in the following order: Zn>Cu>Pb>Cd. The concentration of Zn ranged between 33.56-194.97 mg/kg (with the mean was 100.15±50.83 mg Zn/kg). While, the concentration of Cu, Pb, and Cd in the soft tissues of *A. granosa* ranged between 1.61-14.77 mg Cu/kg (with the mean was 6.94±4.33 mg Cu/kg), 2.97-9.05 mg Pb/kg (with the mean was 5.24±1.95 mg Pb/kg), and 0.41-2.58 mg Cd/kg (with the mean was 1.44±0.70 mg Cd/kg), respectively.

Crassostrea gigas were captured from 6 stations dispersed on three regencies (cities) (i.e.: Kendal regency, Pekalongan city, and Pemalang regency). The concentrations of the metals in the soft tissues of *C. gigas* decreased in the following order: Zn>Cu>Pb>Cd. The concentration of Zn ranged between 366.64-846.81 mg Zn/kg (with the mean was 620.14±222.95 mg Zn/kg). While, the concentration of Cu, Pb, and Cd in the soft tissues of *C. gigas* ranged between 45.94-86.21 mg Cu/kg (with the mean was 59.22±15.29 mg Cu/kg), 3.93-8.09 mg Pb/kg (with the mean was 5.76±1.43 mg Pb/kg), and 0.89-2.98 mg Cd/kg (with the mean was 1.92±0.88 mg Cd/kg), respectively.

Table 1. Bivalve species captured in such locations of study and number of samples collected from northern coastal waters of Central Java, Indonesia according to sampling period in June-July 2009. N: number of animal; - : not sampled

Species	June		July	
	Capture Location	N	Capture Location	N
Anadara granosa	Juwana, Regency of Pati	32	Sambong, Regency of Batang	28
	Morodemak, Regency of Demak	37	Pemali, Regency of Brebes	31
	Banjirkanal, City of Semarang	26	-	-
Perna viridis	Demakan, Regency of Jepara	27	-	-
Crassostrea gigas	Wakak, Regency of Kendal	21	Loji, City of Pekalongan	22
	-	-	Ulujami, Regency of Pemalang	25
Meretrix lyrata	-	-	Pencongan, Regency of Pekalongan	38
	-	-	Wonokerto, Regency of Pekalongan	42
	-	-	Ketiwon Regency of Tegal	38
Amusium pleuronectes	-	-	Sibelis, City of Tegal	32

Meretrix lyrata were found at 4 stations from 2 regencies (i.e.: Pekalongan regency and Tegal regency). The concentrations of the metals in the soft tissues of *M. lyrata* decreased in the following order: Zn>Cu>Pb>Cd. The concentration of Zn ranged between 68.03-128.43 mg/kg (with the mean was 96.38±31.21 mg/kg). While, the concentration of Cu, Pb, and Cd in the soft tissues of *M. lyrata* ranged between 4.80-17.79 mg/kg (with the mean was 11.08±6.68 mg/kg), 1.17-4.86 mg/kg (with the mean was 2.93± 1.99 mg/kg), and 0.48-0.66 mg/kg (with the mean was 0.56±0.08 mg/kg), respectively.

Perna viridis and *Amusium pleuronectes* were only captured from 1 location, Demakan (Jepara Regency) and Sibelis (Tegal City), respectively. The mean concentration of Zn, Cu, Pb, and Cd in the soft tissue of *P. viridis* was respectively, 82.65±3.28 mg/l, 10.88±0.11 mg/l, 1.27±0.28 mg/l, and 0.19±0.02 mg/l. Meanwhile, the mean concentration of Zn, Cu, Pb, and Cd in the soft tissue of *A.*

pleuronectes was 433.71±6.82 mg Zn/l, 7.45±1.57 mg Cu/l, 2.00±0.07 mg Pb/l, and 15.34±1.39 mg Cd/l.

Table 2. Concentrations of cadmium (Cd), lead (Pb), copper (Cu), and zinc (Zn) in the soft tissues of bivalves (in mg/kg), from 24 stations dispersed at 12 regencies/cities along northern coast of Central Java, Indonesia.

Species	Locations	Heavy Metal (in mg/kg)			
		Cd	Pb	Cu	Zn
Anadara granosa	Juwana 1	2.58	4.12	10.38	173.32
	Juwana 2	1.93	6.73	9.56	194.97
	Morodemak 1	1.19	9.05	5.31	80.93
	Morodemak 2	0.90	7.40	4.60	62.79
	Banjirkanal Barat 1	1.71	4.37	10.97	92.40
	Banjirkanal Barat 2	2.25	5.55	14.77	119.47
	Sambong 1	1.52	4.07	1.49	53.96
	Sambong 2	1.27	4.89	1.61	33.56
	Pemali 1	0.41	2.97	4.70	88.27
	Pemali 2	0.60	3.22	5.99	101.78
	Mean ± SD	**1.44 ± 0.70**	**5.24 ± 1.95**	**6.94 ± 4.33**	**100.15 ± 50.83**
Crassostrea gigas	Wakak 1	2.07	4.84	52.11	792.67
	Wakak 2	1.87	3.93	46.51	846.81
	Loji 1	0.94	5.45	58.12	366.64
	Loji 2	0.89	5.77	45.94	370.20
	Ulujami 1	2.98	6.50	66.41	804.99
	Ulujami 2	2.76	8.09	86.21	539.55
	Mean ± SD	**1.92 ± 0.88**	**5.76 ± 1.43**	**59.22 ± 15.30**	**620.14 ± 222.95**
Meretrix lyrata	Pencongan 1	0.66	4.86	4.80	68.03
	Pencongan 2	0.52	4.45	5.89	71.26
	Ketiwon 1	0.56	1.17	15.82	128.43
	Ketiwon 2	0.48	1.25	17.79	117.81
	Mean ± SD	**0.56 ± 0.08**	**2.93 ± 1.99**	**11.08 ± 6.68**	**96.38 ± 31.21**
Perna viridis	Demakan 1	0.18	1.46	10.80	73.26
	Demakan 2	0.21	1.07	10.96	92.04
	Mean ± SD	**0.195 ± 0.02**	**1.27 ± 0.28**	**10.88 ± 0.11**	**82.65 ± 13.28**
Amusium pleuronectes	Sibelis 1	16.32	1.95	6.34	428.88
	Sibelis 2	14.36	2.05	8.56	438.53
	Mean ± SD	**15.34 ± 1.39**	**2.00 ± 0.071**	**7.45 ± 1.57**	**433.71 ± 6.82**

Bivalves are marine animals with low mobility, thus their existence in an area may represent water quality conditions in which they lives. For this reason, bivalves were used as animal samples in this study in order to observe the water quality conditions at each location and also the safety level of the animals themselves as a perishable fishery product from heavy metal pollutants. Average values of metals

concentration in bivalves from northern coast of Central Java showed: 0.56-15.34 mg Cd/kg, 1.27-5.76 mg Pb/kg, 6.94-59.22 mg Cu/kg, and 82.38-620.14 mg Zn/kg (Table 2). The average metal concentrations in the soft tissues of bivalves decreased in the following order: Zn>Cu>Pb>Cd. In the case of cadmium, a higher Cd level was noted on soft tissue of *Amusium pleuronectes* from Sibelis River at Tegal city compared to other locations. This higher level could be due to the sampling stations where *A. pleuronectes* lives were placed at Port of Tegal which is a semi-enclosed area encountered by a concrete sea wall that caused blockage in the exchange process between the seawater in the port with the open seawater. This condition resulted in the high accumulation of cadmium in the port basin.

With lead, high levels of lead found in *A. granosa* and *C. gigas*, with a concentration greater than 5 mg Pb/kg. According to the observation, the location of the sampling stations *A. granosa* are a traffic lanes and fishing boat anchoring area (i.e: Juwana, Morodemak, and Sambong), brass handicrafts industry (i.e: Juwana) and areas with chemical industrial activities (i.e: Banjirkanal Barat), as well as the activity of washing of jeans (Pemali). Meanwhile, *C. gigas* was captured on the area of plywood industry (i.e. Wakak) and batik garment industry area (i.e. Loji) and washing of jeans (i.e. Ulujami). High lead concentrations are evident in association of high concentrations with urban areas near cities with many human activities as like as industry, transportation, etc. There is a strong statistical correlation between human population density and lead concentrations in oysters and mussels [8].

A high concentration of copper was found in the tissue of *C. gigas*, with a concentration reached >59 mg Cu/kg at the station of Wakak, Loji, and Ulujami. High Cu in coastal waters and estuary of Wakak, can be derived from timber preservative treatments in which the main element used is copper. CCA (Copper, Chromium and Arsenic) is intended to protect wood against pests such as decay fungi, wood boring insects or marine borers that can threaten the integrity of wood products.

Moreover, copper is used to control fungi and marine borers, arsenic to control insects and some copper-resistant fungi, and chromium to fix the copper and arsenic in the wood [9]. Based on our findings, there were certain locations with higher levels of metals accumulated in bivalves compared other areas, i.e. *C. gigas* captured from Wakak (Kendal Regency), Loji (Pekalongan City), and Ulujami (Pekalongan Regency). These locations are known as the center of textile and garment industry (Loji and Ulujami) and the one large timber industry (plywood) (Wakak). Both have the potential discharge of heavy metal waste into coastal waters that may be highly accumulated in *C. gigas*. Caution needs to be taken to the Pekalongan City, and Pekalongan Regency which is known as the region with the extensive garment industry (batik and jeans), whether conducted by large industry, nor home industry scale. By observations, the large garment industries generally have a Waste Processing Installation. While, the home industries do not have. This is why, the discharge of waste from the remnants of production tended to be less controlled, and pollute inshore environment, particularly metal contaminants.

Table 3. Maximum Acceptable Limits of cadmium, lead, copper, and zinc ($mg.kg^{-1}$) in marine animals released by some authorities.

Country and Organization	Maximum Acceptable Limit			
	Cd	Pb	Cu	Zn
Indonesia[a]	-	2	20	100
FAO[b]	2	0.3	-	-
WHO[10]	2	5	10	100
European Union[c]	1.0	1.5	-	-
FDA USA[11]	0.2	1.5	100	150

[a] Decree of General Direction of Drug and Food Surveillance No 03725/B/SK/89 concerning acceptable limit of metals in food

[b] FAO : Codex General Standard For Contaminants And Toxins In Food And Feed. Codex Standard 193-1995

[c] European Commission Regulation (EC No 1881/2006**)** concerning setting maximum levels for certain contaminants in foods (mg/kg wet weight)

Using MAL (maximum acceptable limits) from the WHO and FAO (Table 2), the level of Cd in bivalves from the north coast of Java is still below the quality standard (2 mg/kg), except on *Amusium pleuronectes* from Tegal city has exceeded the value of MAL. The use of quality standards of the European Union and the USA FDA will give a different conclusion, given the quality standards of both authorities are much lower than the FAO and WHO. In general, the level of Pb in bivalves from the northern coast of Java is already exceeded the MAL value of the various existing authorities (WHO and Indonesia) (Table 3). High Cu level that almost 3-6 fold exceeded the value of MAL (Indonesian and WHO) was recorded in *Crassostrea gigas* (59.2 mg Cu/kg), but slightly high found in *M. lyrata* and *P. viridis*. Meanwhile, the level of Zn has far exceeded the standard of MAL was found in *C. gigas* (more than 6-fold), and *A. pleuronectes* (more than 4-fold), but slightly exceeded on *A. granosa*. Some important notes in relation with the high content of heavy metals in bivalves in North Coast of Central Java needs to be taken, such as the Cd content in *A. pleuronectes* at Tegal City (15.34 mg/kg), Pb in *A. granosa* and *C. gigas* (5.24 mg/kg and 5.76 mg/kg), copper in *C. gigas* (59.22 mg/kg), and zinc in *C. gigas* and *A. pleuronectes* (620.14 mg/kg and 433.71 mg/kg). The content of the heavy metals in those species have exceeded the maximum acceptable limit released by some authorities (Table 3).

A health based guidance value for cadmium of 0.007 mg/kg body weight per week (Provisional Tolerable Weekly Intake/PTWI) was established by the Joint FAO/WHO Expert Committee on Food Additives and endorsed by the Scientific Committee for Food. With the value of the PTWI for cadmium of 0.007 mg Cd/kg body weight, people with a weight of 65 kg can't tolerate more than 0.455 mg Cd input into his body in a week (Table 4). The concentrations of Cd in *A. granosa* on the North Coast of Central Java was 1.44 mg/kg. People must not consume blood mussel *A. granosa* >0.32 kg in a week. With the same method, then people should not consume oyster: >0.24 kg/week (for *C. gigas*), >0.81 kg/week (for *M. lyrata*), >2.39 kg/week (for *P. viridis*), and>0.03 kg/week (for *A. pleuronectes*). The regulation of PTWI for lead is 0.007 mg/kg body weight [12]. For people with a weight of 65 kg should not be intruding more than 0.46 mg Pb in 1 week. Therefore, on the northern coast of Central Java, people should not consume blood mussel (*A. granosa*) and oyster (*C. gigas*) >0.10 kg and >0.10 kg, respectively. While, the people are allowed to consume *M. lyrata*, green mussel (*P. viridis*) and *A. pleuronectes* : <0.16 kg, <0.36 kg, and <0.23 kg in one week, respectively.

Table 4. Provisional Tolerable Intake of Cadmium, Lead, Copper, and Zinc (mg/kg Body Weight) released by some authorities.

Organization	Provisional Tolerable Intake (mg/kg Body Weight)			
	Cd	Pb	Cu	Zn
FAO/WHO[12]	0.007 (PTWI) 0.025 (PMWI)	0.007 (PTWI)	-	-
FAO/WHO[13]	-	-	0.5 (PTMDI)	0.3 - 1 (PTMDI)

Notes: PTWI: Provisional Tolerable Weekly Intake; PMWI: Provisional Tolerable Monthly Intake; PTMDI = Provisional Tolerable Maximum Daily Intake

Copper is not carcinogenic in humans and highly-exposed populations do not appear to be adversely affected, nor does copper appear to be a cumulative toxic hazard for man, except for individuals with Wilson's disease. On this basis, a provisional value for a maximum tolerable intake of 0.5 mg Cu/kg body weight per day from all sources was established [13]. The level of copper in food meets the nutritional requirements (2-3 mg/day for adults; 0.5-0.7 mg/day for infants). For people with a weight of 65 kg should not be conceded >32.5 mg Cu in a day. With the concentration of Cu in the bivalves found on the northern coast of Central Java, the people should not be consuming blood mussels (*A. granosa*), oyster (*C. gigas*), *M. lyrata*, green mussel (*P. viridis*) and *A. pleuronectes*: >4.7 kg, >0.55 kg, >2.93 kg, >2.99 kg and >4.36 kg in a day, respectively.

Table 5. Acceptable Consumption (kg/week) and Consumption Safely Weight (kg in a week) of some bivalves captured from north coast of Central Java, Indonesia

Bivalves Species	Cd in tissue (mg/kg)	Acceptable Consumption (kg/week)	Pb in tissue (mg/kg)	Acceptable Consumption (kg/week)	Cu in tissue (mg/kg)	Acceptable Consumption (kg/week)	Zn in tissue (mg/kg)	Acceptable Consumption (kg/ week)	Safely Weight (kg/week)
A. granosa	1.44	0.32	5.24	0.10	6.94	32.90	100.15	2.60	0.10
C. gigas	1.92	0.24	5.76	0.10	59.22	2.20	620.14	0.4	0.10
M. lyrata	0.56	0.81	2.93	0.16	11.08	11.72	96.38	2.68	0.16
P. viridis	0.19	2.39	1.27	0.36	10.88	11.96	82.65	3.16	0.36
A. pleuronectes	15.34	0.03	2.00	0.23	7.45	17.44	433.71	0.60	0.03

On the metal zinc, people with a weight of 65 kg, should not be conceded >65 mg Zn in one day. With the concentration of Zn in bivalves found on the northern coast of Central Java, then the people should not be consuming blood mussels (*A. granosa*), oyster (*C. gigas*), *M. lyrata*, green mussel (*P. viridis*) and *A. pleuronectes*: >4.7 kg, 0>.55 kg, >2.93 kg, >2.99 kg and >4.36 kg in a day, respectively. In order to avoid the impact (of Cd, Cu, Pb, and Zn) on people health that could arise as a result of the consumption of bivalves, then safely weight (in kg daily, weekly, or monthly) of bivalves were allowed to be consumed can be determined (Table 5). Concentrations of Cd, Pb, Cu, and Zn in soft tissues of mussels from different localities in the world demonstrated the various values: 0.3-9.5 mg Cd/kg, 0.47-10.67 mg Pb/kg, 3.63-156 mg Cu/kg, and 45.54-542 mg Zn/kg. The Cd levels in bivalves found in the present study (0.19-15.34 mg/kg) were relatively higher than those in bivalves from other locations of the world (Table 6). The concentration of Pb in bivalves from the north coast of Java was lower than the Pb content in the bivalves of various sites, except the bivalves from Oran Harbour (Algeria) [20] and Mediterranean Sea Italy [22]. The levels of Cu in the present study were relatively comparable with those from various locations, except with those from Mediterranean Sea, Italy [22] (Table 6).

Table 6. Comparison of heavy metals concentrations (mg/kg) in soft tissues of mussels from various sampling locations from different localities in the world.

Sampling location	Cd	Pb	Cu	Zn	Authors
Izmir Bay (Turkey)	1.1	1.36	6.92	279	[14]
Canadian coast	3.94	-	9.65	197	[15]
Portuguese coast	1.25	-	13.4	542	[16]
Baltic Sea	-	5.22	13.4	269	[17]
Casablanca (Moroccan coast)	0.78	7.1	17.33	-	[18]
Pekan, Pahang (Malaysian coast)	0.3	0.47	19.05	45.54	[19]
Oran Harbour (Oran bay. Algeria)	0.67	10.67	3.63	89.29	[20]
Montenegro. SE Adriatic (5 sites)	4.25-9.5	1.4-2.3	6.25-15.25	160-221.1	[21]
Mediterranean Sea. Italy	0.6-1.0	2-9	17.9-156	60.9-189	[22]
Northern coast of Central Java	0.19-15.34	1.27-5.76	6.94-59.22	82.65-620.14	Present study

4. Conclusions

The present study revealed that the bivalves from the northern coast of Central Java (Indonesia) were contaminated by the heavy metals (Cd, Pb, Cu, Zn) in their soft tissues. Their concentrations have

exceeded the Maximum Acceptable Limits (MAL). The content of Cd has far exceeded the MAL in *A. pleuronectes*. While, Pb has exceeded the value of MAL in *C. gigas* and *A. granosa*, and Cu content in *C. gigas* has also exceeded the MAL. High content of zinc found in *C. gigas* and *A. pleuronectes*, where its value has exceeded the MAL. These results demonstrated that the bivalves from northern coast of Central Java may be harmful to food. Therefore, it is important to monitor periodically the concentrations of heavy metals in bivalves consumed by people for the reason of public health. The concentrations of heavy metals in the soft tissue of bivalves captured from the northern coast of Central Java increased in the following order: Cd<Pb<Cu<Zn.

References

[1] Mamboya F A 2007 *Heavy metal contamination and toxicity. Studies of Macroalgae from the Tanzanian Coast* (Sweden: Stockholm University) p 48
[2] EFSA (European Food Safety Authority) 2009 Cadmium in food. Scientific Opinion of the Panel on Contam-inants in the Food Chain *The EFSA Journal* **980** pp 1-139
[3] Rochyatun E and Rozak A 2007 Pemantauan Kadar Logam Berat Dalam Sedimen Di Perairan Teluk Ja-karta *Makara, Sains* **11(1)** pp 28-36
[4] Wulandari S Y, Yulianto B, WidiSantosa G and Suwartimah K 2009 Kandungan Logam Berat Hg dan Cd dalam Air, Sedimen dan Kerang Darah (Anadara granosa) dengan Menggunakan Metode Analisis Pengakti-fan Neutron (APN) *Jurnal Ilmu Kelautan UNDIP* **14(3)** pp 170-5
[5] Suyanto A, Kusmiyati S and Retnaningsi C 2010 Residu Logam Berat Ikan Dari Perairan Tercemar Di Pantai Utara Jawa Tengah (Residual Heavy Metals In Fish From Contaminated Water In North Coast Of Cen-tral Java) *Jurnal Pangan dan Gizi* **1(2)** pp 1-7
[6] Arifin Z and Fadhlina D 2009 Fraksinasi Logam Berat Pb, Cd, Cu dan Zn dalam Sedimen dan Bioavailabili-tasnya bagi Biota di Perairan Teluk Jakarta *Ilmu Kelautan* **14(1)** pp 27-32
[7] Soegianto A, Moehammadi N, Irawan B, Affandi M and Hamami 2010 Mercury concentration in edible species harvested from Gresik coast *Indonesia and its health risk assessment. Cah. Biol. Mar.* **51** pp 1-8
[8] National Oceanic and Atmospheric Administration (NOAA) *Chemical Contaminants in Oysters and Mussels" by Tom O'Connor. NOAA's State of the Coast Report. (1998) Silver Spring, MD: NOAA.* Available from http://state_of_coast.noaa.gov/bulletins/html/ccom_05/ccom.html
[9] Read D 2003 *Report on Copper, Chromium and Arsenic (CCA) Treated Timber* (New Zealand: Environmental Risk Management Authority (ERMA)) p 68
[10] WHO 1982 *Toxicological Evaluation of Certain Food Additives* (Geneva: World Health Organization ISBN: 978-92-4-120940-3, p 106
[11] FDA 2001 *Fish and Fisheries Products Hazards and Controls Guidance. 3rd Ed.* (USA: Department of Health and Human Services)
[12] FAO/WHO 2011 *Evaluation Of Certain Food Additives And Contaminants. WHO Technical Report Series 960-JECFA (Joint FAO/WHO Expert Committee on Food Additives). Seventy-third report* p 237
[13] FAO/WHO 1982 *Evaluation Of Certain Food Additives And Contaminants. WHO Technical Report Series 683-JECFA (Joint FAO/WHO Expert Committee on Food Additives)*
[14] Tuncer S and Yaramaz O *Etude des métaux lourds chez les mollusques dans les différentes zones de la baie d'Izmir (Turquie). 5èmes journées d'étude sur les pollutions* (Cannes: C.I.E.S.M.)
[15] Lobel P B, Bajidik C D, Belhode S P, Jackson S E and Longerich H P 1991 Improved protocol for collect-ing mussel watch specimens taking into account sex, condition shell, shape and chronological age *Arch. Envi-ron. Contam. Toxicol.* **21** pp 409-14
[16] Coimbra J, Carraca S and Ferreira A 1991 Metals in Mytilus edulis from the Northern Coast of Portugal *Mar. Pollut. Bull.* **22** pp 249-53
[17] Pempkowiak J, Sikora A and Biernacka E 1999 Speciation of heavy metals in marine sediments vs their bioaccumulation by mussels *Chemosphere* **39** pp 313-21
[18] Bouthir F Z, Chafik A, Benbrahim S, Souabi S, Mardhy H and Messoudi A 2004 Qualité physico-

chimique des eaux côtières du littoral de la Wilaya du grand Casablanca (ocean Atlantique marocain) utilisant la moule Mytilus galloprovincialis comme indicateur de la contamination métallique *Mar. Life*. **14 (1-2)** pp 59-70

[19] Kamaruzzaman B Y, Zahir M S M, John B A, Jalal K C A, Shahbudin S, Al-Barwani S M and God-dard J S 2011 Bioaccumulation of Some Metals by Green Mussel Perna viridis (Linnaeus 1758) from Pekan, Pahang, Malaysia *International Journal of Biological Chemistry* **5** pp 54-60

[20] Rouane-Hacene O, Belhaouari B and Boutiba Z 2012 Trace element concentrations (Zn, Cu, Pb and Cd) in the Mediterranean mussel Mytilus galloprovincialis from Oran Harbour (Oran Bay, Algerian west coast) *J. Appl. Environ. Biol. Sci.* **2(9)** pp 446-52

[21] Joksimovic D, Tomic I, Stankovic R A, Jovic M and Stankovic S 2011 Trace metal concentrations in Mediterranean blue mussel and surface sediments and evaluation of the mussel quality and possible risks of high human consumption *Food Chem.* **127** pp 632 –7

[22] Desideri D, Meli M A, Roselli C and Feduzi L 2009 A biomonitoring study: 210Po and heavy metals in mus-sels *Journal of Radioanalytical and Nuclear Chemistry* **279(2)** pp 591-600

Acknowledgments
Acknowledgment to be addressed to the Research and Development Board of Central Java, Semarang Indonesia, which has funded the realization of this study, and Diponegoro University which facilitated the laboratory for metal analysis.