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Health Risk Analysis of Cd, Pb and Hg in Blood Mussel (*Anadara granosa*) from Demak, Central Java, Indonesia

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Abstract: The famous location of Wedung waters, Demak, Central Java, Indonesia, produces blood mussel, *Anadara granosa*. Anthropogenic activities can lead to contamination of heavy metals such as Pb, Cd and Hg to the living environment of *A. granosa*. This study was done to analyse heavy metals content in the soft tissue of *A. granosa* and health risks arising to Wedung residents from consuming the mussels. Heavy metals were analysed using *atomic absorption spectrometry* (AAS). The result showed that Cd and Pb contents were found in *A. granosa* soft tissue in the range of 0.56 - 0.70 mg/kg for Cd, 0.05 - 0.10 mg/kg for Pb and Hg was not detected. A health risk analysis showed that the HQ value for Pb from *A. granosa* intake was 0.0 <1, so that it was not at risk; whereas for Cd, the value reached 26.5 (HQ > 1). Thus, there was a noticeable health risk for the residents after consuming the mussel. The safety limit in consuming *A. granosa* was 0.33 g/day for children and 1.2 g/day for adults.

Key words: Cd, Pb, Hg, health risk analysis.

Introduction

Blood mussel (*Anadara granosa*) is one of main types of shellfish and is an important source of protein in Southeast Asian countries (Yunus et al., 2014). *Anadara granosa* has an affordable price and can be developed as a source of protein and minerals consumed by the residents (Lindawaty et al., 2016). In 2015, shellfish production in Indonesia reached 59,613 tons and 85.26% of that was *A. granosa* (Ministry of Maritime Affairs and Fisheries, 2015).

A. granosa is a type of bivalve that contributes to the commercial fisheries in the Wedung district, the Demak Regency (Brotohadikusumo, 1994). Activities of Wedung residents, who are mostly farmers, fishermen and housewives, could lead to the entrance of heavy metals of Cd, Pb and Hg to Wedung waters. Agricultural activities such as the use of pesticides and fertilizers and also the runoff from these activities contributed to the increase of heavy metals content in water and sediment (Ansari et al., 2004; Farejiya and Dikhsit, 2016; Soegianto et al., 2010). Garcia et al. (1996) stated that fertilizers and pesticides used in agriculture contained Cd and Pb. Household activities and burning of fossil fuels caused environmental pollution of mercury (Hg) that eventually settled into water or land (USEPA, 2017). Port activities such as painting could spread Cd contamination. Cd is a dangerous heavy metal since it

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can increase the risk of blood vessel disorders (Tuner, 2010). The consumption of blood mussels by the people of Wedung and its surroundings is a way of exposure to various heavy metals contained in the mussels. The level of shellfish consumption, the concentration of heavy metals in shellfish, as well as the duration of exposure, are determinants of health risk hazards.

This study is aimed to analyse contamination of heavy metals Cd, Pb and Hg in *A. granosa* and their impact on the health risk of Wedung residents.

Materials and Methods

Sampling Location

Samples of *A. granosa* were collected from four stations that were chosen using *a purposive sampling* method. The criteria used to determine the stations were they have a significant population of *A. granosa* placed near to the mainland that was considered as the source of contamination. The coordinates of the four stations were as follows:

(1) Station A was located at 6°44′39.20″S – 110°33′4.37″E
(2) Station B was located at 6°44′57.29″S – 110°33′17.02″E
(3) Station C was located at 6°46′1.84″S – 110°32′21.60″E, and
(4) Station D was located at 6°47’14.36″S – 110°33′25.79″E.

The four locations can be seen in Figure 1.

Anadara granosa Collection

A group of samples of *A. granosa* with a total of 100 was collected from each station in February and March 2019. Thus, the total number of samples collected was 400. It was stored in *polyethylene* plastics and put in an iced *coolbox* to keep its maximum temperature of 4°C during transportation. Before getting analyzed, the samples were stored at a temperature below -20ºC (Kumar et al., 2011).

Heavy Metals Analysis

An analysis of Cd and Pb content in *A. granosa* was carried according to SNI 2354.5: 2011. Five grams of samples were destroyed under temperature that was increased gradually by 100 °C every 30 minutes until reaching a temperature of 450 °C in 18 hours. Then, 1 ml of 65% HNO_3 was added to the sample. It was evaporated on a hot plate at 100 °C until dry. The sample was again destroyed under the temperature of 450 °C for 3 hours. Next, 5 ml of 6 M HCl was added to the sample and evaporated on a hot plate at 100 °C until dry. Ten milliliters of 0.1 M HNO_3 were then added to the dried sample, and it was put in a measuring flask

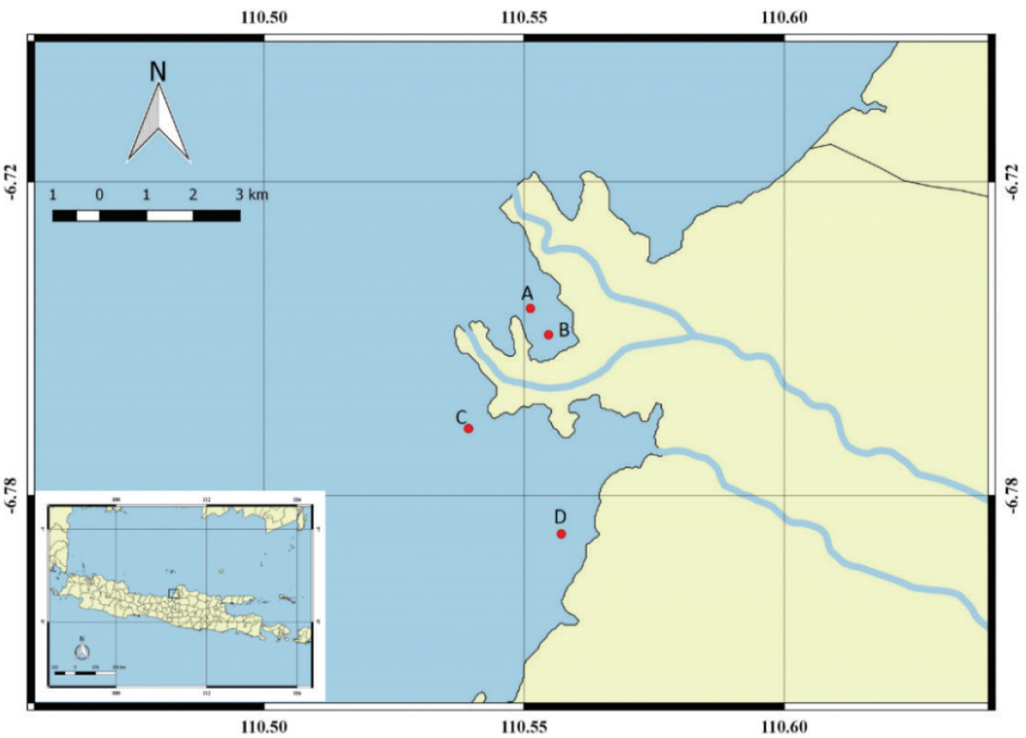

Figure 1: Sampling location.

of 50 ml. Further, 0.1 M HNO_3 solution was added to the sample until reaching the mark. Analysis of heavy metals was continued by reading the sample solution using AAS with a wavelength of 228.8 mm for Cd and 283.3 mm for Pb.

An analysis of Hg content was carried out according to SNI 2354.6: 2016. Five grams of sample was added with 3-5 boiling stones and 10-20 mg V_2O_5. Ten ml HNO_3 65% and 10 ml H_2SO_4 95-97% were then added to the sample, respectively. The sample was then heated until it turned to a yellowish-brown solution. Rinse the sample with 15 ml of deionized water. Add two drops of 30% H_2O_2 through the top end of the cooler, then rinse. The solution was then transferred to a 100 ml measuring flask and then set with deionized water. The solution was read using AAS with a wavelength of 253.7 nm. *R*-value for all heavy metal test is set to > 0.995.

Collection of Consumption Data

To measure the health risk of Wedung residents in correlation with *A. granosa* consumption, interviews were conducted for 400 people in Wedung using a prepared questionnaire. The questionnaire contained questions about daily intake, frequency of exposure, age and weight and resident's ways in consuming mussels.

Health Risk Analysis

Non-carcinogenic health risk caused by Cd, Pb and Hg to the residents who consume blood mussels was estimated by HQ value according to EPA A.S. using formula (1).

$$HQ = \frac{CID}{RfD} \quad (1)$$

Chronic Daily Intake (CDI) was calculated using equation (2) based on (U.S. EPA 1989, 1991):

$$CDI = \frac{C \times IR \times Ef \times ED}{Bw \times AT} \quad (2)$$

where C is heavy metal concentration; IR is intake rate or the weight of blood mussels consumed per day; Ef is exposure frequency; ED is exposure duration (6 years for kid and 30 years for adult); Bw is bodyweight; and AT is average time (6 years x 365 days for kid and 30 years x 365 days for adult).

HQ > 1 indicates a significant non-carcinogenic health risk. Hazard index (HI) caused by Cd, Pb and Hg were calculated using equation (3) :

$$HI = HQ_{cd} + HQ_{pb} + HQ_{Hg} \quad (3)$$

HI > 1 is indicating the need to perform risk management by reducing daily intake. From equation (1) and (2), equation (4) can be derived to calculate a safe daily intake.

$$\text{A safe daily intake} = \frac{Rfd \times BW \times AT}{C \times EF \times ED} \quad (4)$$

Result and Discusion

Heavy Metals in *A. granosa*

Cd and Pb were found in the soft tissue of *A. granosa* in the range of 0.56-0.70 mg/kg for Cd and 0.05-0.10 mg/kg for Pb, while Hg was not detected in *A. granosa*. Those heavy metals spread into waters through agricultural activities such as the use of fertilizers, pesticides and herbicides. Garcia et al. (1996) stated that fertilizers and pesticides used in agricultural activities contained Cd and Pb. It was supported by Lias et al. (2013) that stated that pesticides and herbicides containing Pb and arsenic polluted the ocean, and both were accumulated in sediments and bivalves. Fishing boat fuel contains Pb as an anti-knocking material (Parekh et al., 2002). The residue of fuel-burning was suspected of entering and contaminating Wedung waters.

The test result on the three heavy metals is shown in Table 1, while the hazard identification in the form of the average of Cd and Pb are shown in Table 2.

The concentration of Cd and Pb in *A. granosa* was under the maximum limit issued according to SNI (Indonesia National Standard) No 7387/2009 and European Council Regulation 2006. Thus, *A. granosa* was still safe to be consumed.

Health Risk Analysis

The analysis result on collected questionnaires showed that the intake rate of *A. granosa* was 8.90 g/day for children and 7.30 g/day for adults. The annual exposure frequency for children was 53 times a year, while for adults were 28 times a year. The average body weight of children in Wedung was 31.13 kg, while adults averaged in 59.40 kg. The complete data of *A.granosa* exposure to the residents in 2019 can be seen in Table 3.

Using equation (2), the CDI value for Cd was 0.026 mg/kg bw/day for children and 0.006 mg/kg bw/day for adults. While for Pb, the CDI value was 33E7 mg/kg bw/day for children and 75E7 mg/kg bw/day for adults.

HQ analysis was performed using equation (1). The RfD value for Cd was 0.001 mg/kg-day (EPA 2018), while Pb was 0.0035 mg/kg-day (Khan et al., 2008).

Table 1: Concentration of Cd, Pb, and Hg in *A. granosa* captured from Wedung Coastal Waters, Demak, Indonesia

Content	*Heavy metal content on soft tissue (mg.kg^{-1})*				*Month*	*Standard***
	A	*B*	*C*	*D*		
Cd	0.64 ± 0.18	0.64 ± 0.20	0.60 ± 0.02	0.66 ± 0.08	Feb	1.00[1,2]
Pb	0.08 ± 0.03	0.08 ± 0.02	0.05 ± 0.00	0.10 ± 0.05		1.50[1,2]
Hg	Nd	Nd	Nd	Nd		1.00[1,2]
Cd	0.62 ± 0.05	0.56 ± 0.01	0.70 ± 0.06	0.66 ± 0.13	March	1.00[1,2]
Pb	0.09 ± 0.03	0.09 ± 0.03	0.08 ± 0.04	0.06 ± 0.01		1.50[1,2]
Hg	Nd	Nd	Nd	Nd		1.00[11,2]

1. SNI 7387: 2009; and 2. European Council (2006); Nd = Not Detected

Table 2: Hazard identification in form of the average of Cd and Pb

Source	*Media potential agent*	*Average concentration of hazard Agent (mg.kg^{-1})*	
		Cd	*Pb*
Heavy metal contamination on waters	*Anadara granosa*	0.64	0.08

Table 3: Sampling of the average consumption and frequency of exposure *A. granosa* on Wedung Resident in 2019 (N = 400 persons)

Group of age and profession	*Number of sample*	*Average of consumption g/day*	*Average of age*	*Average of weight (kg)*	*Frequency of exposure A. granosa in one year*
Children	**31.00**	**8.96**	**10.84**	31.13	52.84
Toddler	4.00	3.72	3.50	12.75	39.00
Student	27.00	9.74	11.93	33.85	54.89
Adult	**369.00**	**7.30**	**39.35**	**59.40**	**27.87**
House wife	98.00	5.17	40.78	57.17	22.67
Fisherman	76.00	9.38	44.41	62.00	32.61
Trader	22.00	7.17	36.41	61.73	41.09
Student	23.00	6.23	19.91	47.48	17.52
Village Officials	31.00	5.54	41.52	63.16	28.39
Salt Farmers	23.00	12.73	45.39	61.30	48.61
Farmer	18.00	17.54	45.89	57.44	29.83
Civil Servant	2.00	4.14	32.50	60.00	18.00
Private employee	35.00	4.79	35.97	59.40	20.60
Others	41.00	5.31	34.34	62.27	24.02
Grand Total	**400.00**	**7.43**	**37.14**	**57.21**	**29.81**

From this equation, the HQ value of Cd for children and adults was 26.5 (> 1). The HQ value of Pb was 0.9 for children and 0.2 for adults. Similar results were also obtained by Soegianto et al. (2020), where the HQ index for Cd contained in blood mussels from the coastal waters of East Java, Indonesia was > 1, which indicates that these metals have the potential to cause non-carcinogenic health risk to the consumers.

The hazard index (HI) of Cd and Pb were 27.4 for children and 26.7 for adults. These HI values were > 1, indicating that consumption of *A. granosa* by Wedung residents has a high health risk.

From the HQ analysis, it was found that Cd content in blood mussel consumed by the residents had a risk to health. Thus, it was necessary to perform risk management. Risk management can be realized by reducing the rate of intake of *A. granosa* into the safe limit using equation (4). By using this equation, the safe consumption of *A. granosa* was 0.33 g/day for children and 1.2 g/day for adults.

Cadmium is a very toxic element. It naturally occurs in the soil and spreads in the environment through human activities and can eventually enter the human body through the food chain. Food is the primary access to Cd exposure in humans (Hosseini et al., 2015). Exposure to cadmium could risk human health. The impact of cadmium has been reported to be the cause of "Itai-itai" disease since 1960 (Pan et al., 1960). Cadmium is also carcinogenic for the prostate gland (Tallaa et al., 2007).

Conclusion

Hg content was not detected in the soft tissue of *A. granosa* collected from Wedung waters while Cd and Pb contents were detected in the soft tissue of *A. granosa,* with an average value of 0.64 mg/kg for Cd and 0.08 mg/kg for Pb, respectively. The HQ value of Pb from *A. granosa* was < 1. Thus, it does not risk the resident's health. On the other hand, the HQ value of Cd reached 26.5 (HQ > 1); therefore, it can risk the resident's health. The safe intake of *A. granosa* would be 0.33 g/day for children and 1.2 g/day for adults.

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