Risk assessment of blood clams (*Anadara granosa*) consumption in Hulawa Village, Gorontalo Province, Indonesia

[1]Bun Y. M. Badjuka, [2]Tumartony T. Hiola, [3]Agus Rokot, [4]Rahman Suleman

[1] Department of Environmental Sanitation, Health Polytechnic of Gorontalo, Gorontalo, Indonesia; [2] Department of Environmental Sanitation, Health Polytechnic of Gorontalo, Gorontalo, Indonesia; [3] Department of Environmental Sanitation, Health Polytechnic of Manado, Indonesia; [4] Department of Environmental Sanitation, Health Polytechnic of Gorontalo, Gorontalo, Indonesia. Corresponding author: B. Y. M. Badjuka, bunbadjuka@gmail.com

Abstract. The community activities in gold processing using mercury have led to the environmental issue in Hulawa Village, Gorontalo Province, Indonesia. The aim of this study is to identify the public health risks of consuming blood clams (*Anadara granosa*) containing significant mercury (Hg) concentrations in the village previously mentioned. A design of observational analysis in environmental health risks was applied. There were ten environmental samples from one location and 82 human samples related to particular terms. All data were obtained through direct interviews with respondents and laboratory sample analysis. The results lead to the sample of blood clams taken at the estuary point of the river containing an average Hg concentration of 0.567 ppm. The lowest intake rate (IR) of mercury (Hg) is for consuming blood clams in the amount of 50 g/day with 12 respondents, and the highest is for consuming 100 g/day with 70 respondents. The lowest amount of mercury (Hg) exposure (intake rate/IR) for consumption of blood clams is 70-90 days/year with 35 respondents, and the highest is >114 days/year with a total of 37 respondents.

Key Words: aquatic contamination, artisanal wastewater, mercury exposure.

Introduction. Mercury has been widely applied worldwide in human activities, either in medical tools (Chávez et al 2012; IPEN 2011), or in gold mining (IPEN 2016; Peycheva 2014). Problems revolving around mercury are prevalent in Europe (Marnane 2018; EEA 2017), Asia (Abeysinghe et al 2017), Australia (Bramwell et al 2018; Maher et al 2020; Stinton et al 2020), South Africa and East Africa (Campbell 2014; Walters et al 2011). Of its application in Artisanal Small-scale Gold Mining (ASGM), mercury has become a severe environmental issue over the last few years. The notorious problem is Minamata disease, which affects the neurological system, and in Japan it is caused by mercury contamination in seafood or organic mercury consumption (Minamata City Planning Division 2007; Noriyuki 2006; Semionov 2019). Another problem worth mentioning is the gold mining process, which is one of the contributing factors to mercury contamination in various countries (Arifin et al 2014; Arifin et al 2020a).

In developing countries, ASGM is easy-to-access and is vital to workers' economies. It is also one of the primary sources of mercury contributors to the environment through its gold processing, either by ore and gold separation step or in the burning step known as the amalgamation process, which produces wastewater and air pollution. The waste is directly released into the environment without any filter (Heumasse et al 2019; Marnane 2018; Verma et al 2018; Funoh 2014). ASGM has widely spread in almost all provinces of Indonesia, such as West Nusa Tenggara Province (Krisnayanti 2018), Java Province (Barkdull et al 2019), North Sulawesi (Palapa & Maramis 2015), and Gorontalo (Arifin et al 2020b; Hiola & Badjuka 2021).

The mercury pollution produced by the ASGM gold processing leads to a declined aquatic system (Azad et al 2019; Maher et al 2020). In the tailing of the amalgamation process, the mercury is distributed at a low level to the environment (soils and sediments) around the ASGM areas. The mercury is evaporated and disbanded by the wind to wider areas before the residues are stored by particular processes (dry and wet) and become naturally available. When the mercury contaminates the environment, it may react with oxygen to a particular form (amongst the chemical and physical conversion) (Abbas et al 2020; NHDES 2019). In Hulawa Village, 1 kilogram of mercury was applied to process 120 kilograms of ore. Moreover, 30 kilograms monthly, or 360 kilograms annually, of mercury were directly released in the wastewater to the river that flows into the sea (Abubakar & Katili 2014). This fact has proven that environmental mercury contaminants in Hulawa Village are undeniable. Some research has been done to investigate the effect of mercury in Hulawa Village. One example is a study by Mallongi et al (2015), which revealed that the mercury concentration in this village had been deposited in the aquatic system. Mallongi et al (2014) previously found mercury contamination in some rice grain samples. A mercury contamination research has also been conducted by Hiola and Badjuka (2021), which found that mackerel scads (*Decapterus macarellus*), the most common fish consumed by inhabitants of Hulawa Village, have been contaminated by mercury. Therefore, this study aims to investigate the mercury contamination in blood clams (*Anadara granosa*), and to perform a health risk assessment of the people of Hulawa Village consuming blood clams. According to previous research, mercury contamination has also existed in the inhabitant's hair and reached the maximum limit value (Arifin et al 2014; Arifin et al 2020b).

Material and Method

Description of the study site. This study was conducted in 12-15 July 2021, in Hulawa Village, East Sumalata Sub district, North Gorontalo Regency, Gorontalo, Indonesia, where the ASGM activities are operated (Arifin et al 2014). Blood clams were collected at the mouth of the Hullawa river to the sea (Figure 1). The exposure of mercury wastewater that emerged from the ASGM may have a significant possibility of exposing the inhabitants of Hulawa Village (Arifin et al 2020a).

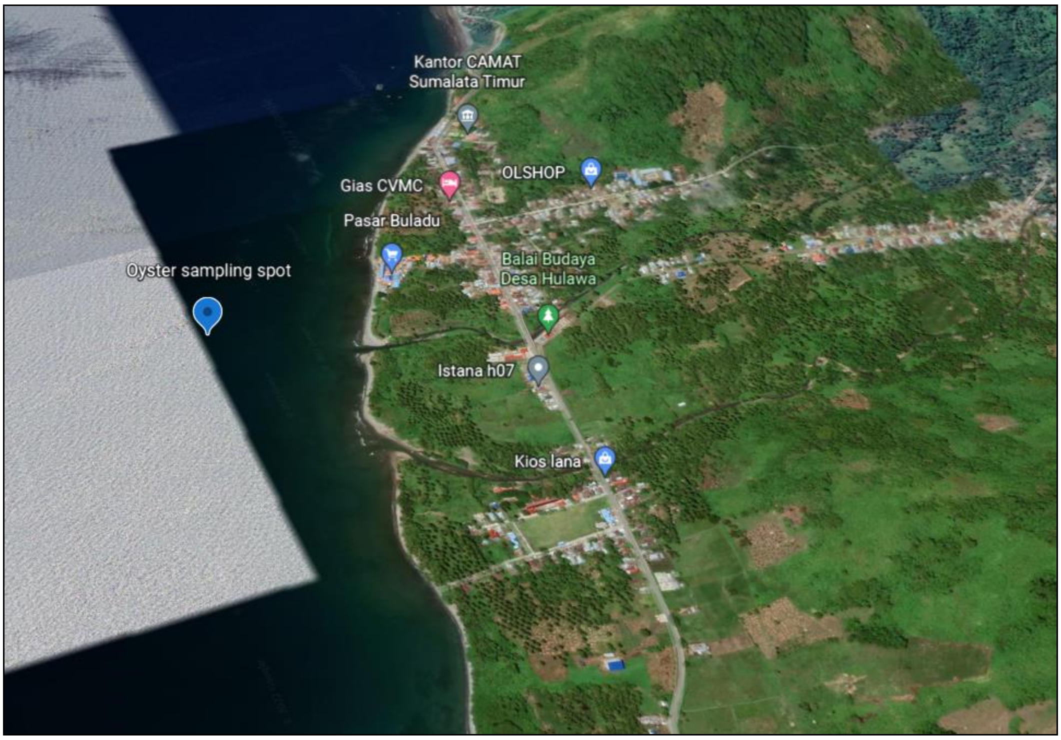

Figure 1. Location of blood clams sampling (map generated using Google Earth).

Ethical statement. The Commission of Ethics of the Health Polytechnic of Gorontalo has formally examined and approved this study, as evidenced by the registration number LB.03.02/KEPK/31/2021.

Research samples. The samples in this study were divided into two categories. The blood clams represented a data source for the environment; meanwhile, the Hulawa Village population represented the second category. The interrogated members of the community should meet the following criteria: (1) having lived in Hulawa Village for at least five years; (2) having worked for at least 15 years and consumed blood clams from Sumalata waterways. The number of samples was calculated using the following formula (Hidayat 2010):

$$n = \frac{\mathrm{N.}\ Z_\alpha^2\ \mathrm{P.q}}{\mathrm{d}^2\ (\mathrm{N}-1) + Z_\alpha^2\ \mathrm{P.q}}$$

where n is the sample size, P is the estimated proportion of the population (0.5), d is the absolute false tolerance (0.05), Z_α^2 is the number of curves dependent on (1.96), and N is the number of population units. There are 570 people in Hulawa Village and 82 people were interrogated.

For the blood clam samples, 15 blood clams were obtained from the research station in Sulawesi waters. The blood clam samples were kept in a box and transferred to a laboratory to be tested for mercury.

Analysis of mercury. The blood clam samples were delivered to the Center of Fishery Products Quality Development and Assessment Center laboratory's certified facility, South Sulawesi, to identify the mercury concentration. The atomic absorption spectrophotometry (AAS) method was applied in this procedure, which SNI-01-2354.6-2016 standardized. The AAS method was chosen because of its precise mercury identification, even in low concentrations, water or sediment (Armon & Hänninen 2015), and fish tissue (Coufalík et al 2014).

Age and duration of stay. This variable has criteria that refer to the inhabitants who have lived a minimum of 15 years in Hulawa Village. Such reasoning is also able to affect the mercury accumulation from blood clams consumption within human organs (United States Department Of Health And Human Services 1999; Vieira et al 2013).

Health risk assessment. This part discusses the risk assessment of human health based on the present or future pollution and chemical exposure in humans. The Hg intake from one-year blood clam consumption and absorption by the respondents is referred to as exposure in this study. The risk quotient (RQ) requires the reference dose for Hg (RfD mg/kg/day). RQ is also approximated.

When the RQ < 1, there is no evidence of adverse health effects from eating blood clams. However, if RQ > 1, negative health effects are probable after consuming the blood clams. According to the U.S. EPA (2007), the reference dose (RfD) for mercury (Hg) is 0.0001 mg/kg/day.

This research applied the Environmental Health Risk Assessment method, based on U.S. EPA (2007), as follows:

$$I=(CxRxfExDt)/WbxTavg$$

where: I - intake (mg/kg/day)
C - concentration of risk agent (mg kg-1)
R - rate of intake or consumption (kg day-1)
fE - frequency of annual exposure (day year-1)
Dt - duration of exposure (real-time or projection, 30 years for residential default value)
Wb - weight (kg)

Tavg - average period (Dt x 365 days per year for non-carcinogenic substances, 70 years x 365 days per year for carcinogenic substances).

For calculating Risk Rate, the formula used is:

$$RQ = I\ (mg/kg/day) / RfD\ (mg/kg/day)$$

where: I - intake
RfD - oral reference dose

Meanwhile, for managing the health risk, the formulas are:

1. To decrease pollutants:

$$C = (RfD \times Wb \times Tavg)/(R \times fE \times Dt)$$

2. To decrease intake:

$$R = (RfD \times Wb \times Tavg)/(C \times fE \times Dt)$$

3. To calculate the time contact:

$$Dt = (RfD \times Wb \times Tavg)/(C \times R \times fE)$$

where: C - concentration of risk agent (mg kg-1)
R - rate of intake or consumption (kg day-1)
fE - frequency of annual exposure (day year-1)
Wb - weight (kg)
Dt - duration of exposure, year (real-time or projection, 30 years for residential default value)
Tavg - average time period (Dt x 365 days per year for non-carcinogenic substances, 70 years x 365 days per year for carcinogenic substances)
RfD - oral reference dose).

Results and Discussion. Intake rate of blood clams in Table 1 below depicts that the highest intake rate of blood clams is 100 grams/day, and the lowest one is 50 grams/day. In addition, the finding indicates that there are 12 respondents with the lowest intake rate of blood clams of 50 grams/day and 70 respondents with the highest intake rate of 100 grams/day.

Table 1

Intake rate of blood clams

Blood clams intake (g/day)	*Number of respondents*	*Percent (%)*
50	12	14.6
100	70	85.4
Total	82	100.0

The distribution of average intake in blood clams. Table 2 denotes that the average intake rate of blood clams 92.68 grams/day where the highest intake rate is 100 grams/day and the lowest one is 50 grams/day.

Table 2

The distribution of average intake rate in blood clams

Intake rate	*Mean*	*Median*	*Min*	*Max*	*SD*
Blood clams	92.68	100	50	100	17.781

The distribution of average amount of exposure (days/year) of mercury (Hg) from blood clams. Based on Table 3, it can be observed that the respondents who consumed the blood clams have an average amount of exposure of 113.68 days/year, with the highest number of 180 days/year and the lowest number of 76 days/year.

Table 3

The distribution of average amount of exposure (day/year) to mercury (Hg) from blood clams

Sample	*Mean*	*Median*	*Min*	*Max*	*SD*
Respondents	113.68	114.00	76	180	24.756

The distribution of average body weight (kg) of respondents consuming blood clams. Table 4 depicts the average body weight for respondents consuming blood clams, which is 57.57 kg, with the highest body weight of 83 kg, and the lowest one of 38 kg. The distribution of respondents' body weight is also confirmed by the linear line of graphic in Figure 2; therefore, all respondents have fulfilled the criteria for assessment.

Table 4

The distribution of average body weight (kg) of respondents consuming blood clams

Sample	*Mean*	*Median*	*Min*	*Max*	*SD*
Respondents	57.57	58.00	38	83	9.639

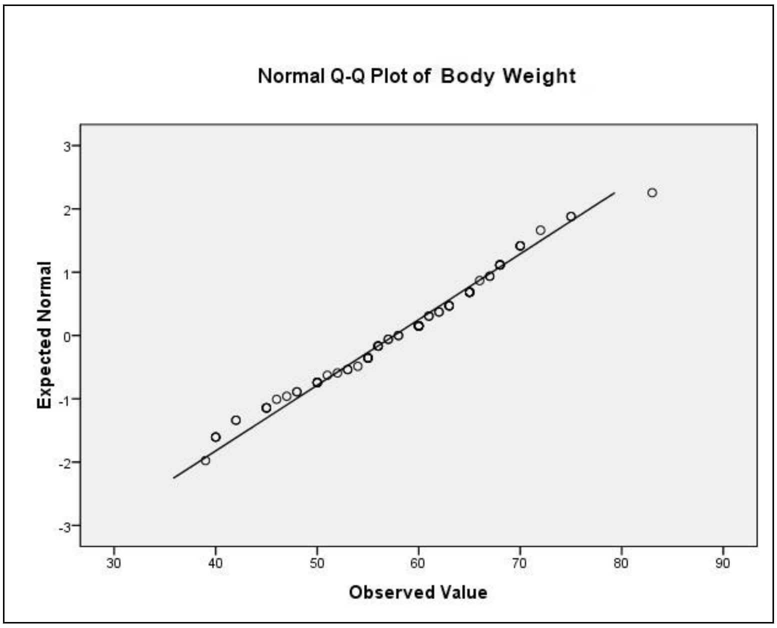

Figure 2. Distribution plot of respondents' body weight.

Intake (I) level of consuming blood clams with 30 years-exposure duration (Dt) in the Hulawa Village community. Table 5 below obviously denotes that the respondents consuming clams have an average intake value of 0.00028997 mg/kg/day with a minimum value of 0.000100 mg/kg/day and a maximum value of 0.0000524 mg/kg/day.

Table 5
Intake average value (I) of blood clams' consumption with 30 years-exposure duration (Dt)

Sample	*Min*	*Max*	*Mean*	*Median*	*SD*
Respondents	0.000100	0.000524	0.00028997	0.0002803	0.0000945

Average risk quotient (RQ) value of blood clams with 30 years-exposure duration (Dt). Table 6 below indicates that respondents who consume blood clams have an average RQ value of 2.899736 with a minimum value of 0.9986 and a maximum value of 5.2428.

Table 6
Average risk quotient (RQ) value of consuming blood clams with 30 years-exposure duration (Dt)

Sample	*Minimum*	*Maximum*	*Mean*	*Median*	*SD*
Blood clams	0.9986	5.2428	2.899736	2.803562	9.4543

Risk analysis. Risk analysis includes identifying and quantifying risks that may occur due to exposure to hazardous substances by considering the possible effects that are harmful to human health. The benefit of risk analysis is to protect humans from the potential adverse effects of blood clams from a hazardous material. Most importantly, this risk analysis aims to estimate the risks that may occur due to consuming blood clams containing heavy metals found in Hulawa Village (Daud 2011).

Risk analysis of consuming blood clams. The survey result reveals that one of the respondents (respondent no. 59) with a bodyweight (BW) of 70 kg is known to consume clams with an intake rate (IR) of cockles or clams as much as 100 grams/day. At the same time, the exposure frequency (EF) is 135 days/year because s/he does not consume blood clams every day. The amount of intake (I) in the 30 years-exposure duration (Dt) for the lowest mercury (Hg) level (C=0.0005634 mg/g) is 0.0000992 mg/kg/day.

In the meantime, the risk level of the population who consumed blood clams with the lowest mercury (Hg) level (C= 0.0005634 mg/g) for 30 years is 0.992 mg/kg/day.

The risk quotient of respondent no. 75 who has a bodyweight of 40 kg with the amount of intake (I) in the 30 years-exposure duration at the highest mercury (Hg) level (C = 0.0005701 mg/g) is 0.0001004 mg/kg/day.

The risk quotient (RQ) of the population who consumed the blood clams with the highest mercury (Hg) level (C=0.0005701 mg/g) for 30 years is 5.271 mg/kg/day.

The amount of intake (I) in the 30-year exposure duration period at the average Hg level (C=0.000567 mg/g) is 0.000524 mg/kg/day.

Likewise, the risk quotient (RQ) of the population who consumed blood clams, with the highest mercury (Hg) level (C=0.000567 mg/g) for 30 years, is 5.24 mg/kg/day.

Risk quotient value is greater than 1 (RQ>1) for the blood clams' consumption, thus, it is necessary to proceed to risk management.

Risk management of consuming blood clams containing mercury. The following table (Table 7) denotes that the minimum concentration of mercury (Hg) in blood clams is C=0.000563 mg/g, maximum of C=0.0005701 mg/g, and the average is C=0.000567 mg/g. In addition, it indicates that the safe concentration of consuming blood clams containing Hg in Hulawa Village with the average body weight of the respondents who consume blood clams of 57.57 kg, in the 30 years-exposure duration is 0.0001994 mg/kg/day. For this reason, to gain a safe concentration in consuming blood clams with the 30 years-exposure duration, the concentration of Hg in blood clam is declined to 0.000194 mg/kg/day.

In brief, the blood clams are safe to consume (RCS=100%) if it consumed in the amount of 92.68 g/day for 113.68 days/year within a period of 30 years by people weighing 57.57 kg or more if the Hg level in the blood clams is same or lower than 0.0001994 mg/g.

Risk management for safe concentration of mercury from consuming blood clams (BW average, IR average, and EF average). In compliance with Table 8, the risk level (RQ) of respondents with a bodyweight of 57.57 kg consuming blood clams of 92.68 g/day for 113.68 days/year is 5.271 mg/kg/day (RQ>1), meaning that it can be detrimental to the health of the person concerned. At the same time, the risk management in blood clams can be carried out by lowering the intake rate as follows:

$$IR = \frac{0.0001\ \text{mg/kg/day}\ \times 58\ \text{kg}\ \times 10950\ \text{days}}{0.000563\ \frac{\text{mg}}{\text{gram}} \times 113.68\ \text{days/years} \times 30\ \text{years}}$$

$$R = 32.8028658\ \ \text{mg/gram/day}$$

So, blood clams with Hg levels of 0.000563 mg/g will be safe to be consumed (RCS=100%) by someone with an average body weight of 57.57 kg or more for 113.68 days/year within a period of 30 years if the daily intake rate is not more than 32.802 g/day.

Risk management for intake exposure duration (C minimum and maximum, IR average, BW average). Table 9 shows that the risk management of the average concentration for the exposure duration to intake blood clams has a safe exposure duration (Dt) not exceeding 0.010550 mg/kg/day.

Table 7

Risk management for safe concentration of consuming blood clams (average BW, average IR, average EF, and average RfD)

Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*RfD (mg/kg/day)*	*Safe concentration (mg/g) at any exposure duration (years)*					
					5	10	15	20	25	30
Respondents	57.57	92.68	113.68	0.0001	0.0012	0.0006	0.000399	0.000299	0.00024	0.0001994

Table 8

Risk management for Hg intake rate (C minimum, average EF, average BW)

Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*C min (mg/g)*	*RfD (mg/kg/day)*	*Safe quantity (g/day) at any exposure duration (years)*					
						5	10	15	20	25	30
Blood clams	57.57	92.68	113.68	0.000563	0.0001	196.81719	98.408597	65.605732	49.204299	39.363439	32.8028658

Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*C max (mg/g)*	*RfD (mg/kg/day)*	*Safe quantity (g/day) at any exposure duration (years)*					
Blood clams	57.57	92.68	113.68	0.00057	0.0001	194.50414	97.252068	64.834712	48.626034	38.900827	32.4173558

Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*C mean (mg/g)*	*RfD (mg/kg/day)*	*Safe quantity (g/day) at any exposure duration (years)*					
Blood clams	57.57	92.68	113.68	0.000567	0.0001	195.5676	97.7838	65.1892	48.8919	39.1135	32.59459

Table 9
Risk management for intake exposure duration (C minimum and maximum, IR average, BW average)

Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*C min (mg/g)*	*RfD (mg/kg/day)*	*Safe Dt (years)*
Blood clams	57.57	92.68	113.7	0.000563	0.0001	10.61810501
Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*C max (mg/g)*	*RfD (mg/kg/day)*	Safe Dt
Blood clams	57.57	92.68	113.7	0.00057	0.0001	10.4933176
Sample	*BW (kg)*	*IR (g/day)*	*EF (days/year)*	*C mean (mg/g)*	*RfD (mg/kg/day)*	Safe Dt
Blood clams	57.57	92.68	113.7	0.000567	0.0001	10.550

Conclusions. The sample of blood clams taken at the estuary point of the river contained an average Hg concentration of 0.567 ppm. The lowest intake rate (IR) of mercury (Hg) is in blood clam consumption of 50 g/day was seen in 12 respondents, and the highest is 100 g/day with 70 respondents. The lowest amount of mercury (Hg) exposure (intake, rate/IR) for consumption of blood clams is 70-90 days/year (35 respondents), and the highest is > 114 days/year (37 respondents). The level of risk (RQ) that can cause health problems due to exposure to mercury (Hg) in the Hulawa Village community shows an RQ value of more than 1 (RQ>1). Meanwhile, the risk level of respondents with a body weight of 57.57 kg who consumes blood clams is 92.68 grams/day, for 113.68 days/year, with an RQ value of 5.271 mg/kg/day (RQ > 1), meaning that it poses a risk for health. Management of health risk reduction needs to be performed because it handles the risk of mercury (Hg) exposure by reducing the intake of blood clams that grow in Hulawa Village.

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Conflict of interest. The authors declare that there is no conflict of interest.

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Authors:
Bun Y. M. Badjuka, Department of Environmental Sanitation, Health Polytechnic of Gorontalo, Gorontalo, Indonesia, e-mail: bunbadjuka@gmail.com
Tumartony T. Hiola, Department of Environmental Sanitation, Health Polytechnic of Gorontalo, Gorontalo, Indonesia, e-mail: tumartony@poltekkesgorontalo.ac.id
Agus Rokot, Department of Environmental Sanitation, Health Polytechnic of Manado, Indonesia, e-mail: agusrokot@gmail.com
Rahman Suleman, Department of Environmental Sanitation, Health Polytechnic of Gorontalo, Gorontalo, Indonesia, e-mail: rahman_suleman@poltekkesgorontalo.ac.id

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