

Arsenic Exposure to Fish and Shellfish Consumption among Community in Makassar, Indonesia

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Article Info

Article history:

Received Jul 29, 2016

Revised Sep 20, 2016

Accepted Sep 26, 2016

Keyword:

Arsenic

Environmental health risk analysis

Gafrarium tumidum shellfish

Leiognatus equulus fish

ABSTRACT

Fish and shellfish contaminated by Arsenic (As) heavy metals from people activity discharge into the coast. Coastal community was exposure risk by As due to consumption of marine products. This study aimed to determine of human health risks level who consume fish and shellfish that contain As in Kaluku Bodoa and Untia coastal, Makassar. This research designed by observational and Environmental Health Risk Analysis (ERHA) approach. Human sample were 49 people and 8 environmental sample that selected based on certain criteria. Data collected through environmental assessment, interview and anthropometric data measurement. Data analyzed with ERHA methods. The results showed that the highest mean As levels found in *Leiognatus equulus* fish, i.e. 1.589 mg / kg and *Gafrarium tumidum* shellfish, i.e. 4.244 mg/kg of Untia coastal. The mean level of the carcinogenic risk for fish and shellfish consumption were unacceptable because they contain As that demonstrated value of more than exponent 4 ($ECR > E-4$) and non-carcinogenic risk mean level was value of more than 1 ($RQ > 1$). Fish and shellfish consumption considered unsafe and will impact health problems for the community. Community should be restrict the frequency and amount of fish and shellfish consumption.

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1. INTRODUCTION

Heavy metal pollution cases was make the world community shocked because arsenic contamination. Arsenic poisoning impact for 85 million people due to consumption of arsenic-contaminated drinking water in the coastal areas of Bangladesh. The mass poisoning case reports of environmental pollution was the biggest recorded in world human history in the 20th century. Drinking water levels of 20 mg/L has been impact for human health problems. Therefore, WHO establishes the levels of As in drinking water that allowed to less than 10 mg/L [1].

Several studies in the UK also found arsenic levels in seafood like oysters, shrimp and fish. Samples of aquatic biota were examined. It was found arsenic levels in the sponge (8-24 mg/kg wet weight), mollusks (1-68 mg/kg wet weight), crustaceans (10-79 mg/kg). While the fish varies, i.e. 2-15 mg/kg wet weight or 2-25 mg/kg dry weight content was quite high and indicated may be distributed to the humans who consume them, and accumulated in the body to be carcinogenic to humans [2].

Previous study related to heavy metals pollution from the north east of Bengal Bay, India showed toxic levels of heavy metal mercury (Hg) and arsenic (As) specified in the muscle tissue of six species of marine fish were collected. Arsenic levels were in the range of 0.02 to 2.34 mg/g dry weight. United State of

America set the levels of As that can be consumed in seafood less than 0.04 mg/day, while the number of total As levels that can be consumed at 0.2 mg/day [3].

Arsenic pollution in Buyat Bay, North Sulawesi Minahasa Regency has also been an environmental case in Indonesia. This occurs due to the location of tailings disposal which empties into the ocean waters, the mean level of As in the fish was 1,37 mg/kg total levels in Buyat Bay exceed the quality standards set by Food and Drug Administration of Indonesia which equal to 1 mg/kg [4]. Arsenic absorbed by 90% in the body that stored in the liver, kidneys, digestive tract wall, spleen and lungs. It also stored in small amounts in the hair and nails as well as undetected for a long time, a few years after chronic poisoning [5].

Results study have also been conducted in Buyat North Sulawesi [6], there were found arsenic levels in water 0.098 mg/L, and fish of 3.867 mg/kg. Furthermore, the acquired diagnosis cases of disease in the population showed not strong dose-response relationship arsenic causes skin lesions [6]. These results were consistent with high concentrations of arsenic founded which 25 wells inspected the water, the number of cases as many as 54 people who suffer from disorders of the skin (keratosis or hyperkeratosis). The results obtained, 90% of water wells polluted Buyat village with minimum concentrations (0.0063 mg/l), maximum (0.1040 mg/l) and the mean \pm SD (0.040 \pm 0.030 mg/l). Health risk (RQ) has exceeded 1, duration time was 1.5 years and a maximum consumption rate of 53 ml/day/person weighing 35 kg. Based on these data findings, Buyat community should not drink water from wells exceeds 53 ml/person/day as an environmental health risk management [7].

The Environmental Protection Agency (EPA) has determined that the metal arsenic was harmful to human health and it can lead to non-carcinogenic and carcinogenic illness. Inorganic arsenic can cause skin cancer, lung, liver and bladder. High levels of exposure can cause the death of human, while long-term exposure can cause darkening of the skin, warts on the palms, soles, and torso [8-9].

Heavy metal pollution in the waters at the Losari Makassar (near Makassar Golden Hotel) and Losari (front of Fort Rotterdam) sampling point has exceeded safe limits. In September 2012 the results obtained successively at the two points [i.e. 0.0081 mg/L; 0.0372 mg/L; 0.0185 mg/L] and [0,0045mg/L; 0.0659 mg/L; 0.0141 mg/L]. Heavy metal levels have exceeded the standard of South Sulawesi Governor Regulation No. 69 Year 2010 regarding Standard and Environmental Damage Criteria Appendix No. I.c.2. Sea Water Quality Standard for Marine tourism [10], wherein 0.002 mg/L for the metal mercury (Hg), 0.002 mg/L for Hexavalent Chromium (Cr VI), and 0.005 mg/L for lead (Pb) [11].

Characteristics of metals in an aquatic environment were highly dependent on the speciation of metals. It had affected the presence of these metals in biological tissues (bioavailability) and toxicity to biota, transportation and mobilization, as well as interaction with sediment or soil [12]. Heavy metals settle to the sediment has a residence time to thousands years while the inside of living organisms, heavy metals will be concentrated through the bioaccumulation process. Heavy metals can presence into the body organism in three ways items, namely food chain, gills and diffusion through the skin surface. Thus, indicators of heavy metal pollution in coastal waters use the sediment and marine life that presence in the waters around the sea [13-14].

Presence of heavy metals in coastal Makassar will be lowering the environment quality and toxic to marine life even to humans. Types of marine life such as fish and shellfish consumed by community because of the high protein content. The mussels were found in the coastal area and sold by fishermen on the edge. In addition, affordable prices by the community. The high level of fish and shellfish consumption which metal contaminants impact to public health in acute and chronic disease. Metal accumulated in marine life and human tissue [14-16].

The coastal community as a people affected the risk of heavy metal pollution in the waters. There were 14 districts in Makassar city, but the selected area just two sub-district which have coastal community, potentially exposed to arsenic due to industrial activities, maritime integrated region, yag agricultural pesticides and commercial fertilizers namely Kaluku Bodoa Coastal in Tallo sub-district and Untia Coastal in Biringkanaya sub-district.

Based on data about presence of heavy metals in Makassar, a source of heavy metal pollution, as well as the nature of the carcinogenic and non-carcinogenic in humans, the researchers interested to measure the levels of arsenic contained in fish and shellfish consumed by Kaluku Bodoa and Untia coastal Community. Observational study was using Environmental Health Risk Analysis approach related to Arsenic (As). These approach become limited research study and never been done before in this coastal region. This study aimed to determine the health risk of arsenic contamination in fish and shellfish due to seafood consumption for Kaluku Bodoa and Untia coastal community in Makassar City.

2. RESEARCH METHOD

2.1. Research design

This type of research was an observational study with an Environmental Health Risk Analysis (EHRA) approach which risk factors were measured at the same time to provide a magnitude prediction of the health risks caused by Arsenic in fish and shellfish.

2.2. Location and time research

The study conducted on May-June 2014 in Makassar coast, sampling site are Tallo District (Kaluku Bodoa Coastal) and Biringkanaya (Untia Coastal), South Sulawesi, Indonesia. The selected location based on the consideration that the coastal areas had experienced sedimentation and water pollution from two rivers namely, Jeneberang and Tallo river. There were also a variety of activities that potentially contaminate aquatic environments such as industrial, maritime integrated area, residential, hospitality and agricultural activities.

2.3. Population and sample

The environmental population were fish and shellfish in the Makassar coastal. The sample in this study taken from Kaluku Bodoa and Untia Coastal. The tissue samples of fish/shellfish that has been obtained, then taken to the laboratory and Research Center for Food Technology of Maros laboratory analysis of Arsenic. Sample analysis was using AES (Atomic Emission Spectroscopy).

The human population was all the fisherman who live in Untia and Kaluku Bodoa Coastal area. The sample in this study was a fishermen population as much as 49 people. Sampling was done by proportional random sampling method.

2.4. Data analysis

This study used Environmental Health Risk Analysis (ERHA) and Excel programme. Environmental health risk analysis carried out by the risk characterization procedures which calculated using the formula:

$$RQ = \frac{I}{RfD} \quad (1)$$

Annotation :

RQ : Risk Quotient (Non-carcinogenic Risk characterization)

RfD : Analysis of dose-response

I : Intake

The level of health risks for carcinogenic effects expressed in notation Excess Cancer Risk (ECR). To characterize the risk for carcinogenic effects calculation by folding intake with Slope Factor (SF). The formula for determining the ECR was as follows:

$$ECR = Ink \times SF \quad (2)$$

Annotation :

ECR = Excess Cancer Risk (carcinogenic risk characterization)

SF = reference value risk agent with carcinogenic effects (mg/kg/hari)

I = Intake (mg/kg/hari)

Arsenic has a value of SF according to international standards that was equal to 1.5 mg/kg/day, equivalent to the reference dose ($0.0001/SF = 0.0001/1.5 = 6.66667E-05$). The risk characteristics expressed by the level of risk (Risk Quotient "RQ") for the effects of non-carcinogenic was dividing between intake (I) and reference dose (RfD) while the carcinogenic risk levels expressed in notation Excess Cancer Risk (ECR), which was the multiplication of intake (I) and the slope factor (SF). Risk exists and needs to be controlled if $RQ > 1$ and $ECR > E-4$ [17].

3. RESULTS AND ANALYSIS

The results of this study showed that the risk of Arsenic in location study areas was very high levels because the fish and shellfish consumption as well as supported by the characteristics of respondents who contribute to occur the health problems.

3.1. Distribution of respondents characteristics in the Kaluku Bodoa and Untia Coastal

The distribution of respondents characteristics in the Kaluku Bodoa and Untia Coastal can be showed below in Table 1. The number of fishermen who frequently eat fish and shellfish caught of about 49 people with the majority age and length of stay for respondents both in the range of 30-40 years with a percentage of 57.1%. Table 1 also shows that respondents who had elementary education as much as 49%. Health problems were often experienced by the majority of respondents are skin complaints of itching and diarrhea as much as 28.6% of respondents to the nature of the disease constantly as much as 95.9%. The efforts treatment of the majority respondents was doing their own treatment. Community should not be allowed if the symptoms have appeared already. We recommend that the respondent should check to a doctor their health or the nearest healthcare. Low education level of respondents lead to a lack of awareness or initiatives to report health complaints to health services. Thus, the community decided to treat themselves to a generic drug with dose not fit even ignoring complaints. Major community was consider a common disease that can heal itself without further intervention.

Table 1. Distribution of Respondents Characteristics in the Kaluku Bodoa and Untia Coastal

Variable	N	Percentage (%)
<i>Age</i>		
a. <30 years old	8	16.3
b. 30-40 years old	28	57.1
c. >40 years old	13	26.5
<i>Level of Education</i>		
a. Not graduated elementary school	6	12.2
b. Elementary school	24	49.0
c. Junior High School	17	34.7
d. Senior High School	2	4.1
<i>Length of stay</i>		
a.<30 year	8	16.3
b. 30-40 year	28	57.1
c. >40 year	13	26.5
<i>Health problems</i>		
a. Skin (irritation/itching)	14	28.6
b. Muscle ache	10	20.4
c. Asthma	2	4.1
d. Rheumatism	5	10.2
e. Cough	4	8.2
f. Diarrhea	14	28.6
g. Others..etc.	0	0
<i>Characteristic of disease</i>		
a. Chronic	47	95.9
b. Recurrence	2	4.1
<i>Treatment efforts</i>		
a. Self-medication	28	57.1
b. To the health service	13	26.5
c. To witchdoctor	8	16.3

Source : Primary Data

Health problems according to Table 1 that occur such as skin complaints of itching and diarrhea that are constantly can be early symptoms due to the nature toxic of arsenic that contaminated fish and shellfish which consumed by Kaluku Bodoa and Untia coastal community. It related consistent with study that successfully confirmed to the diagnosis cases and arsenic exposure assessment in fish provided important data about dose response of arsenic can cause skin lesions that exposure to ingestion from Buyat residents amount to 0.011 mg/kg wet weight and the discovery of the high value of the Target Hazard Quotient (THQ) amount to 0.365 [6-7].

3.2. Levels of arsenic (As) from fish and shellfish in Kaluku Bodoa and Untia Coastal

The fish samples of this research founded in Kaluku Bodoa and Untia coastal areas that consumed by local people or community were *Siganus sp.* and *Leiognatus equulus* while species of mussels samples were *Anadara grandis* and *Gafrarium tumidum*. The results study from Table 2 shows that the higher mean arsenic levels in fish and shellfish found in Untia than Kaluku Bodoa coastal. The mean Arsenic levels of *Leiognatus equulus* fish was 1.589 mg / kg and *Gafrarium tumidum* shellfish of 4.244 mg/kg Arsenic.

Table 2. Levels of as Metals from Fish and Shellfish in the Kaluku Bodoa and Untia Coastal

Location	fish and shellfish species	As (mg/kg) Level
Kaluku Bodoa	Fish	
	<i>Siganus sp.</i>	1.627 1.327
	Mean	1.477
Untia	<i>Leiognatus equulus</i>	1.249 1.928
	Mean	1.589
	Shellfish	
Kaluku Bodoa	<i>Anadara grandis</i>	3.708 2.527
	Mean	3.118
	Untia	<i>Gafrarium tumidum</i>
	Mean	4.244

Source : Primary data

The results on Table 2 also shows that the mean levels of arsenic in fish and shellfish was higher in Biringkanaya Sub-district precisely in the Coastal Untia than Tallo Sub-district. The high levels arsenic in Untia coastal due to land had use as paddy fields and plantations, and also as the shipping busy traffic area. Pesticides and fertilizers used by farmers contributed greatly high levels of heavy metals arsenic discharged into the environment and eventually empties into the sea [19]. Presence of heavy metals in fish and shellfish might indicate health problems to humans through the fish and shellfish consumptions. Health problems caused by heavy metals become systemic effects with clinical symptoms appear after consuming for a long time. Based on the results of fish and shellfish samples assessments in Table 2, Kaluku Bodoa and Untia were categorize unsafe for consumption by the public because it was above the maximum threshold allowable to 1 mg/kg Arsenic for food like seafood [4], [18].

Heavy metals founded to be higher level in shellfish than fish because the tendency of all types of shellfish to store or accumulate heavy metals for a long period time and patterns of movement are limited when compared to fish. The other study showed that the order of sequence the higher cadmium levels found in the Lake Sentani was a row of benthic (*Plectropomus Leopardus*) > pelagic fish (*Pilchards*) > Shellfish (*Anadara trapezia*), the maximum value that still exist, namely (4.34 > 2.89 > 2.40) mg/kg wet weight [20].

3.3. Carcinogenic risk (ECR) and non-carcinogenic risk (RQ) level due to heavy metal exposure through consumption of fish and / or shellfish

Carcinogenic risk level was expressed in exponent without units (for example: 1,3E-4). The risk level was being acceptable or safe when $ECR \leq E-4(10^{-4})$ or expressed by $ECR \leq 1/10,000$. The risk level was being unacceptable or unsafe when $ECR > E-4(10^{-4})$ or expressed by $ECR > 1/10,000$. Non-carcinogenic risk level was expressed in terms of Risk Quotient (RQ). If $RQ \leq 1$, it means exposure remained below normal limits and the people who consume the shellfish and fish within still safe limits from a health risk by arsenic exposure throughout his life. If $RQ > 1$, it means that the exposure was above normal limits and people who eat fish and shellfish that pose a health risk by arsenic exposure throughout his life [21]. To determine the level of risk, it was necessary to include the intake value to see how much daily intake of fish and shellfish consumption that contain heavy metals than the reference dose ($As=0.0003$ mg/kg/day). Exposure duration default value used was 30 years [18].

The results study on Table 3 shows that the carcinogenic risk level (ECR) and non-carcinogenic risk level (RQ) consumption of fish and shellfish in the coastal city of Makassar residents. The means level of carcinogenic risk (ECR) fish and shellfish consumption contain arsenic (As) from Kaluku Bodoa and Untia coastal which value more than exponent 4 ($ECR > E-4$) that mean value in Kaluku Bodoa i.e. 1.19E-03 for *Siganus sp.*, 2.20E-03 for *Anadara Grandis* and 1.10E-03 for *Leiognatus equulus* and 2.31E-03 for *Gafrarium tumidum* in Untia. The mean level of Risk Quotient (RQ) for fish and shellfish consumption contain arsenic contamination was value more than 1 ($RQ > 1$) that mean value in Kaluku Bodoa i.e. 6.2 for *Siganus sp.*; 11.42 for *Anadara Grandis* and 5.71 for *Leiognatus equulus* and 11.97 for *Gafrarium tumidum* in Untia.

Thus, fish and shellfish consumption become very risky for community. Arsenic exposure impact carcinogenic and non-carcinogenic risk because all risk characterization were above the safe limit. They necessary to control the exposure pathway, intake, fish and shellfish sustainable consumption frequency.

Table 3. Level of Carcinogenic Risk (ECR) and Non-Carcinogenic Risk (RQ) Arsenic (As) by Fish and Shellfish Consumption (Dt = 30 years) from Kaluku Bodoa and Untia Community

Risk type	Location	Fish /shellfish species	Carcinogenic Risk (ECR) level / Non-carcinogenic (RQ) riskLevel		
			Mean	Min	Maks
Carcinogenic	Kaluku Bodoa	Fish			
		<i>Siganus sp.</i>	1.19E-03	4.71E-04	2.50E-03
	Untia	<i>Leiognatus equulus</i>	1.10E-03	3.42E-04	2.14E-03
		Shellfish			
	Kaluku Bodoa	<i>Anadara grandis</i>	2.20E-03	0	5.43E-03
		<i>Gafrarium tumidum</i>	2.31E-03	0	4.01E-03
Non-Carcinogenic	Kaluku Bodoa	Fish			
		<i>Siganus sp.</i>	6.2	2.44	12.95
	Untia	<i>Leiognatus equulus</i>	5.71	1.77	11.09
		Shellfish			
	Kaluku Bodoa	<i>Anadara grandis</i>	11.42	0	28.14
		<i>Gafrarium tumidum</i>	11.97	0	20.8

Source : Primary data

Organ target toxicity of arsenic were the nervous, heart, endocrine system, and haematopoietic system. The toxic effect of arsenic especially depending on the species, oxidation states and chemistry. Inorganic arsenic was considered carcinogenic and relate primarily to the lungs, kidneys, bladder, and skin disorders [1]. The evidence, people who eat fish, rice, and drinking water containing arsenic can experience systemic effect (dermal effect), immunological and lymphoreticular effects (cellular limpatik), neurological effects, reproductive effects, developmental effects, and cancer [22-23].

The findings in this study supported with other previous study [6], which showed that long-term arsenic exposure in Buyat residents who consume fish and rice had a significant relationship with an increased incidence of skin disorders or hyperkeratosis. Skin disorders as a result of exposure determinants of oral arsenic through biota such as fish from the groundwater was polluted by heavy metals arsenic. The other study also confirmed by a research report that inorganic arsenic through the ingestion can affect various effects such as dermal; hyperkeratosis, hyperpigmentation and hypopigmentation; periorbital swelling; the occurrence of spontaneous abortion and nervous system damage (if in high doses) [23-25].

Arsenic can be easily absorbed roughly 95 percent if it swallowed. Arsenic will distributed and stored in all tissues of the body and metabolized and eliminated through two sequential process. The first was the oxidation/reduction were arsenate into arsenite and arsenite into arsenate. Reduction of arsenate (As V) to arsenite (As III) required before methylation occurs. This reaction requires glutathione (GSH). Glutathione was known to form complexes with arsenic and mediates the reduction of arsenate into arsenite. Glutathione complex can be eliminated in the bile and a positive correlation has been found between glutathione and arsenic in the bile [22-23].

The second stage was the metilation, which occurs mainly in the liver, require s-adenosymetionin (SAME) and possibly other methyl donors (choline, cysteine, glutathione, lipoic acid reduced) to produce monometilarsenic acid (MMA) and dimetilarsenic acid (DMA). Both MMA and DMA are found in human urine and are expected as the end product of the metabolism of arsenic. Because DMA was removed from the cell faster than MMA or inorganic arsenic, and arsenic methylation reduces the amount retained in the tissues by the increased solubility in water arsenite, methylation was considered by some researchers as a mechanism detoxication [16], [22]. Speciation studies of human exposure to arsenic in the urine indicates that the metabolite was composed of 10-15% inorganic arsenic and acid monomethylarsenic and most (60-80%) of dimetilarsenic acid [13].

The risk level of carcinogenic and non-carcinogenic caused by fish and shellfish consumption contain Arsenic showing signs of insecurity so they were need required risk management. Risk management was done in an effort to reduce of health risk for locally and systemically effect. There are several ways to reduce levels (C) of arsenic in fish and shellfish, such us reducing the intake (R) fish and shellfish rate, reducing the frequency of arsenic exposure (FE) and reducing the duration of arsenic exposure (Dt).

4. CONCLUSION

The higher mean levels of Arsenic (As) in fish and shellfish consumed by communities were found in Untia than Kaluku Bodoa coastal areas. Carcinogenic risk level value was more than exponent 4 (ECR > E-4) and non-carcinogenic risk more than 1 (RQ > 1) at all study sites. It can be concluded that fish and shellfish were not safe for consumption and impact health problems that are non-carcinogenic even carcinogenic. Risk management need to be control it. We recommended that people, especially community

who settled in the Kaluku Bodoa and Untia coastal always restrict the daily consumption offish and shellfish by reducing the intake rate of fish and shellfish, reducing levels of arsenic during the food processing, as well as reducing the frequency of As exposure (fE) and the default duration time of arsenic exposure (Dt).

ACKNOWLEDGEMENTS

Authors highly appreciate and would like to thank for the Makassar Municipality who have given a very kind cooperation during the research commencement. Hence, we thank to National Chemical Laboratory Standardization in Maros Research Center for Food Technology, Environmental Agency of Makassar Regions, and the District Health Office of Makassar, Indonesia for their supporting and assistance during data collection and analysis.

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