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Toxic Metal Concentrations of Human Hair in Downstream of ASGM Sites in Bone Bolango Regency, Gorontalo Province, Indonesia

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Abstract. Bone Bolango is a regency that has a lot of ASGM sites that use elemental mercury for gold processing compare to the other regencies in Gorontalo province. This activity produces waste containing hazardous substances dumped in Bone River. This river has been used to support human life also a source of drinking water in the city of Gorontalo. This research aims to investigate the source of toxic metal in the human hair of Tulobolo ASGM sites in Bone Bolango Regency, Gorontalo Province, Indonesia. Hair samples were collected from the inhabitants who are living near Bone River. Total of toxic element concentration in the samples was analyzed by using Particle-induced X-ray emission (PIXE) which derives from Iwate Medical University. The maximum concentration of Hg, As, and Pb are 12.5 µg/g, 11.1 µg/g, and 13.5 µg/g, respectively. Those results show that Hg, As, and Pb are in danger level. Also, the results reveal that natural and anthropogenic sources have regionally polluted the areas.

Keywords: Toxic metals; Human hair; ASGM.

1. Introduction

Toxic metal occurs naturally in the earth's crust. However, the human activities such as industrial, agrochemical application, medical application, mining, and fossil fuel combustion also led to the high global emission of toxic metal [1–4]. Natural phenomena such as weathering and volcanic eruptions also contribute to toxic metal pollution [5,6]. The toxic metal which is inter-related with heavy metal and metalloids such as arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), and mercury (Hg) classified as human carcinogens [6]. Excessive levels of toxic metal are hazardous to the organism which enters the food chain and damage to human health [6,7]. In an epidemiological study, hair can be used to measure toxic metal levels in the body as a firmly established method in several group studies [8–11]. The Hg concentration in hair as an indicator of total mercury in the human body. When toxic metal ions come in contact with human body, they are absorbed and form complexes with a carboxylic acid, amine, and a thiol of proteins resulting in malfunctioning or death of the cells and consequently lead to

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1. Introduction

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a variety of diseases [12]. In recent years, many of the scientists concerned about the toxic metal concentration of and human health problems [13–15].

The ASGM in the developing countries become the main target for increasing their livelihood thus played in a political sphere. As well as in Ghana, ASGM has played a substantive role in the socio-economic and political life of indigenes for a long time [16,17]. By the case in Ghana, Gorontalo province may have the same case because of ASGM activities for the economic reason. According to Ministry of Forestry and Mining of Gorontalo province 2012, Gorontalo province has an of ASGM sites where Bone Bolango is the most regency which has a lot of ASGM sites and also miners [18]. According to Limbong and Agusa in Arifin 2013, Hg pollution in hair, and also fish and sediment are related to the ASGM activities [8]. The possibility of another toxic metal also will consist of groundwater or surface water when the rocks are due to the oxidation processes [19]. Impact of ASGM activities on the environment also human scalp hair in Gorontalo has been reported [8,15,20–22]. This research was undertaken to investigate the source of toxic metal by a human hair in the downstream area of Bone river, Gorontalo province, Indonesia.

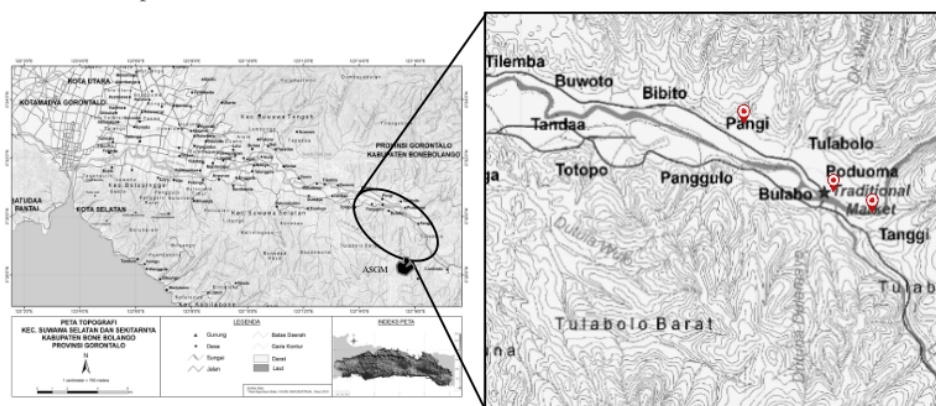


Figure 1. Map of area of research on human hair (Reconstruct from Bankonsurtanal Edition I 1991).

2. Sampling and Method

2.1. Sample locality

This present study was conducted in Bone river have to consistent use capital R or small r that located in eastern part of Gorontalo, Indonesia. The Artisanal Small-Scale Gold Mining (ASGM)'s activities are located an upper stream of Bone river, which is in the eastern part of Gorontalo. These activities cause pollution from toxic metals to the environment or human as well.

2.2. Sampling

Hair samples (n=24) in this present study were collected from the residents of three villages in the upper stream to a downstream area along Bone River (river) that is one of disposal of waste from the mining area, Bone Bolango regency, Gorontalo Province. The names of those three villages are Tulabolo (August 2015), Poduoma, and Pangli (August 2017). Collected hair samples were stored immediately in plastic zip-lock bags and identified the names of the persons.

2.3. Preparation sample

The human hair samples washed with deionized water a Sharp ultrasonic cleaner bath for five minutes to remove dust, bacteria, dirt, hair cosmetics, and other possible contaminants. After that washed again with acetone (Wako Pure Chemical Industries, Ltd., Osaka, Japan) to wash out the attached material that did not purify water. Then the clean hair samples were dried in a ventilated oven at 40 °C for 48

hours for particles-induced X-ray emission (PIXE) analysis in the Cyclotron Research Center, Iwate Medical University, Japan.

2.4. Analytical method

A particle-induced X-ray emission (PIXE) analysis was used to determine the toxic metal concentrations in human hair samples. For PIXE analysis the hair sample was as much as eight hairs per person were stuck on a target holder, and bombarded with 2.9 MeV proton beam energies from a cyclotron [23].

3. Results

3.1. Toxic metal concentrations in human hair

3.1.1. *Mercury (Hg)*. Mercury concentration of human hair of all participants are more than $0.01 \mu\text{g/g}$ which indicates that toxicity is in danger level based on German Human Biomonitoring Commission revision 2011 [24]. The average concentration with highly mercury levels over $0.01 \mu\text{g/g}$ was $6.38 \mu\text{g/g}$, $0.87 \mu\text{g/g}$, and $0.88 \mu\text{g/g}$ in Tulabolo, Pangi, and Poduoma, respectively. The Hg has regionally polluted by the processes of smelting gold (change the sentences is not clear what do you mean). This method still used for centuries to process gold in ASGM in the world. These processes lead to vaporized the elemental mercury into a toxic by atmospheric. The Hg in gold mining processes may absorb through the skin by going the transport of mercury across the epidermis and via the sweat glands, sebaceous glands, and hair follicles [25,26].

3.1.2. *Lead (Pb)*. ASGM relied on several techniques such as panning, shallow pit or deep shaft mixed with other heavy metals which associated with gold such as arsenic (As), copper (Cu), silver (Ag), and lead (Pb) [27–29]. In this research, the result showed that the Pb concentration of hair samples are more than $0.15 \mu\text{g/g}$ which indicates that toxicity is in danger level as well according to German Human Biomonitoring Commission revision 2011 [24]. Gold mining activities could occur this Pb contamination. Several researchers have been reported that Pb concentration range in gold mine tailings are about 80 mg/kg to 510 mg/kg whereas the standard mean concentration of Pb in surface soils range is 10 to 67 mg/kg [30–32]. Even though the Pb does not dissolve easily to the environment, but it can mix with soil particles or dust and enter underground water or drinking water following rain and surface water. The exposure of Pb can be able through by several eating food which cultivated on soil with high Pb concentration, drinking water, polluted air and so on [29,31,32].

3.1.3. *Arsenic (As)*. Arsenic concentration ranging from hair samples in this research is about 0.14 to $10.4 \mu\text{g/g}$. This concentration is higher than the reference value of As according to German Human Biomonitoring Commission 2003 [33]. The source of Arsenic pollutant has a possibility appearing from gold mining process which occurs as arsenopyrite (FeAs) or by hydrothermal processes [29,34]. The mine tailing which contains substantial of As can release to surface water, groundwater used for drinking water, or the other aquatic environment through geothermal water [34,35].

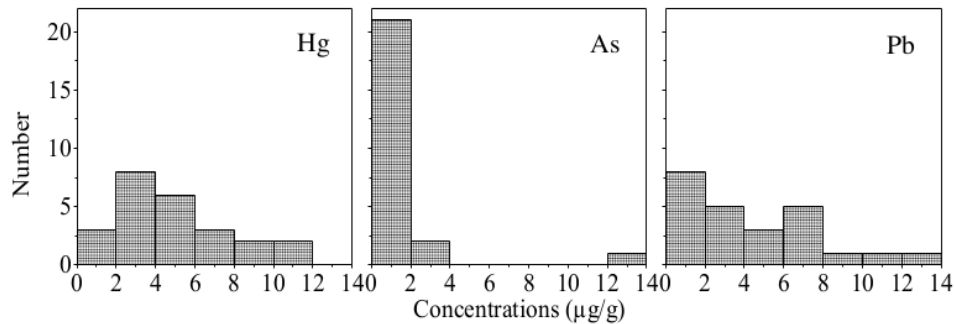


Figure 2. Histograms of Hg, As, and Pb concentrations.

4. Discussion

Toxic metal from ASGM associated with the released of harmful elements from the tailing on gold processing. The intoxication of human by Hg due to ASGM activities has been examined and having elevated levels of Hg in urine and hair samples in several places in Indonesia [36]. Miner is working using Hg in the process of milling the materials, panning processes, until burning the amalgam which can expose the elemental of Hg and then transformed to inorganic Hg in the human body [36,37]. This research area, the atmosphere has been polluted by elemental Hg which possible to have direct contact with humans through the skin or breathing.

Table 1. Total Hg, Pb, and As concentrations ($\mu\text{g/g}$) in scalp hairs of inhabitants in three villages in the downstream area of Bone River, Bone Bolango regency, Indonesia.

Village	Hg ($\mu\text{g/g}$)			Pb ($\mu\text{g/g}$)			As ($\mu\text{g/g}$)		
Tulabolo	7.43	\pm	1.88	5.01	\pm	2.03	0.00	\pm	0.00
	6.53	\pm	1.66	2.79	\pm	1.64	0.00	\pm	0.00
	7.11	\pm	1.41	2.95	\pm	1.46	0.44	\pm	0.49
	2.10	\pm	1.48	5.52	\pm	2.26	2.43	\pm	0.76
	2.96	\pm	1.72	2.59	\pm	2.07	1.14	\pm	0.69
	9.35	\pm	1.87	4.40	\pm	1.94	0.00	\pm	0.00
	6.18	\pm	1.61	4.02	\pm	1.66	0.26	\pm	0.56
	5.57	\pm	1.34	1.87	\pm	1.40	0.83	\pm	0.47
	4.11	\pm	1.16	2.28	\pm	1.31	0.72	\pm	0.44
	3.55	\pm	0.84	1.01	\pm	0.84	0.00	\pm	0.00
	6.03	\pm	1.25	1.89	\pm	1.28	0.56	\pm	0.43
	11.04	\pm	1.37	7.62	\pm	1.52	1.48	\pm	0.51
	5.10	\pm	0.91	3.55	\pm	1.13	2.06	\pm	0.38
	12.27	\pm	0.73	4.26	\pm	1.05	13.48	\pm	0.53
Min	2.10	\pm	0.73	1.01	\pm	0.84	0.00	\pm	0.00
Max	12.27	\pm	1.88	7.62	\pm	2.26	13.48	\pm	0.76
Pangi	0.00	\pm	0.00	9.28	\pm	1.76	0.00	\pm	0.00
	2.62	\pm	1.61	6.82	\pm	1.52	0.00	\pm	0.00
	0.00	\pm	0.00	5.31	\pm	1.50	0.00	\pm	0.00
Min	0.00	\pm	0.00	5.31	\pm	1.50	0.00	\pm	0.00
Max	2.62	\pm	1.61	9.28	\pm	1.76	0.00	\pm	0.00
Poduoma	0.05	\pm	1.64	2.06	\pm	1.56	0.55	\pm	0.51
	0.00	\pm	0.00	10.17	\pm	2.24	0.14	\pm	0.74
	1.79	\pm	1.69	8.02	\pm	1.75	0.00	\pm	0.00
	2.39	\pm	1.65	2.29	\pm	1.70	0.28	\pm	0.50
	1.91	\pm	1.42	3.24	\pm	1.27	0.00	\pm	0.00
	0.00	\pm	0.00	11.11	\pm	1.94	0.00	\pm	0.00
	0.00	\pm	0.00	6.96	\pm	2.34	0.16	\pm	0.62
Min	0.00	\pm	0.00	2.06	\pm	1.27	0.00	\pm	0.00
Max	2.39	\pm	1.69	11.11	\pm	2.34	0.55	\pm	0.74

Human hair sample analysis results in this research showed the total concentration of Hg with another harmful toxic element as well as As and Pb are in alarm level. The nearest village to the mine had the most massive pollution of toxic metals. The emission and accumulation of heavy metals to the environment have caused by the disruption of the natural sources of the geochemical cycle through the anthropogenic activities include ASGM [38]. Unlike other toxic metal such as Zn, Cu, and Ni, the Hg,

As, and Pb are not biological importance to a living organism and the most toxic to the ecosystem [29,38].

By the ASGM activities not only produce the existing waste of Hg by amalgamation processes but also produce other toxic elements such as As and Pb. That toxic element also tends to bioaccumulate in human beings and an environmental media such as water, soil, sediment or food crops [39,40].

5. Conclusions

ASGM have played an essential role in the livelihood of economic development in developing countries despite releasing toxic wastes into the environment. The concentration of toxic metals such as Hg, Pb, and As in human hair samples of ASGM sites is immensely higher according to German Human Biomonitoring Commission. The use of elemental Hg in amalgamation processes of gold mining is one of the toxic sources to the environment. This Hg concentration of human hair is the consequence of ASGM activities and potential risk to human health. In addition to that, the possibilities of Pb and As contamination also related to the mining activities and also mixing to the natural sources by hydrothermal alteration processes of gold mineralization. According to this research, the finding shows there are two primary sources: natural and anthropogenic sources of toxic metals release to the environment.

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