Arsenic, lead, and mercury concentrations of scalp hairs in ASGM miners and inhabitants of Gorontalo Utara regency, Gorontalo province, Indonesia

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1 Introduction

The Gorontalo Utara Regency is home of the oldest ASGM site in the Gorontalo Province and has now three active major ASGM sites which is estimated produces 286 kg of gold and emits 572 kg of Hg per year¹. Impacts of emission of more than half ton Hg from ASGM sites to the environment and human health are reported recently². Impacts of similar ASGM activities on health condition of miners and inhabitants of mining area and polluted rivers and soils are reported elsewhere^{3,5}.

In this report, we presented the evidence of other toxic heavy metals (As and Pb) in scalp hair of miners and inhabitants of Gorontalo Utara Regency. The distributions and correlations of heavy metals concentration in scalp hairs are interested to study.

2 Experimental

2.1 Study Area

Samples were collected from five districts in the Gorontalo Utara Regency: Anggrek, Kwandang, Monano, Sumalata, and Tolinggula (figure 1). Geographically, Tolinggula, Sumalata, Monano, Anggrek, and Kwandang are situated on hills and mountains along the coastline of the Gorontalo Utara Regency. Inhabitants of the Gorontalo Utara Regency mainly work as farmers and fishermen. Marine fish are commonly part of their diets, along with rice, corn and vegetables, which are also produced on the nearby hills alongside the coastline.

The ASGM activities in the Sumalata and Anggrek districts are located along the Wubudu and Anggrek riverbanks, respectively. The bioaccumulation of Hg, which may occur in living organisms such as paddy rice, corn, and marine fish, become agents that spread Hg contamination through the food web of inhabitants of the Gorontalo Utara Regency. The Hg concentration in river sediments and fish will be used as background information about the Hg in the biotic and abiotic environments.

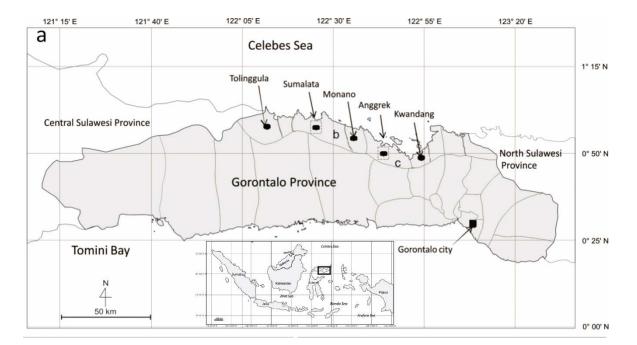


Fig. 1. (a) Gorontalo Province map showing sampling locations (●) of human hair from Gorontalo Utara Regency, showing Tolinggula Sumalata, Anggrek, Monano and Kwandang districts. Location of Gorontalo Province in Indonesian map is shown (inset)

The Sumalata and Anggrek districts are locations with ASGM activities, while Kwandang, Monano, and Tolinggula are districts without mining activities. The residents of Anggrek and Sumalata are considered the ASGM miners group, while the residents of Kwandang, Monano, and Tolinggula are considered the control group.

2.2 Sampling

Human scalp hair samples were taken from 95 participants from inhabitants of Anggrek (n=25), Sumalata (n=23) and other regions of the Gorontalo Utara Regency (Kwandang (n=7), Monano (n=37), and Tolinggula (n=4)) between 2012 and 2013. Of the 95 participants, 53 were female, and the mean age was 23 years (range: 8 months – 63 years). Among the 95 participants, 19 were ASGM workers, 15 were housewives, six were unemployed, one was teacher, one was university student and 38 were children (participants with ages below 18 years old).

The As, Pb, and Hg concentrations in the hair samples from Anggrek, Kwandang, Monano, Sumalata, and Tolinggula were determined to understand the status of contamination. The distribution of participants according to sex, location and occupation are summarized in Table 1. Approximately 10-20 strands of hair were cut close to skin from the right backside (mastoidal region of the temporal bone) and then labeled and stored in a sample plastic bag⁵.

The Hg concentration in hair samples will be used to characterize the risk through a comparison with reference values published by the German Human Biomonitoring Commission in 1999 (Commission Human – Biomonitoring of the Federal Environmental Agency Berlin, 1999)⁶. The German Human Bio-Monitoring (HBM) commission established toxicology threshold limits, which can be put into three categories. The first category is below normal or HBM I, where the Hg level in hair is below 1 µg/g. The

above normal category is an alert level between HBMI and HBM II, where the Hg hair content is from 1 to 5 μ g/g. Meanwhile, above 5 μ g/g is categorized into the high level or over HBM II. Permissible limit of Pb in hair is about 70 μ g/g⁷, Pb and As concentrations in hair is correlated with Pb and As concentrations in blood^{8,9}.

Residence	Sex	N
Anggrek	F	11
	Μ	14
	Total	25
Kwandang	F	6
	\mathbf{M}	1
	Total	7
Monano	F	22
	Μ	15
	Total	37
Sumalata	F	11
	Μ	12
	Total	23
Tolinggula	F	3
	М	0
	Total	3
Total	F	53
	Μ	42
	Total	95

Table 1. Distribution of information on scalp hair donors among inhabitants of Gorontalo Utara Regency

2.3 Analytical Procedure

Elemental analysis for the scalp hair samples was performed by particle induced x-ray emission (PIXE) in the Cyclotron Research Center, Iwate Medical University, Japan. The precision and accuracy of this method have been reported elsewhere¹⁰⁻¹⁴. Hair samples were washed using Milli-Q water and shaken in an ultrasonic bath for 1 minute. Then, the samples were dried by wiping them with a tissue. The dried hair samples were washed again by being stirred in acetone for 5 minutes. Then, they were washed again using Milli-Q water, wiped well with tissue and left to dry at room temperature. The hair samples (approximately seven hairs per person) were stuck on a target holder. A 2.9 MeV-proton beam

hit the target after passing through a beam collimator of graphite, whose diameter was 6 mm. X-rays of energy higher than that of the K- Ka line were detected by a Si (Li) detector (25.4 mm thick Be window; 6 mm active diameter) with a 300 mm-thick Mylar absorber. For measurements of X-rays lower than the K-Ka line, a Si (Li) detector (80 mm Be; 4 mm active diameter), which has a large detection efficiency for low energy X-rays, was used. Descriptions of the data acquisition system and the measuring conditions are reported elsewhere⁸. The typical beam current and integrated beam charge were 100 nA and 40 mC, respectively. The procedure for the standard-free method for untreated hairs is almost the same as that reported in the previous studies⁶.

3 Result and discussion

Table 2 shows the summary of statistical results of As, Pb, and Hg concentrations in hairs of the Gorontalo Utara Regency inhabitants. There is clear that mean concentrations of As, Pb, and Hg for inhabitants of Anggrek and Sumalata are higher than inhabitants of Kwandang, Monano, and Tolinggula.

We perform Spearman correlation on the total sample and we found the tree cofficients which is tabulated in Table 3. There are strong and significant correlations of Pb and As concentrations to Hg concentration in hairs of inhabitants of the Gorontalo Utara Regency. While weak and non-significant correlation is found between As and Pb.

Region	A	As		0	Hg	
	$Mean \pm SD$	range	$Mean \pm SD$	range	$Mean \pm SD$	range
Anggrek	1.7 ± 2.5	0.0 -11.3	15.8 ± 16.4	0.3 - 67.6	16.9 ± 29.2	2.1 - 144.8
Kwandang	1.1 ± 0.8	0.0 - 2.5	4.9 ± 5.1	0.0 - 13.1	6.9 ± 4.0	3.5 - 14.6
Monano	0.4 ± 0.9	0.0 - 4.2	4.6 ± 3.8	0.0 - 16.8	6.2 ± 4.1	2.8 - 28.1
Sumalata	2.7 ± 4.4	0.0 - 19.9	10.4 ± 7.9	2.8 - 32.0	10.9 ± 13.2	2.5 - 69.8
Tolinggula	0.3 ± 0.3	0.0 - 0.7	3.3 ± 3.0	0.1 - 5.9	5.1 ± 0.8	4.3 - 6.0
Total	1.3 ± 2.7	0.0 - 19.9	8.7±10.4	0.0 - 67.6	10.2 ± 16.9	2.1 - 144.8

Table 2. Arsenic, Lead and Mercury concentrations (μ g/g) in hairs of inhabitants of the Gorontalo Utara Regency

Table 3. Correlation coefficient between elements	Table 3.	Correlation	coefficient	between	elements
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Element	As		Hg		
	coeff	sig	coeff	sig	
Pb	0.07	0.47	0.24	0.015	
Hg	0.25	0.012			

4 Conclusion

Concentration of As, Pb, and Hg in average of miners and inhabitants of exposed group (Anggrek and Sumalata) are elevated than contro group (Kwandang, Monano, and Tolinggula). According to HBM that in general inhabitants of Gorontalo Utara Regency are already in alert level, while mean values for inhabitants exposed groups are already in danger level. The amount of Pb in hair of inhabitants of Gorontalo Utara Regency are still in save level, while for As are already higher than limit.

We found also significant and strong correlation between concentration of As and Pb to Hg in scalp hair of inhabitants of the Gorontalo Utara Regency. The importance of such correlation may reveal the unknown sources contamination, routes of contamination and impact on health status of individual hair donors.

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