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Heavy Metals in Water of Stream Near an Amalgamation Tailing Ponds in Talawaan – Tatelu Gold Mining, North Sulawesi, Indonesia

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Abstract

Heavy metals exposed to the environment as a result of Talawaan–Tatelu mining activity (North Sulawesi, Indonesia) potentially contaminated the surrounding area. The purpose of this study was to determine the content of As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg in water of stream near an amalgamation tailing pond in Talawaan–Tatelu mining. The metals were determined using ICP/MS, except Hg using CVAFS method. In general, the metals content in water of stream near Talawaan–Tatelu mining is higher than one found in the river water near the other traditional mining in Indonesia and the world's metals average content in the river water, but lower than the Indonesian standard of water quality.

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Keywords: amalgamation tailing; artisanal and small-scale gold mining; heavy metals; stream; Talawaan-Tatelu gold mining; water

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Nomenclature	
g t $\mu g \cdot L^{-1}$ m	weight (1 g equal 1 000 mg and equal 0.001 kg) weight (1 tonne equal 1 000 kg) mass concentration (microgram per liter) length (1 meter equal 0.001 km) area (1 hostare equal 10 000 square meter)
h	time (hour)
d	time (day)
mo	time (month)
yr	time (year)

1. Introduction

Artisanal and Small-scale Gold Mine (ASGM) in Indonesia has lasted long enough which in practice is done by manual workers who use a simple method, known as amalgamation. In mining, the amalgamation is used to recover precious metals from ore using mercury. One of ASGMs in Indonesia which is still in operation up to present, namely Talawaan–Tatelu mining. This traditional mining located in Talawaan Village and Tatelu–Rondor Village, respectively located in the Talawaan District and Dimembe District, two districts bordering in North Minahasa Regency, North Sulawesi, Indonesia.

At first, residents surrounding this mine depended their lives on farming (paddy, coconut, cloves, nutmeg) and livestock (freshwater fish) activities. These activities had been the main economic activities of Talawaan people before 1998 when an Australian company *Aurora Mining Co.* (hereinafter known as the *Archipelago Resources Pty. Ltd.*) discovered gold in this area. Along the way, due to the pressure of the people in the surrounding area of Talawaan, the Australian company's mining concessions handed over to the traditional miners of around 10 000 people¹.

Gold ore in Talawaan mining was obtained manually from a narrow tunnel with a depth of 20 m to 30 m. The obtained ore was placed in a sack and transported to a processing center. The miners generally obtained 0.5 t to 1 t of ore $\cdot d^{-1}$ with a gold content obtained ranged from 5 g to 10 g $\cdot t^{-1}$ of ore. Each processing unit operated 12 to 48 grinding machine (local people call it *tromol*) with a diameter of 48 cm and a length of 60 cm. One *tromol* grinded 40 kg of ore for 4 h. After 4 h passed, milling paused and added approximately 0.5 kg to 1 kg of mercury into the *tromol* then grinding was continued for 1 h. One unit of milling processing 400 kg of gold ore before the amalgam was taken. This procedure released mercury into the environment about one third of the total mercury that was added earlier in the *tromol*. Mercury – gold amalgam was separated from the mud ore by means of panning and filtering then the amalgam was burned in the open air¹.

In July 2000, it was estimated there were about 200 to 250 gold processing units operating in Talawaan¹. This activity increased to around 400 units in June 2001. Each processing unit operated about 12 gold grinding machines which was able to earn 4 g to 6 g of gold per cycle (0.3 g to 0.5 g gold per machine). In general, milling lasted two cycles per d (one cycle operates about 4 h), 6 d per week².

Based on the monitoring results in around 133 units operating in Talawaan area, Veiga and colleagues¹ found that gold milling operators used about 10 kg to 30 kg of mercury per mo (average of 15 kg per milling unit). The ratio of mercury lost to gold obtained was quite high in this area which was about 40 : 60. This ratio was similar to that proposed by Filho et al.³. From the comparison of the results they obtained was found that the ratio of mercury lost to gold obtained were found in Talawaan area reaching 30 times to 40 times higher than the mean ratio of ASGM mining activity around the world. This fact was confirmed by Ayhuan et al.⁴ who reported that the traditional gold mining activity in Talawaan generally used 1 kg of mercury for every 30 kg of gold ore.

Environmental monitoring results reported in a few references generally state that ASGM of Talawaan has polluted the environment in any significant degrees. At the end of 2000, Limbong et al.² examined all drinking water

installations in the city of Manado (Manado is located about 20 km to 30 km from the Talawaan mine site) and found that total mercury content ranged from $0.05 \ \mu g \cdot L^{-1}$ to $0.13 \ \mu g \cdot L^{-1}$. Later in the May 2011, they found that the content of mercury has increased about 5 times to 18 times in two installations which are located adjacent to the mining area. Another study of Limbong et al.⁵ also noted that in May to June 2001 there was an increase in levels of mercury in water, sediment, and fish in the three rivers that flow into the city of Manado which is the tributaries of the Talawaan River.

Mercury disposed to the environment from Talawaan ASGM activity is estimated at $24 \text{ t} \cdot \text{yr}^{-1}$ to $90 \text{ t} \cdot \text{yr}^{-1}$. Even Aspinall⁶ estimates that mercury disposed each year ranged from 96.05 kg \cdot d⁻¹ to 285.12 kg \cdot d⁻¹ or approximately 35.06 t \cdot yr⁻¹ to 104.07 t \cdot yr⁻¹. When compared with the disposal of mercury carried by the Chisso Corporation in Minamata beach (which is estimated at around 27 t over a period of 37 yr, it has been phenomenal which caused *Minamata disease*)⁷, the disposal of mercury which is derived from ASGM activity in Talawaan is much higher.

Data on the content of heavy metals in the environment around Talawaan–Tatelu ASGM activities which successfully traced through the literature review was last reported a decade ago. In fact, the data only reported the content of heavy metals, especially mercury. Monitoring such as the characterization of various heavy metals in the environment around the mine activity especially around Talawaan Watershed is very important to do in sustainable manner. This study aims to determine the content of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), and mercury (Hg) in water of a small river near the amalgamation tailing ponds in Talawaan–Tatelu ASGM activities.

2. Material and methods

2.1. The time and place of study

The study was conducted from September to December 2013 in Talawaan–Tatelu (TT) ASGM activities which is located in Talawaan Village and Tatelu–Rondor Village, respectively in Talawaan District and Dimembe District, two districts bordering in North Minahasa Regency, North Sulawesi, Indonesia (Fig. 1).

2.2. Water sampling and pretreatment

Water samples were taken on a small stream near the amalgamation tailing ponds using glass bottles sampler. Previously the bottles sampler were washed and rinsed with demineralized water. Water samples were taken directly using the bottle sampler with position of the bottle's mouth facing the direction of flow of stream water. Acidification was done directly in the field on water samples to analyze metal content by adding concentrated nitric acid until the pH is less than 2. Bottle which already had contained water samples was labeled according to sampling station and time. Bottle samples were then put in a cool box and taken to the laboratory for further treatment.

2.3. Heavy metals determination

Determination of heavy metal content in water samples carried out by a testing laboratory, *Water Laboratory Nusantara* (WLN, *PT. Water Laboratory Nusantara Indonesian*, www.wln.co.id). This laboratory has received accreditation as a testing laboratory by the National Accreditation Committee of Indonesia (*Komite Akreditasi Nasional*, KAN) with registration number: LP-433-IDN. This laboratory (WLN) is a subsidiary of WLN BV, Glimmen, Netherland (www.wln.nl) which has been holding an ISO/IEC 17025 since 1993 and ISO 9001:2000 since 2008. Determination of dissolved arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn) in water samples were done using the reference method developed by the American Public Health Association (APHA), by Inductively Coupled Plasma/Mass Spectrometry method (ICP/MS, APHA-3125-B, 2005). The determination of mercury (Hg) used the reference method developed by US Environmental Protection Agency (USEPA), through Cold Vapor Atomic Fluorescence Spectrometry method (CVAFS, USEPA-245.7, 2005).

3. Result and discussion

Talawaan–Tatelu ASGM activities have total area of \pm 822 ha (2004 data) which include several villages, namely: Wasian, Tatelu, Tatelu–Rondor, Warukapas, Talawaan, Kolongan, Tetey, and Mapanget. These villages are the upstream regions of Talawaan watershed which cover area of 34 000 ha. In Talawaan watershed there are three major rivers, namely: Talawaan River, Kima River, and Bailang River⁹. This study focused on a small river where the upstream is a small natural spring, while the downstream is Talawaan River. When processing took place on milling and amalgamation unit, its wastewater flowed directly into this stream. Based on previous observation⁸ around the downstream there are fresh waterfish ponds, paddy fields, hog raising pens, as well as housing of Talawaan residents (Fig. 2). In addition, the surrounding communities utilize Talawaan River for daily activities such as bathing and washing¹⁰.



Fig. 1. Research site in: (a) map of Indonesia; and (b) map of North Minahasa Regency⁸.

Fig. 3 presents the data of heavy metals content in the water of stream near the tailing ponds of mill and amalgamation processing units in Talawaan–Tatelu mining. The content of some heavy metals in the stream water is relatively very low, below the detection limit. The metals that are below the limits of detection i.e., cadmium, copper, nickel, zinc, and mercury, with the detection limit i.e., $0.1 \ \mu g \cdot L^{-1}$; $5 \ \mu g \cdot L^{-1}$; and 0.05 $\ \mu g \cdot L^{-1}$, respectively. The metals that can be detected i.e., arsenic, chromium, and lead, with the mean of concentration i.e., $(10.00 \ \pm 1.73) \ \mu g \cdot L^{-1}$; $(5.00 \ \pm 1.00) \ \mu g \cdot L^{-1}$; and $(23.00 \ \pm 3.46) \ \mu g \cdot L^{-1}$, respectively. For comparison, in Table 1 are presented the data of heavy metals content in the water of stream near Talawaan–Tatelu mining, along with the data from the river that is closer to some of the other ASGM mining in Indonesia, the world's average metal content in the water of the river¹¹, and the water quality criteria for class I and IV based on Indonesian Government Regulation No.82, 2001^{12} .



Fig. 2. The stream delta (red arrow), the mouth of stream where the stream flows into Talawaan River (this image shows the location of some inhabitants' economic activities, modified from www.maps.google.co.id).



Fig. 3. Bar graph of heavy metals concentration in water of stream near Talawaan-Tatelu mining.

The arsenic in the water of stream near Talawaan–Tatelu mining is fairly high, $(10.00 \pm 1.73) \ \mu g \cdot L^{-1}$, when compared with the water of Batudulanga River near the Mount Pani mining in Gorontalo (less than 0.01 $\ \mu g \cdot L^{-1}$)¹³. The arsenic in water of stream near Talawaan–Tatelu mining is also higher than the world's average arsenic content in the river's water (0.62 $\ \mu g \cdot L^{-1}$)¹¹. However, arsenic in stream water near Talawaan–Tatelu mining is lower than the Indonesian water quality criteria for class I (water that is intended to be used for drinking water) and class IV (water that is intended to be used to irrigate crops), where the content of criterion i.e., 50 $\ \mu g \cdot L^{-1}$ and 1 000 $\ \mu g \cdot L^{-1}$, respectively¹².

Table 1. Heavy metals content (μg/L) in water of stream near Talawaan–Tatelu (TT) mining, others ASGM mining, world's average of metals in river water, and Indonesian water quality index.

No	Metals	Metals content ($\mu g \cdot L^{-1}$) in river near ASGM mining			World's average	Water qual. index		
	dissolved	Stream near TT mining	Batudulango R. Gorontalo	Citeluk R. Banten	Ciupih/Cisoka R. Banten	of metals in river water	1 st class	4 th class
1.	Arsenic	$10.00 \pm 1.73^{*}$	< 0.01	-	_	0.62	50	1 000
2.	Cadmium	< 0.01	< 0.01	-	< 0.01 to 0.04	0.08	10	10
3.	Chromium	$5.00\pm1.00^*$	-	-	-	0.7	50	1 000
4.	Copper	< 5	< 0.01	-	< 0.01	1.5	20	200
5.	Nickel	< 1	-	-	-	0.8	-	-
6.	Lead	$23.00 \pm 3.46^{\ast}$	< 0.05	-	< 0.05	0.08	30	1 000
7.	Zinc	< 5	< 0.01	-	< 0.01 to 0.02	0.6	50	2 000
8.	Mercury	< 0.05	< 0.05	$3.26\pm4.34^*$	_	-	1	5
	References	This research	13	14	15	11	12	

Noted : * (metals content \pm se) μ g \cdot L⁻¹

Cadmium in stream water near Talawaan–Tatelu mining is relatively the same as found in the water of Batudulanga River (less than 0.01 μ g · L⁻¹)¹³ and in the water of Cisoka and Ciupih River near Ciberang mining, Banten (ranging from less than 0.01 to 0.04 μ g · L⁻¹)¹⁵ because the content of cadmium in these rivers are generally below the detection limit. Cadmium in stream water near the Talawaan–Tatelu mining is also relatively similar with the world's average content of cadmium in the river's water (0.08 μ g · L⁻¹)¹¹, but lower than the Indonesian water quality criteria for class I and IV (both 10 μ g · L⁻¹).

For chromium, is not obtained the reference comparison about chromium content in water of rivers near ASGM mining areas in Indonesia. The content of chromium in stream water near Talawaan–Tatelu mining can only be compared with the world's average content of chromium in the river's water¹¹ and the Indonesian water quality criteria¹². Chromium in river water near Talawaan–Tatelu mining is relatively higher than the world's average content of chromium in the river's water¹¹ and the Indonesian water quality criteria¹². Chromium in the river's water (0.70 $\mu g \cdot L^{-1}$) but lower than the Indonesian water quality criteria for class I (50 $\mu g \cdot L^{-1}$) and class IV (1 000 $\mu g \cdot L^{-1}$). Both chromium criteria have the chromium content in the speciation form of Cr⁶⁺.

In comparison to copper, its concentration in the stream water near Talawaan–Tatelu mining is relatively the same as in the water of Batudulanga River (less than 0.01 μ g \cdot L⁻¹)¹³ and in water of Ciupih and Cisoka River (less than 0.01 μ g \cdot L⁻¹)¹⁵, because the copper in these streams is generally below the detection limit. Copper in the water of these rivers is relatively lower than the world's average content of copper in the river's water (1.5 μ g \cdot L⁻¹)¹¹ as well as Indonesian water quality criteria for class I (20 μ g \cdot L⁻¹) and class IV (200 μ g \cdot L⁻¹)¹².

For nickel, there is no comparative reference from the rivers near the ASGM mines in Indonesia and also with the Indonesian water quality criteria¹². Nickel content in the water of stream near Talawaan–Tatelu mining is relatively the same or even lower (because it is below the limit of detection, $1 \ \mu g \cdot L^{-1}$) than the world's average content of nickel in the river's water (0.80 $\mu g \cdot L^{-1}$)¹¹.

Lead content in the water of stream near Talawaan–Tatelu mining is high, $(23.00 \pm 3.46) \ \mu g \cdot L^{-1}$, when compared with the water of Batudulanga River¹³ and Cisoka–Ciupih River¹⁵ (both less than 0.05 $\ \mu g \cdot L^{-1}$) as well as

the world's average content of lead in the river's water¹¹ (0.08 μ g · L⁻¹), but lower than the Indonesian water quality criteria for class I (30 μ g · L⁻¹) and class IV (1 000 μ g · L⁻¹)¹².

For zinc, the concentration in the water of stream near Talawaan–Tatelu mining is relatively similar to the water of Batudulanga River (less than 0.01 μ g · L⁻¹)¹³ and Cisoka–Ciupih River (ranging from less than 0.01 μ g · L⁻¹ to 0.02 μ g · L⁻¹)¹⁵, because it is below the detection limit. Zinc in these rivers is relatively lower than the world's average content of zinc in the river's water (0.60 μ g · L⁻¹)¹¹ and the Indonesian water quality criteria for class I (50 μ g · L⁻¹) and class IV (2 000 μ g · L⁻¹)¹².

Finally, the content of mercury in the water of stream near Talawaan–Tatelu mining is relatively similar to the water of Batudulanga River (less than 0.05 μ g · L⁻¹)¹³. However, the mercury in the water of the two rivers is lower than in water of Citeluk River near Ciberang mining, Banten (ASGM mining activities actively doing excavation on the cliffs of the river) which ranged from 3.26 μ g · L⁻¹ to 4.34 μ g · L⁻¹ ¹⁴. For mercury, no comparative data of the world's average content of mercury in the river's water is available¹¹. Mercury in water of stream near Talawaan–Tatelu mining is lower than the Indonesian water quality criteria for Class I (1 μ g · L⁻¹) and class IV (5 μ g · L⁻¹)¹².

In Talawaan–Tatelu mining, amalgamation technique was still the only option until mid of 2002 although the government of North Sulawesi had facilitated technology transfer from mercury to cyanide in 2000 that began with a demonstration of making gold processing technology with cyanide. Over time, the processing of gold which was originally only using the amalgamation technique began to be combined with cyanide techniques. Most miners stored amalgamation tailing in sacks for sale on processors that have a cyanide tank, while some miners directly sold the gold ore which had been milled (without amalgamation)¹⁶.

The world's average content of heavy metals in river's water¹¹ can be used to describe the level of natural availability of each metals in the river water. The availability of arsenic, cadmium, chromium, copper, nickel, lead, zinc, and mercury, specialized both in river water and in the surrounding environment should be considered for mining processing especially for those who use cyanide tank because these metals can form complexes with cyanide. The availability of these metals could affect the efficiency of cyanide to bind the gold^{17–20}.

From the comparison of the water quality criteria, at first glance it seems that the content of all these heavy metals does not encroach on the water quality criteria. However, when examined more deeply especially when viewed verse-by-verse explanation of this rule there is a provision which in fact has been overlooked. In Explanation of Government Regulation No.82 of 2001¹², especially the explanation of Article 4 Section 1 states that: "Water quality management is intended to maintain the quality of water for the purpose of preserving the function of the water, through preservation or control. Preservation of water quality conditions intended to maintain water quality as their natural condition." Furthermore, in the explanation of Article 4 Section 3 states that: "The nature of water in the water sources in the protected forests, springs and deep ground water aquifer, the quality is generally very well. The water in these water sources will also be difficult to rectify the quality if contaminated and it took many years to recover. Therefore, it must maintain its quality as natural condition. The springs water quality should be preserved as natural condition both the springs inside and outside the protected forests..."

It has been argued earlier, that there are springs in the upstream of small rivers that became the focus of this research. From the results of field observations⁸, we found that the area around the spring often flowed with liquid waste from tailing ponds during the amalgamation processing or when the water flow increases due to rain. With these circumstances, it is difficult to determine the condition of the natural springs that exist at this point. The information can be used as an approach to the natural condition of the springs is the world's average of metal content in the river's water¹¹.

From the comparison, three of the eight heavy metals indicating higher rate than the world's average content of heavy metals in the river's water i.e., arsenic (16.13 times), chromium (7.14 times), and lead (287.5 times). Five other metals can not be compared because their concentration were below the detection limit. When the world's average content of metals in the river's water is used as an approach toward natural conditions, the maintenance of water quality measures (especially for arsenic, chromium, and lead) should be done to minimize the threat of a negative impact on the availability, efficiency, carrying capacity, holding capacity, and productivity of the river's springs. This action needs to be done, keeping in mind that the water in the downstream of this small river is used directly by the surrounding community.

4. Conclusion

In general, the metals content in water of stream near the Talawaan–Tatelu mining is higher than in the rivers near the other ASGM mining in Indonesia and the world's average content of metals in the river's water, but lower than the Indonesian water quality standard. As a recommendation, the maintenance of water quality measures should be done to minimize the threat of negative impact toward the surrounding residents.

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