

Article

A Case Study of Heavy Metal Pollution in Water of Bone River by Artisanal Small-Scale Gold Mine Activities in Eastern Part of Gorontalo, Indonesia

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Abstract: Bone River, which is located in the eastern part of Gorontalo, Indonesia, has an issue of contamination by heavy metals due to artisanal small-scale gold mine (ASGM) activities. This river is used as a source of water by inhabitants living along the river, due to the lack of another clean water supply. The purpose of this study is to investigate the concentration of As from the mining site alongside Bone River, and to reveal the As pollution source. Water and sediment samples were collected from ASGM and along Bone River to the delta. The concentrations of As, Hg, and Pb in water samples were identified by inductively coupled plasma mass spectroscopy (ICP-MS), while concentrations in sediment samples were determined by particle-induced X-ray emission (PIXE). Results showed that the concentrations of As, Hg, and Pb in water ranged from 66 to 82,500 µg/L, 17 to 2080 µg/L, and 11 to 1670 µg/L, respectively. These levels exceeded, by 1000 to 10,000 times, the safe limits of drinking water defined by the World Health Organization, which indicated that Bone River water is not safe for drinking or cooking purposes.

Keywords: heavy metal; metalloid; ASGM; river water

1. Introduction

Heavy metals are produced from natural and anthropogenic sources and can build-up in sediments, having significant environmental implications for local communities, as well as for river water quality [1]. Heavy metals are known to be toxic and containing carcinogenic metalloids that can cause cancer in the skin, lungs and urinary tracts; cardiovascular disease; neurotoxicity; and diabetes [2–7]. Gold mining is a source of heavy metals, one of the most serious environmental problems in the world. Specifically, artisanal small-scale gold mines (ASGM) are an important source of heavy metals because the extracted material may contain harmful metals, such as As and Pb. ASGM commonly use Hg for gold extraction because it is quite cheap, can be handled easily, and can form an amalgam with gold. Due to the lack of attention to health awareness, the miners usually ignore the health risk associated with Hg during amalgamation [8]. Miners in ASGM have also used cyanide instead of mercury for gold extraction since 1889 [8–11], as was the case in Huanca village, Peru, where mercury and arsenic concentrations were found in surface soil samples of the area of Huanca settlement and the agricultural land adjacent to the Acarí River. It was suggested that those concentrations were linked to the gold ore as they reached maximum concentrations (48.4 mg·kg⁻¹)

at the site of an old grinding process in the mining site [8]. People in Bone Bolango Regency are concerned that the water was contaminated by heavy metals and metalloids due to ASGM.

Within the aquatic environment, the sediments have been broadly utilized as natural pointers for the assessment of metal contamination in the water [12]. Sediments settle, diminishing their bioavailability. However, dangerous substances like heavy metals and metalloids are shown in a number of a chemical forms trapped in the sediments. As these can build up to create a reservoir, they constitute a possible peril for the downstream valley [12–14].

The objectives of this study were (i) to investigate the concentration of heavy metals such as As, Hg, and Pb in water from mining sites along the Bone River; and (ii) to reveal the heavy metal pollution source in water from the upper course of Bone River, close to ASGM, to the lower course of the river, near the delta.

2. Materials and Methods

2.1. Research Locality

The present study was conducted along Bone River, which is located in the eastern part of Gorontalo, Indonesia. Artisanal small-scale gold mining activities are located in the upper course of Bone River. Bone River characteristics are 1/1800 to 1/1250 of riverbed slope, 120 m to 220 m of river width, ± 2.3 m of water depth, and about 210 to 870 m³/s of carrying capacity [15].

2.2. Sampling

Water and sediment samples were collected from mine tailings, along with the main stream of Bone River, and the delta of the river (Figure 1). After determining the location of the sampling points, 11 water samples, consisting of two samples from gold mining wastewater and nine water samples from the upper course to the delta of Bone River, were collected in different bottles from the laboratory. Samples of water were acidified with 3% nitric acid, along with other samples, and were transferred to the laboratory for As and other metals analysis.

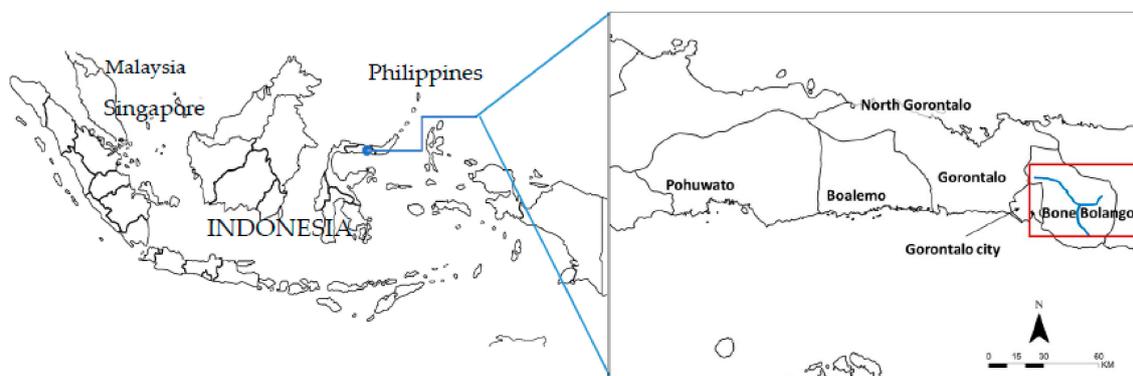


Figure 1. Field site characteristics.

A total of 50 g of sediment samples, from a depth of 20 cm under the riverbed, were collected [5]. The collected samples were coarse sand to clay material (as much as 2 to 0.002 mm) with the wet density from 1 to 2 g/cm³. All the samples were preserved in autoclaved sample bags. The sediment samples were imported under the permit of the minister of Agriculture, Forestry, and Fisheries, Japan in accordance with the Plant Protection Law. The sediment samples were dried in a laboratory for about 48 h at 120 °C. Following this, samples were homogenized by grinding in an agate mortar and stored in labeled bottles until chemical analysis.

2.3. Analytical Method

Each of the collected water samples was first filtrated from the suspension by 0.45 µm membrane filter (Whatman Merck Millipore Corporation, Darmstadt, Germany). The filtrated water samples were diluted with 3% HNO₃ to a final volume of 10 mL for analysis. The concentrations of heavy metal in water samples were determined using a Varian 820-MS instrument (Agilent Technologies, Santa Clara, CA, USA) analysis with inductively coupled plasma mass spectroscopy (ICP-MS) using a Varian 820-MS instrument (Agilent Technologies, Santa Clara, CA, USA) at the Integrated Center for Sciences, Ehime University. All treatments were replicated three times. Reagent blanks and internal standards were used where appropriate to ensure accuracy and precision. A certified reference material (NIST1634f) [16] provided by the National Institute of Standards and Technology (NIST) of Japan was used for analytical accuracy and precision. The analysis of the reference material was accurate, indicating a high level of reproducibility. The analytical calibration curve was created using a multi-element calibration standard (PerkinElmer, Inc., Massachusetts, MA, USA). Rhodium standard solution (Wako Pure Chemical Industries, Ltd., Osaka, Japan) was used as the internal standard. The sediment samples were taken at about 20 cm. They were dried in a ventilated oven at 120 °C for 48 h. The dried samples were ground to a fine size, and homogenized for analysis of particle-induced X-ray emission (PIXE) at Cyclotron Research Center, Iwate Medical University, Japan. For PIXE analysis, the homogenized sediment samples were mixed with Pd-C as an internal standard with the ratio of 3:1, then set up onto a thin film holder by using collodion solution and bombarded with a 2.9 MeV proton beam energy from a cyclotron [17].

2.4. Quality Control

National Institute of Standard and Technology (NIST) CRM 1643f standard reference material [16] and Multi-element standard XSTC-13 from SPEX CertiPrep (NZ, USA) [18] were used to validate the results of As, Hg and Pb in water. The As, Hg and Pb concentrations in sediment were determined in the certified reference material National Institute of Standard and Technology (NIST) SRM 2782 [19].

3. Results

Figure 2. below shows the location of water and sediment samples which derived from ASGM site. In this study, the process of getting gold from ASGM in Bone Bolango Regency includes using two types of gold process (i.e., elemental Hg and cyanide). After digging, crushing and grinding the rock to fine materials, miners mix the Hg for amalgamation. The pH of an aquatic system, and the quantity of trace metals or metalloids present, indicate its water quality. The acceptable normal range of pH, per guidelines suggested by WHO, is about 6.5 to 8.5 [20,21]. The obtained results of pH values of mining (W1) and tailing waters (W2) were 1.98 and 4.00, respectively. This result indicates that the acidic water may release other metal ions such as Pb. On the other hand, the pH value of river water ranges from 7.53 to 7.82 (W3–W11), which indicates the slightly alkaline nature of the river water. The pH in this research is close to the limits recommended by WHO for drinking water.

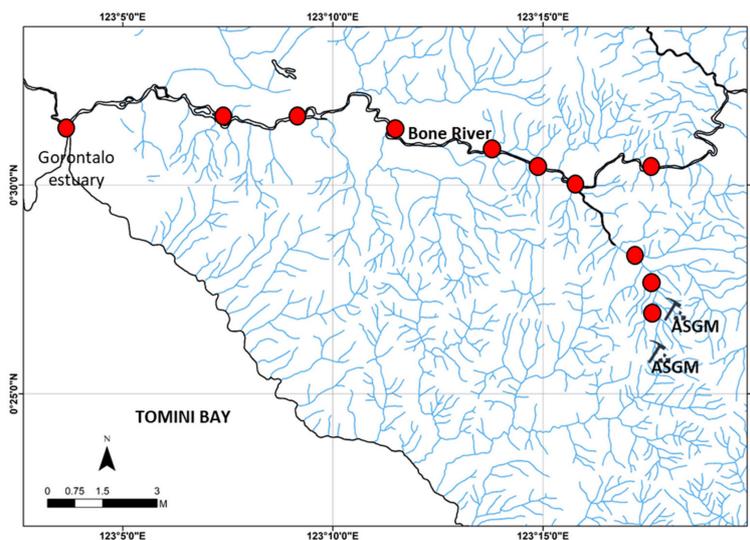


Figure 2. Water and sediment sampling points from the upper course to the lower course of Bone River.

The As, Hg, and Pb concentrations in water and sediment samples from the studied sites are presented in Tables 1 and 2. Concentrations of As, Hg, and Pb in water samples ranged from 66 to 82,500 $\mu\text{g/L}$, 17 to 2080 $\mu\text{g/L}$, and 11 to 1670 $\mu\text{g/L}$, respectively. The results of this study revealed that the water contained As, Hg, and Pb concentrations exceeding the threshold value established by WHO guidelines for water (i.e., 10 $\mu\text{g/L}$, 6.0 $\mu\text{g/L}$, and 10 $\mu\text{g/L}$, respectively) [21]. These results also indicate that concentrations surpassed the limits established by Indonesian regulations for water [22]. The concentrations of As, Hg, and Pb in sediments are shown in Table 2. Concentrations of As, Hg, and Pb in sediment samples ranged from 16 to 120,000 $\mu\text{g/g}$, not-detected (ND) to 790 $\mu\text{g/g}$, and 24 to 10,700 $\mu\text{g/g}$, respectively. These levels indicate that the sediment in Bone River is contaminated by those heavy metals and metalloids. The ND data can be obtained when a concentration is lower than the detection limit of the analytical instrument.

Table 1. Heavy metal concentrations in water samples from the upper course to the lower course of Bone River.

Sample	As		Hg		Pb	
	Concentration ($\mu\text{g/L}$)	SD	Concentration ($\mu\text{g/L}$)	SD	Concentration ($\mu\text{g/L}$)	SD
W1	118	7	176	35	135	1
W2	66	5	489	21	22	7
W3	103	3	102	27	83	3
W4	220	3	2080	6	1670	2
W5	82,500	8	28	26	135	5
W6	1690	1	744	11	581	2
W7	10,800	2	37	7	26	4
W8	1050	7	16	37	18	6
W9	148	2	71	13	69	3
W10	300	4	24	21	27	6
W11	196	11	24	13	23	6

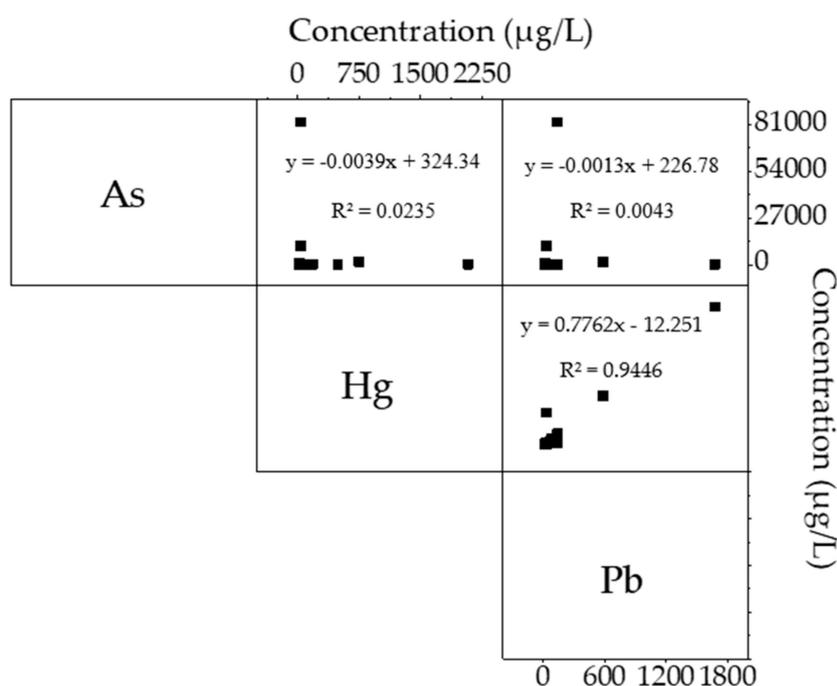
SD: Standard Deviation.

Table 2. Heavy metal concentrations in sediment samples from the upper course to the lower course of Bone River.

Sample	As		Hg		Pb	
	Concentration ($\mu\text{g/g}$)	Error	Concentration ($\mu\text{g/g}$)	Error	Concentration ($\mu\text{g/g}$)	Error
W1	62,100	5020	790	230	2660	1210
W2	27,600	1380	ND	ND	843	891
W3	120,000	23,100	ND	ND	10,700	4192
W4	2030	77.4	57.9	22.3	4030	177
W5	16.7	20.3	21.3	14.3	130	27.3
W6	42	6	ND	ND	132	25
W7	23.1	1.8	ND	ND	24.1	6.2
W8	4.6	5.5	ND	ND	109	22
W9	23.7	7.1	33	12.4	82.2	27.2
W10	1250	61	ND	ND	1320	149
W11	25	9	26	7	59	26

Error: analytical error; ND: Not-detected.

The correlation between each element, which is shown in Figure 3, indicates that there is a positive correlation between Hg and Pb concentrations in water. Hg concentrations increase as Pb concentrations increase. In this study, the positive correlation between Hg and Pb is not well understood.

**Figure 3.** Relationship between As, Hg, and Pb in water samples from the upper course to the lower course of Bone River.

4. Discussion

4.1. Heavy Metal Risk to the Aquatic System Due to ASGM Activities

ASGM is currently practised in more than 70 countries, where it is mostly little acknowledged, unregulated, or illegal. This represents a large global emission source of Hg to the atmosphere, with estimates ranging from 20 to 37% of anthropogenic Hg emissions [23–25]. The above result clearly shows that the Hg contaminations were originally from anthropogenic sources, which come from the use of mercury in the gold process. Most ASGM activities take place alongside streams and rivers because these activities require access to water for operations; this can pose a significant threat

to aquatic ecosystems [26]. In this case, the Hg could be released into the aquatic system (e.g., at a watercourse near the ASGM site) and Hg could accumulate at the delta. The heavy metals and metalloids, such as Hg, Pb, As and sulfate ions, could also be leached into the stream and river through the surface and underground workings waste disposal by ASGM activities [27,28]. Thus, the high level of Hg, Pb, and As obtained in this study exist in the environment from those processes in ASGM. These heavy metals and metalloids are harmful to humans and environment.

4.2. Mercury Pollution

This research shows that Bone River has been polluted by Hg, which is commonly known to come from elemental Hg, which is used for gold extraction during ASGM activities. The Hg from mining sites could be released from the gold process and may find its way, in another form like methylmercury, into the water of Bone River and finally into the estuary of Gorontalo city. Hg is harmful to the human body, as it attacks the central nervous system and can lead to Minamata disease [29–32]. In this study, the Hg pollution in Bone River water comes from anthropogenic sources (i.e., from ASGM activity).

4.3. Arsenic Pollution

Either by natural or anthropogenic causes, high As concentrations in the environment are usually related with As-bearing parent rock materials also commonly present in sulfide rock deposits as As-sulfide minerals [28]. As concentrations in water from natural processes can be found in high concentrations, from 0 µg/L to more 5000 µg/L, but are rarely found in much higher concentrations, especially in groundwater [33]. Commonly, As is mobilized in water when the pH value is about 6.5 to 8.5, under both oxidizing and reducing conditions [33]. Gold mining activities, which extract the ore from these rock deposits, will produce acid rock drainage (ARD), which has high acidity and can result in elevated levels of sulfate (SO_4^{2-}), As, Fe and other toxic metals [34]. It may be a natural source of As pollution into the groundwater.

The gold ores and concentrates, containing some minerals that include metals and metalloids, need to be destroyed to get the gold [9,35]. The gold-bearing gold ore also contained As, crushed and spread over liquid mercury to remove the gold. In this study, the gold extraction processes not only used an elemental Hg, but also cyanide. Other miners use both to produce the gold where, after crushing and milling the rocks into a fine grain and mixing with Hg, the fine materials are put into a big box and are mixed with cyanide. Results of this study show extremely high concentrations of As in river water, compared to the natural As contamination of underground water in Bangladesh. The As concentration of underground water in Bangladesh reached up to about 500 to 4000 µg/L, which is above the 50 mg/L limit set for drinking water in Bangladesh, and the 10 mg/L recommended by the World Health Organization [9,20,36]. The As concentration in water during this study reached up to 82,600 µg/L, suggesting that the As contamination in this river water was not just from natural sources that occurred in a gold ore which contains As. Many other signs and symptoms have been reported in patients that have chronically ingested As, such as in West Bengal, India and Bangladesh [37–39]. In West Bengal, respiratory symptoms, crepitation, liver enlargement, and evidence of chronic arsenical dermatosis and hepatomegaly were prevalent [40,41]. Previous studies mentioned that skin cancer caused by As is usually accompanied by non-cancerous skin reactions and that the most common early symptoms are gastrointestinal, including diarrhea and abdominal pain [37,42].

In the cyanidation process of getting gold, arsenic sulfides in the presence of oxygen are oxidized to arsenite and arsenate, with the distribution of each dependent on oxidation potential and pH [9]. The possibility of As being leached from the gold-cyanidation process is also related to the pH, where in this study the miner is setting up the water and material conditions to be alkaline. Under alkaline conditions, gold can be found by leaching cyanidation, the arsenites could be formed on the dissolution of their sulfides [43].

4.4. Lead Pollution

ASGM activity, which includes techniques such as digging, crushing, panning and mixing that can be mixed with heavy metals and metalloids associated with gold such as As, Hg, and Pb [27,44–46]. In this research, the concentration of Pb in water was higher than the WHO regulation, which is about 10 µg/L [20]. This result shows that Pb is toxic to human as well because it can cause severe gastrointestinal symptoms initially although no abnormalities were found on physical examination [47]. Even though Pb does not dissolve easily into the environment, it can mix with soil particles or dust and enter underground water or drinking water following rain and surface water runoff. Exposure of Pb can occur when eating food cultivated on soil with high Pb concentrations, drinking contaminated water, breathing polluted air and so on [48,49].

4.5. Practical Implication

The contamination of rivers by chemicals leads to accumulation of these chemical substances in sediment. Thus, deposits act as a major sink of chemical pollutants such as heavy metals and metalloids. Sediments, therefore, contain an environmental record of pollutants that can be useful to establish the pollution history of a domain [50,51]. In this case, the pollutant could be solved to mitigate by the several solutions such as chemical precipitation, cementation, electrodeposition, solvent extraction, reverse osmosis, ion exchange and phytoremediation. Furthermore, constructing a lateral embayment in the river bank could serve as a control area where the pollutants could be naturally stored and then removed [52–54].

5. Conclusions

In this study, Bone River water has been polluted by Hg, high As, and Pb from ASGM activity. We also showed that there are two sources of heavy metal and metalloid pollution in water of Bone River, with both a natural source of pollution and an anthropogenic source (i.e., ASGM). In the case of gold cyanidation, it has been restricted to arsenic and a lesser amount of Hg. The As minerals may dissolve through oxidation processes and in alkaline water, and the As can also be present in solution. However, much about the leaching of As to the environment from the gold processing through cyanidation processes, remains poorly understood. More research is required to elucidate their role and understand the harmful metals and metalloids in the gold process in ASGM.

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Conflicts of Interest: The authors declare no conflicts of interest.

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