Characterization of potential mercury contamination in the ASGM area of Mandailing Natal, North Sumatera

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Abstract. A large amount of Hg is used for gold extraction through the amalgamation process in the Mandailing Natal derived from artisanal and small-scale gold mining (ASGM) area located in the North Sumatera Province, Indonesia. The objective of this work was to characterize the potential contamination of total Hg in water, sediments, and soil in this ASGM area. Sampling was done in two locations in West Panyabungan and Huta Bargot Districts using grab sampling method. Results showed that accumulation of Hg in the studied soils and sediments was the highest, but Hg in aquatic solution was below the detection limit of the analytical method of the studied community wells and rivers. The highest concentration of Hg was found in the river sediments of the Saba Padang irrigation (1,63 mg/kg), and in the soil of the plantation area of Huta Bargot (1,62 mg/kg) respectively. These concentrations may pose a serious problem for aquatic and land life, related ecosystems, and human health. For further study, there is a need to study Hg availability in riverine biota to better understand the cycling of Hg in this ASGM area. Understanding the ecological impacts can assist in the prioritizing of impact mitigation efforts.

1. Introduction

The Mandailing Natal artisanal and small-scale gold mining (ASGM), located in North Sumatera Province, is an important ASGM region in Indonesia. Currently, mercury-gold amalgamation is the most common method of gold extraction used by artisanal miners that represents a highly economical, easy to use, and mercury is readily available for centuries [1,2]. In mining, the amalgamation technique is used to recover precious metals from ore using mercury [3]. Hg amalgamation is based on the formation of Au-Hg alloy, which is subsequently heated to volatile Hg to obtain pure gold. Unfortunately, this method commonly leads to massive Hg contamination in the mining area.

Actually the use of Hg in ASGM already banned by Indonesian government which committed to eliminating and reducing the use of mercury in ASGM as manifested by signing the Minamata Convention in 2013 and through Law Number 11 of 2017 concerning Ratification of the Minamata Convention and its derivative regulations with the Presidential Regulation Number 21 of 2019 concerning the National Action Plan for Mercury Reduction and Elimination (RAN PPM) [4].

Hg is among the most highly toxic trace metals in the environment [5–7]. Many national and international agencies and organizations are interested in investigation of mercury contamination in Indonesia [8–11]. Heumasse, Omar and Demmallino [9] studied the Hg content of the Buru ecosystem...
in Maluku, indicated that Hg originated from the amalgamation process showed higher bioavailability than Hg naturally present in soil minerals. These authors found that Hg in the water has a correlation with Hg in the sediment and macrozoobenthos. Harianja et al. [10] investigated Hg contamination effect and socioeconomic factors in communities living around ASGM operations in Sukabumi Regency. The study found that the inhabitants had elevated hair Hg content, with the majority classified as alert and high levels. Another research conducted by Barkdull et al. [11] compared mercury contamination in four Indonesian watersheds affected by ASGM of varying scale, revealed that Hg concentrations in stream water increased by orders of magnitude from upstream to downstream of ASGM activities.  

In general, Hg is relatively inert with strong volatility under most atmospheric conditions. Thus, Hg is not easily oxidized to less volatile forms by major oxidants, so its residence time is quite long time, estimated in the range of 1–2 years [12,13].

The studied area of Mandailing Natal has around 200 miners and they are all located in alluvial plains in the Sumatera plateau. Mining operations use very traditional equipment (steel milling/drum) and are carried out by men, women, and children. Miners work day and night, and even in abandoned mines to be able to survive. The houses are located next to the mines and along the river. The inhabitants of these areas consume fish from the rivers, and they use the waters for livestock, domestic purposes, and irrigation. Uncontrolled Hg application occurs due to the high usage of mercury as practiced by many miners. Several previous efforts have pointed out the importance of determining the most suitable preventive measures in the area, but local and national authorities have had great difficulties controlling this ASGM operation. This is due to the lack of qualified staff and infrastructures. All these factors collectively contributed to the development of the present critical environment conditions. ASGM Mandailing Natal has been operated for more than a decade but data and information regarding Hg contamination on the environment at this site is rare. The objective of this study was to trace Hg exposure in water, sediment, and soil in two selected areas affected by ASGM in comparison to regulatory threshold levels. In addition, some heavy metals were also measured since most heavy metals in the ores tend to be associated with the release of mercury as the result of ore gold processing. MacDonald [14] pointed out that although mercury remains a concern in ASGM impacted rivers, it is not the only contaminant of concern. Sedimentation and particulate bound elements such as Al, As, Cu, Fe, Hg, and Pb were the main river pollutants resulting from ASGM operations. The outcome of this study might help for taking precautionary steps in monitoring the mercury and other heavy metal contamination in the water and sediment, and to advice the authorities of the health impact that can occur for inhabitant. Furthermore, the research findings might be valuable for local and national governments for proposing Hg phase-out in ASGM as a part of the National Action plan of Indonesia under the ratification of the Minamata Convention.

2. Materials and Methods

The study used survey methods that describe the actual condition of the study area. Sample collections, field observations, and laboratory analysis were carried out.

2.1. Sampling

The study area is located at Mandailing Natal District, North Sumatera, Indonesia. Two different locations in two sub-districts affected by Hg pollution from gold mining and processing activities were selected for this study: West Panyabungan and Huta Bargot. In order to determine the extension of Hg pollution in the study area, water, sediment, and soil sample were collected using grab sampling method [15]. A total of 15 sampling points (11 within the area of Huta Bargot and 3 in West Panyabungan) were sampled, taking into account the position of possible sources of Hg contamination such as areas of storage, production, discharge as well as the possible sinks as can be seen in Table 1 and Figure 1.

Temperature and pH were measured directly in the field using portable pH meter equipment type Orion Star A221, while other parameters were analyzed in the laboratory. Water samples for dissolved metals analysis was filtered at the field with 0.45 μm membrane filter paper and placed in a polyethylene bottles. Reagen grade nitric acid (HNO₃) was added to a pH ≤ 2 for sample preservation. Soil and
sediment samples were taken using a soil spade with caution to avoid contamination between samples. Soil and sediment samples were stored in polyethylene bags. All samples were stored at 4°C and kept in light dark conditions.

### Table 1. Sampling location of water, sediment and soil.

| Location                  | Sample code | Sample type                          | Sampling point                                      | Coordinate                |
|---------------------------|-------------|--------------------------------------|-----------------------------------------------------|---------------------------|
| Huta Bargot               | SUM B1      | Well water                           | Near river                                          | 99° 29' 43.44" 0° 52' 17.94" |
|                           | SUM B2      |                                      | Inhabitant area                                     | 99° 29' 44.7" 0° 52' 24.816" |
| Amalgamation area         | SUM B3      | Well water                           | Close to amalgamation process                       | 99° 29' 40.016" 0° 52' 19.884" |
|                           | T1          | Soil                                 | Plantation area                                     | 99° 29' 38.832" 0° 52' 16.896" |
|                           | T2          |                                      | Near amalgamation area                              | 99° 29' 37.14" 0° 52' 22.944" |
| Aek Latong River          | SAL         | River water and sediment             | Downstream from the gold mining area                | 99° 30' 19.656" 0° 53' 26.808" |
| Aek Siaporas River        | SAS         | River water and sediment             | Downstream from Aek Latong River                    | 99° 29' 37.824" 0° 53' 17.52" |
|                           | SSM 1       |                                      | Upstream (before amalgamation area)                  | 99° 29' 35.736" 0° 52' 11.172" |
| Simalagi River            | SSM 2       | River water and sediment             | Close to amalgamation area                          | 99° 29' 43.44" 0° 52' 17.328" |
|                           | SSM 3       |                                      | Downstream (after amalgamation area)                | 99° 30' 35.388" 0° 53' 17.268" |
| Batang Gadis River        | SBG         | River water and sediment             | Downstream far away from amalgamation area          | 99° 31' 19.272" 0° 53' 51.252" |
| Saba Padang Village       | ISP         | River water and sediment             | Irrigation from Batang Gdis river                   | 99° 30' 14.22" 0° 52' 3.432" |
| West Panyabungan          | SAR 1       | River water and sediment             | Upstream                                            | -                          |
|                           | SAR 2       |                                      | Longat Village                                      | -                          |
| Aek Salir River           | SR          | River water and sediment             | Irrigation in Runding Village                       | 99° 30' 14.292" 0° 52' 3.504" |

#### 2.2. Analytical treatment

Soil and sediment samples were dried at air temperature until the weight was constant. Dry samples were ground to 200 mesh using a ring mill and then sieved. Strong acid digestion was used based on the USEPA standard method 3050B [16]. The samples were digested with repeated addition of nitric acid and hydrogen peroxide and repeated heated on a hotplate without boiling, then followed by the addition of hydrochloric acid. Flame atomic absorption spectroscopy (Flame-AAS) was used for heavy metals determination.

To analyze heavy metals, such as Pb, As, Cr, Cu, Zn, Ni, and Cd in water sample; filtered water samples were analyzed using Flame-AAS instrument based on SNI 6989-84:2019 standard method, and for Hg analysis was based on SNI 6989-78:2019 with Cold Vapour AAS.

#### 3. Results and Discussion

#### 3.1. Chemical analysis results of well water samples

The drinking water quality standard under the Ministry of Health Regulation Number 492/Menkes/Per/IV/2010 regarding the requirement of drinking water quality [17], stated that mercury levels permitted in drinking water is 1 µg/L. Well water sample was collected near the amalgamation
area in Huta Bargot sub-district, and the results of mercury and heavy metals analysis are shown in Table 2. It can be seen that the dissolved mercury and other heavy metals in well water around the houses in Huta Bargot District were below the required quality standard for metal content in drinking water. The total mercury content in SUM B3 which is close to the mercury amalgamation area, was 0.483 µg/L, and this concentration was still below the required threshold.

![Figure 1. Sampling location map.](image)

**Table 2.** The mercury and heavy metals concentration in well water in Huta Bargot Sub-district.

| Sample code | pH  | Hg  | Hg Total | Pb  | As  | Cr  | Cu  | Zn  | Ni  | Cd  |
|-------------|-----|-----|----------|-----|-----|-----|-----|-----|-----|-----|
| SUM B1      | 6.40| <0.346 | <0.346 | 1.900 | <1 | 6.099 | <0.014 | <0.015 | <0.054 | <0.005 |
| SUM B2      | 7.21| <0.346 | <0.346 | 1.252 | <1 | 0.695 | <0.014 | <0.015 | <0.054 | <0.005 |
| SUM B3      | 6.58| <0.346 | 0.438 | <1 | 1.120 | 9.791 | <0.014 | <0.015 | <0.054 | <0.005 |

Ministry of Health Regulation No. 492/2010

6.5 - 8.5 1 - 10 10 50 2.0 3.0 0.07 0.003

Based on information obtained from the community in research area, people use well water for daily needs such as bathing, washing, watering the plantation, and some people use it for drinking water. The quality of well water in the three wells met the water quality for drinking water.
3.2. Heavy metal analysis results of river water and sediment samples

Resident used the rivers in the study area for bathing, washing, agricultural irrigation, farming and drinking for cattle, and catching freshwater fish for consumption. Therefore, the water quality classification falls into the second class of Government Regulation of the Republic of Indonesia Number 22 of 2021 Annex VI about National Water Quality Standard for River Water and the Like [18].

Mercury enters the river in the form of the high density elemental Hg⁰. It will sink to the bottom of the river water and accumulate in the sediment at a depth of 5-15 cm below the sediment surface. The sediment reflects the quality of the surface water and provides important information regarding mobilization of pollutants entering water. The metal content in sediment plays an important role in detecting the sources of pollutants in the aquatic system. Indonesia does not have a sediment quality standard yet for sediments. In this study, Canadian sediment quality guidelines (CSQG) for the protection of aquatic life were used as a sediment quality standard [19], as presented in Table 3.

| Table 3. The Canadian sediment quality standard for the protection of aquatic life. |
|----------------------------------|----------------------------------|----------------------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| Effect for aquatic life          | Hg     | Pb     | As    | Cr    | Cu    | Zn    | Cd    |
| Minimal effect range             | mg/kg  | mg/kg  | mg/kg | mg/kg | mg/kg | mg/kg | mg/kg |
| Possible effect range            | 0.13-0.7| 30.2-112| 7.24-41.6| 52.3-160| 18.7-108| 124-271| 0.7-4.2|
| Probable effect range            | >0.7   | >112   | >41.6 | >160  | >108  | >271  | >4.2  |

Canadian Council of Ministers of the Environment (CCME) use this standard to protect living things in the water. The guidelines consist of threshold effect levels (TELS) and probable effect levels (PELS) to identify the following three ranges of chemical concentrations concerning biological effects [19], which is below the TEL, the minimal effect range within which adverse effects rarely occur; between the TEL and PEL, the possible effect range within which adverse effects occasionally occur; and above the PEL the probable effect range within which adverse effects frequently occur.

Sampling was conducted in November 2019 when entering the rainy season. It is estimated that there has been a dilution by rainwater. Rainwater is able to dissolve various chemical compounds and dilute pollutants that entering rivers. The analysis results of mercury and heavy metal content in river water are presented in Table 4. As for sediments are shown in Table 5.

| Table 4. The mercury and other heavy metals concentration in river water. |
|----------------------------------|----------------------------------|----------------------------------|------------------|------------------|------------------|------------------|
| Sample Code                      | pH     | Hg     | Pb     | As    | Cr    | Cu    | Zn    | Cd    | TSS  |
| Huta Bargot                      | 7.90   | <0.346 | <0.346 | <1    | 23.02 | 4.148 | <0.014| <0.015| <0.054| 9.438| 52.3| 18.7 | 124 | 7.24 | 0.7-4.2|
| SSM 1                            | 7.82   | <0.346 | <0.346 | 10.57 | <1    | 1.80  | <0.1 | <0.014| <0.015| <0.054| <0.005| 29.5 |
| SSM 2                            | 7.56   | <0.346 | <0.346 | <1    | 89.70 | 3.041 | <0.014| <0.015| <0.054| <0.005| 19.5 |
| SSM 3                            | 8.27   | <0.346 | <0.346 | <1    | 2.36  | 9.292 | <0.014| <0.015| <0.054| <0.005| 7.5  |
| SAL                              | 7.69   | <0.346 | <0.346 | <1    | <1    | 3.912 | <0.014| <0.015| <0.054| <0.005| 6.5  |
| SAS                              | 7.47   | <0.346 | <0.346 | <1    | <1    | 9.272 | <0.014| <0.015| <0.054| <0.005| 6    |
| SBB                              | 7.31   | <0.346 | 0.414  | <1    | <1    | 10.77 | <0.014| <0.015| <0.054| <0.005| 140  |
| ISP                              | 8.53   | <0.346 | <0.346 | <1    | <1    | 2.709 | <0.1 | <0.014| <0.015| <0.054| <0.005| 31.2 |
| West Panyabungan                 | 8.41   | <0.346 | <0.346 | <1    | <1    | 9.438 | <0.014| <0.015| <0.054| <0.005| 6    |
| SAR                              | 6      | 0.02   | 0.05   | -     | 10    | 2000  | 50   | 50   | -    | 50   |
| Standard                         | PP 22/2021 | 30 | 1000 | 50 | 0.02 | 0.05 | 10 | 50 |
Table 5. The concentration of mercury and other heavy metals in river sediment.

| Sample Code | Heavy metal content |
|-------------|---------------------|
|             | Hg mg/kg | Pb mg/kg | As mg/kg | Cr mg/kg | Cu mg/kg | Zn mg/kg | Ni mg/kg | Cd mg/kg |
| Huta Bargot |          |          |          |          |          |          |          |          |
| SSM 1       | 0.094    | 16.60    | 83.44    | 46.54    | 21.44    | 80.91    | 12.19    | <0.059   |
| SSM 2       | 0.072    | 14.28    | 98.74    | 47.84    | 21.99    | 84.03    | 12.64    | <0.059   |
| SSM 3       | 0.042    | 15.92    | 72.14    | 62.19    | 21.39    | 79.06    | 16.36    | <0.059   |
| SAL         | 0.182    | 28.45    | 461      | 71.06    | 22.97    | 37.89    | 1.74     | <0.059   |
| SAS         | 0.138    | 21.00    | 90.9     | 47.27    | 26.89    | 70.64    | 4.09     | <0.059   |
| SBG         | 0.024    | 9.66     | 40.11    | 80.63    | 15.59    | 56.84    | 11.75    | <0.059   |
| ISP         | 1.630    | 26.89    | 37.74    | 22.67    | 130      | 141      | 17.36    | 0.507    |
| West Panyabungan |      |          |          |          |          |          |          |          |
| SAR 1       | 0.004    | 16.67    | 8.92     | 61.00    | 19.92    | 78.86    | 20.98    | <0.059   |
| SAR 2       | 0.075    | 13.89    | 11.27    | 43.08    | 22.96    | 85.11    | 21.23    | <0.059   |
| SR          | 0.958    | 20.04    | 33.15    | 22.92    | 76.77    | 135      | 18.46    | 0.586    |          |
| Standard CCME (2001) | 0.13 | 30.2 | 7.24 | 52.3 | 18.7 | 124 | - | 0.7 |          |

Table 4 shows that the content of mercury and other heavy metals in river water was below the class II quality standard (PP 22 of 2021). The highest Pb concentration was found at Simalagi river (SSM 2) that close to residential areas and amalgamation gold processing activities of 10.57 mg/L, but this was still below the threshold limit of 30 µg/L. The total suspended solids (TSS) in Batang Gadis river was found to be high, reached 140 mg/L that exceeded the threshold value of 50 mg/L. Suspended particles can be formed from soil erosion and waste discharge. Batang Gadis river is an estuary for several rivers in Panyabungan, West Panyabungan, Huta Bargot, North Panyabungan, Naga Juang, Bukit Malintang, Siabu and others. Many gold mining and processing activities directly dispose of their wastes into the river. Other sources were from resident activities along the river, such as disposal of domestic waste and agricultural.

From the results of river sediment analysis in Table 5, the rivers in the Huta Bargot area have been polluted by heavy metals that exceed the limit of the CCME quality standards, particularly As. The Aek Latong river (SAL) and Aek Saporas River (SAS) have been polluted by mercury at low level with possible effect range. A high level of Hg was found in the irrigation sediment of Saba Padang Village (ISP). The Hg content in the sediment reached 1.63 mg/kg, indicating that the river was severely polluted by Hg with the probable effect range.

Elevated As concentrations were detected in almost all sampling points. Heavily polluted by As as indicated by very high concentration in sediment with the probable effect range exceeding the value of 41.6 mg/kg were in Aek Latong, Aek Saporas, and Simalagi river. The highest As concentration was found at the downstream of the artisanal gold mining area, namely Aek Latong sediment river which reached 461 mg/kg.

Arsenic levels were highest in arsenide form from amalgam of copper, lead, silver and sulfide from gold. Gold mining and processing activities that have been going on for a long time in this area are considered to be the cause of the high concentration of heavy metals in the sediment.

A high Cu content was also found in almost all sediment samples in Huta Bargot. The highest concentration found was 130 mg/kg, indicating a heavy Cu contamination in the irrigation sediment of Saba Padang Village.

In West Panyabungan, a high Hg content was also found at the irrigation of Runding Village (SR), which indicates that the river has been contaminated with Hg. The high metals content of As and Cu was also found at the irrigation channel in Runding Village and Aek Salir river at Longat Village, which indicates that the river was also polluted by the heavy metals from gold mining activities.
The quality of river water is largely determined by its discharge. The high river flow discharge contributes to the pollutant concentration to be small, but on the other hand, it can result in a high sedimentation rate so that the pollutant level in the sediment is high.

3.3. Mercury content in the soil

Soil samples were taken around the gold processing area using mercury in Huta Bargot Sub-district. Soil analysis results are presented in Table 6.

| Sample Code | pH  | Hg Total mg/kg | Sampling location           |
|-------------|-----|----------------|----------------------------|
| T1          | 6.67| 0.095          | plantation area            |
| T2          | 7.72| 1.62           | near amalgamation area      |

In general, the pH values of the soil at the study location were 6.67 and 7.72, classified as neutral to slightly alkaline. Indonesia does not have any legislation for the classification of polluted soil according to Hg concentration. Therefore, reference values from some researchers of several countries have been used to evaluate the degree of Hg enrichment of the soil samples [20–23].

Generally, the average background (natural) concentration of Hg in soil ranges from 0.03 to 0.1 mg/kg, with an average value of 0.06 mg/kg, whereas Hg-contaminated sites often have soil concentrations that are 2 to 4-orders of magnitude higher [21,22]. The soil microbial community is overly sensitive to Hg concentrations, and this sensitivity is influenced not only by soil properties but also by the plant species growing in the soil. A level of 0.36 mg/kg of Hg in soils is proposed to be a critical concentration in which plant and soil organisms will be affected [23]. A high Hg concentration of 1.62 mg/kg was found in the soil around the processing site using mercury, indicating that the soil has been heavily contaminated by Hg. The Hg value exceeded 1 mg/kg may indicate a threat to food production due to the need to protect human health in areas with higher Hg soil levels [20].

4. Conclusions

The present study has shown the extent of contamination of water, sediment, and soils surrounding ASGM operation in Mandailing Natal. Analytical data have confirmed a high level of Hg pollution in two areas studied of Huta Bargot and West Panyabungan. Hg contents of the majority of water samples from wells and rivers of the two areas were below the detection limit. However, Hg concentrations found in some sediments were high, indicating that enrichment of Hg in the sediments has occurred. Among the study sites, the Saba Padang irrigation and plantation area in Huta Bargot must have received a very high amount of atmospheric deposition of Hg as well. This may be due to the open topographic position of these areas. Future study on the impacts of this ASGM should not be limited to the physical and chemical properties of riverine water and sediment, but also include impacts on riverine biota; the studies should not solely concentrate on Hg. Other pollutants as trace heavy metals (i.e. As, Cu) in the sediment should also be carefully assessed because their impacts on riverine ecosystems can also be significant. Identification and control of sediment release at each step of mining and processing in ASGM operations should be institutionalized and a part of requirements if ASGM is to be formalized as one of the obligations in the environmental impact assessment document.

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