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Analysis of Mercury (Hg) Levels in the Patipulu River Ecosystem, Kaiely Village, Buru Island

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Abstract. Gold deposits on Gunung Botak, Buru Island, were discovered in 2011. Illegal miners extract gold using mercury (Hg) in rotating drums (trommel). For flushing, a lot of water is used so that the drum is placed along the river bank and the resulting waste is disposed of directly into the environment so this is very dangerous for humans and the environment. One of the areas where the drum is located in the river bank of the Patipulu River, Kaiely Village. This research was conducted to determine the levels and distribution of mercury at the ex-trommel location and the Patipulu River ecosystem. The results showed that tromol waste had a mercury concentration of 22 ppm or 22 times higher than the standard (SNI for mercury levels in the sediment was a maximum of 1 ppm) while river sediments were 16 times higher. From the data obtained, it can be concluded that the Patipulu River ecosystem has been contaminated with mercury.

INTRODUCTION

Many developing third-world nations have an abundance of gold imbedded in rural areas of the country. Artisanal Small Scale Gold Mining (ASGM) generally done in Asia, Africa, nd Latin America, for example in Mindanao, Filipina [1], Colombia [2], Chili [3], Sub-sahara, Africa [4], and Zimbabwe [5]. The same problem also occurs in several ASGM in Indonesia, such as in North Sulawesi [6] and Palu-Central of Sulawesi [7].Gold was discovered in 2011 at Mount Botak on Buru Island, in the Mollucas Province, Indonesia. During 2012, artisanal gold mining in the Wamsait village area of the Wae Apu district had become rampant and uncontrolled with large population influxes to the Island [8].

The amalgamation technique is a gold processing technique by mixing rock/material containing gold and mercury using trommel (steel drums).The use of the amalgamation method has an impact on the environment and human health through the accumulation of mercury in the environment [8]. Mercury (Hg) is a heavy metal that is classified as the most dangerous pollutant, because it is a neurotoxin, both for organisms and humans [9,10]. As a heavy metal, mercury that enters the aquatic environment will be difficult to degrade.Besides being concentrated in water, the mercury will also settle in the sediment.

Mercury can undergo various transformations in the environment, and Hg ion can be converted into one of its most toxic forms, methyl mercury (MeHg), by both abiotic and biotic pathways [11], and furthermore, it can be accumulated through the process of bioaccumulation and biomagnification, namely through the food chain in the body tissues of aquatic animals, so that mercury levels can reach dangerous levels for both aquatic animal life and human health who consume aquatic animals that have been contaminated with mercury. Rivers that have been contaminated with mercury flow through several villages along the riverbanks and empty into Kaiely Bay.Some rivers on Buru Island that have been contaminated with mercury are the Waeapo, Waelata, Anahoni, and the Marloso River [8,12]. The water of Kaiely Bay is covered with various mangroves with very high density.The

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mangrove ecosystem is ideal as a place to spawn various marine biota (fish, shrimp, shellfish, squid, etc.) as well as a center for seaweed cultivation.Kaiely Bay is connected to the Banda Sea, which is the spawning center for various types of tuna from around the world.Maluku Region will also be designated as the National Fish Stock. Research conducted shows that marine biota in Kaiely Bay has been polluted by mercury [13].

The Patipulu River is a river that flows through Kaiely Village - the largest and oldest village on the Kaiely Bay Coast.Prior to mining activities, the Patipulu River was used as a source of fresh water by residents of Kaiely Village.When the mining activity was started, villagers who did not understand the impact of using mercury built drum in backyards of their houses or in gardens that were planted with staple food. Even though mining activities have been stopped since 2017 and there is no more trommel operating, the waste from ore processing still contains high concentrations of mercury. Research result from Reichelt *et al* [14] show that trommel waste from the Marloso area closed to the Gogrea mine site has mercury levels of 203 mg kg of total Hg (THg).

The location of the trommel in Kaiely Village is on the Patipulu's River banks so that when it rains, the water runoff from the trommel sites that contains mercury will pollute the waters of Kaiely Bay through the Patipulu River.To determine the pollution status of the Patipulu River, a study was conducted to determine the concentration of mercury at the former tromol location (tailings) and river sediments.Parameters were measured include total organic carbon (TOC), analysis of elemental composition in samples using X-Ray Fluoresence (XRF), analysis of the crystallinity of metals compounds using X-Ray Diffraction (XRD) and determination of total mercury content (THg) using CV-AAS (Cold Vapour-Atomic Absorption Spectrometry).

FIGURE 1. Site sampling on the Patipulu River

EXPERIMENTAL

Taillings and River Sediment Samples Collection

Samples of River sediments were collected in September 2019. Tailings samples were collected from near the Pattipulu's River bank (Fig.1). Tailings samples were collected from the surface (0–10 cm) sediments of a settlement pond adjacent to a trommel site (ore processing plant) in the Kayeli Village. River, estuary, and marine sediment samples were collected downstream of trommel sites. River samples were collected by hand and by van Veen grab at deeper marine locations (Table 1). Samples were stored in polyethylene bags and transported on ice to the University of Pattimura, Ambon where they were immediately frozen.

TABLE 1. Details of samples taken from along Patipulu River, Kaiely Vilage

Sample Code	Sample Type	Location
SK-1	River sediments	S -3.38570 E 127.11156
SK-2	Trommel waste	S -3.38545 E 127.11146
SK-3	River sediments	S -3.38529 E 127.11135
SK-4	Trommel waste	S -3.38513 E 127.11142
SK-5	River sediments	S -3.38501 E 127.11145
SK-6	River sediments	S -3.38487 E 127.11151
SK-7	River sediments	S -3.38469 E 127.11155
SK-8	River sediments	S -3.38262 E 127.11111
SK-9	River sediments	S -3.38206 E 127.11158
SK-10	The estuary of Patipulu River (offshore sediments)	S -3.38154 E 127.11164

Taillings and River Sediment Samples Processing

Samples were dried in an oven at 40 ºC. The dried samples were crushed using a mortar and pestle and then sieved using a 100 mesh sieve.The sample was weighed 5 g, put in a plastic vial for further analysis.The sample was put in a beaker glass, added with 5 mL of concentrated H_2SO_4 solution and 5 mL of HNO_3: $HClO_4$ (1:1) solution.The glass beaker is heated on a hot plate to produce a clear solution.Once cool, the solution is filtered and diluted in a volumetric flask.Blank solutions are prepared in the same way but without samples. Ten milliliters of the sample solution was piped and added 0.1% 0.1 mL of $KMnO_4$ solution, and while shaking, adds 0.1 M hydroxyl-amine hydrochloride and 0.5 mL of $SnCl_2.2H_2O$ solution.Mercury content analysis was performed using a CV-AAS (Cold Vapor-Atomic Absorption Spectrophotometer).

Determination of Total Organic Carbon (TOC)

Total organic carbon (TOC) was measured using the method was adopted from US-EPA [15]. Five grams of each sample was placed in a 50 mL beaker and 30% H_2O_2 was added until the sample frothing ceases.The samples were heated to 90 ºC during peroxide addition to increase the speed and completeness of the peroxide digestion. After the digestion process is completed, the sample is dried at 105 ºC, cooled in a desiccator and weighed.

RESULTS AND DISCUSSION

Metals in Sediments and Trommel Wastes

Total mercury (THg) concentrations in trommel wastes and rivers sediments from Patipulu River, Buru Island, Indonesia was exceeded SNI and ANZECC/ARMCANZ) [16] limits in all samples, and by up to 22 times. The highest concentration was founded in the trommel waste sample (site SK-8; 22.106 mg/kg, on site SK-4; 22.11 mg/kg), and the lowest concentration was measured in Patipulu estuary/offshore sediment (site SK-10; 0.142 mg/kg). Kayeli Village is located on the coast of Kayeli Bay, approximately 5 km from the ASGM area on Gunung Botak, but the ASGM operators transported material from Gunung Botak to Kaiely Village and set up many trommel units to extract gold.This method requires large volumes of water for flushing and results in the deposition of mercury into the upper catchment of the Patipulu river system. Although ASGM activities on Gunung Botak officially ceased in 2017, but at the time of sampling in June 2019, there was one trommel unit still operating in Kayeli Village, on the of Patipulu River bank (on SK-4 site, with THg 22.11 mg/kg).Whereas in the ex-trommel site was abandoned for four years (SK-2) the THg concentration was 16.5 mg/kg. The Hg concentration in the waste at tromel site waste is very high, this is probably due to rain. Rainwater runoff will carry mercury deposits so that it will pollute the Patipulu River and thus flow into Kaiely Bay.

TABLE 2. Total Organic Carbon (%TOC) and Total mercury (THg) in River sediments and trommel waste samples

Sample Code	Sample Type	%TOC	THg (mg/kg)
SK-1	River sediments	5.47	10.34
SK-2	Trommel waste	4.20	16.15
SK-3	River sediments	1.60	5.506
SK-4	Trommel waste	2.69	22.11
SK-5	River sediments	2.50	1.10
SK-6	River sediments	2.65	1.32
SK-7	River sediments	0.99	0.35
SK-8	River sediments	1.57	2.46
SK-9	River sediments	1.84	0.18
SK-10	The estuary of Patipulu River (offshore sediments)	0.54	0.14

Percentage of Total Organic Carbon (TOC) (Table 2) linear with THg conccentrations. Total organic carbon (TOC) are the most important components of sediments and soils as it can be used to distinguish marine and terrestrial sources of organic matter, environmental depositional conditions, pollution indices, and soil quality and productivity indicators [17]. The soil and sediments organic carbon is mainly derived by decomposition of the plants and animals or plankton or anthropogenic sources such as chemical contaminants, fertilizers or organic-rich waste. Organic matter is one of the important components of bottom sediments and it is mostly responsible for binding metals. It has been estimated that organic matter can bind up to 95% of the divalent mercury species [18].

All waste trommels samples and some river sediment samples exceeded the acceptable Hg concentrations of 1 mg/kg recommended for aquatic sediments in Indonesian (SNI), Australian and New Zealand (ANZECC/ARMCANZ 2000) waters by between 5 and 22 times (Table 2). Comparisons with the Male et al. (2013) study show that THg concentrations in Kayeli Village Estuary samples which collected in May 2013, at the same site (SK-10) was 1.17 mg/kg but in the current study, after seven years, THg concentrations still remain (0.14) (Table 2; [8]. This study shows high enough of THg in receiving aquatic environments of Patipulu River at Kayeli Village, Buru Island, Indonesia. From this, there is potential for food chain contamination and because of the close relationship of the villagers with the environment in relation to food resources and food security there is a further risk of human health concerns.

X-ray Diffraction and X-ray Fluorescence Analyses

Selected samples (SK-1, SK-4 and SK-8) based on the XRD and XRF analysis results show the primarily composed of quartz (SiO_2) (Table 3). Site SK-1 (River sediment) were composed of between 5–30% Muscovite ($KAl_2(Si_3Al)O_{10}(OHF)_2$, Cronstedtite ($Fe_3FeSiO_4(OH)_5$ and trace amounts of titanite ($Ca(TiO)(SiO_4)$). SK-8 (River sediment) were composed of between 5–30% Ilite $(K,H_3O)Al_2Si_3AlO_{10}(OH)_2$, Phillipsite $(K_2)CaAl_2Si_4O_{12}xH_2O$ and trace amounts of Biotite $K_2(Fe_2MgTi)(Al_2SiO(OH)_4)$. The trommel site samples (SK-4, Figure 2) were composed of between 5–30% Muscovite ($KAl_2(Si_3Al)O_{10}(OHF)_2$, Biotite $K_2(Fe_2MgTi)(Al_2SiO(OH)_4$and trace amounts of kaolinite $(Al_2(Si_2O_5)(OH)_4$.

TABLE 3. XRD analysis of river sediments and trammel waste samples from Patipulu Rivers, Buru Island, Indonesia

Sample	Major (100-30%)	Minor (5-30%)	Trace (<5%)
SK-1	Quartz (SiO_2)	Muscovite Cronstedtite	Titanite
SK-4	Quartz (SiO_2)	Muscovite Biotite	Kaolinite
SK-8	Quartz (SiO_2)	Ilite Phillipsite	Biotite

XRD results are supported by XRF; quartz (SiO_2) was the major component of all samples. River sediments samples (SK-8 and SK-1) were lower in SiO_2 content (59.9% and 49.3% respectively) and the trommel site (SK-4) had the highest SiO_2 content of 66.14%. Aluminum oxide (Al_2O_3) was the second most abundant component in all samples; River sediments samples (SK-1 and SK-8) contained 18.7% and 15.2% Al_2O_3 respectively. Trommel waste sample (SK-4) was lower in Al_2O_3 content (9.7%), but contain HgO (0.1%). Iron oxide (Fe_2O_3) and potassium oxide (K_2O) were the only other compounds with concentrations above 1%. Fe_2O_3 concentrations were similar at the two river sediment sites (SK-1 and SK-8) of 7.4% and 8.6% respectively, and the trommel waste sample (SK-4) contained 7.8% Fe_2O3.

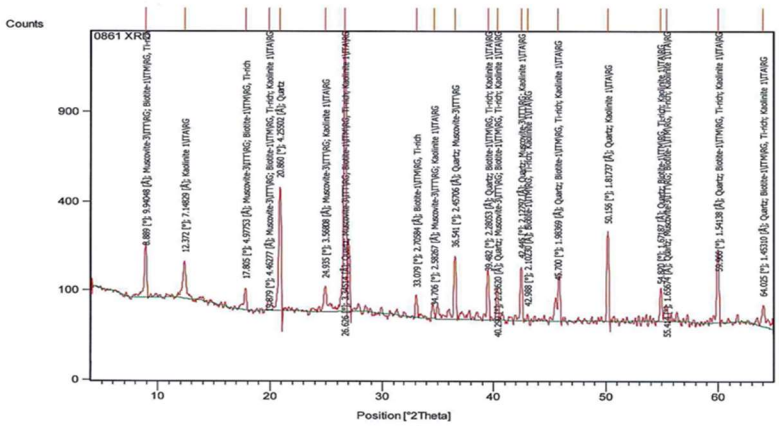

FIGURE 2. XRD Diffraction patern of trommel waste sample (SK-4)

CONCLUSION

This study shows high enough of THg in receiving aquatic environments of Patipulu River at Kayeli Village, Buru Island, Indonesia. From this, there is potential for food chain contamination and because of the close relationship of the villagers with the envrionment in relation to food resources and food security there is further risk of human health concerns. This study demonstrates the potential magnitude of ecological and human contamination issues on Buru Island in the short and long term, warranting urgent scientific and governmental investigation and intervention.

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