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Mercury Contamination in Selected Edible Plants and Soil from Artisanal and Small-scale Gold Mining in Sukabumi Regency, Indonesia

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Abstract

Artisanal and small-scale gold mining (ASGM) activities often pollute soil, water, and air, thereby achieving widespread proliferation, and contaminating the surrounding biota including plants. Mercury contamination on agricultural land around ASGM areas has been widely reported. This study aims to determine the total mercury contamination in plants and soil around active ASGM sites in Sukabumi Regency, Indonesia, namely, Waluran, Lengkong, and Ciemas Districts. Total mercury (Hg) content was measured from 27 plant samples (including cassava [*Manihot utilisima*], rice [*Oryza sativa*], and papaya [*Carica papaya*]), 7 rhizosphere soil samples, and 7 non-rhizosphere soil samples. Data were analyzed using Kruskal–Wallis test. Results showed no significant difference in total mercury concentrations among locations or plant parts, between rhizosphere and non-rhizosphere soils ($p > 0.05$), and among cassava plant parts, papaya, and rice. The highest mercury level was found in cassava (0.33–43.27 ppm). Mercury contamination in rice and papaya was relatively low at 0.03–1.22 and 0.06–5.11 ppm, respectively. According to the Regulation of the Head of BPOM of the Republic of Indonesia Number 23 of 2017 concerning the Maximum Limit of Heavy Metal Contamination in Processed Food, 0.03 ppm is the maximum limit of mercury contamination in fruits, vegetables, and cereals. Therefore, all plant samples around the ASGM sites have exceeded the maximum mercury contamination and thus are not suitable for consumption.

Keywords: ASGM, cassava, mercury, soil, Sukabumi

Introduction

Artisanal small-scale gold mining (ASGM) is carried out by individual miners or small businesses with limited capital investment and production. Gold ore processing using mercury is a common ASGM practice because of its effectiveness and low cost [1]. Mercury is mixed with the ore to form an amalgam, which is then heated or burned to release its mercury component and extract the gold [2]. Elemental mercury released during this process can bind with other elements to create different compounds and become soluble in sediment and aqueous substance [3]. Humans can experience various health problems due to mercury exposure through oral, skin, and inhalation. Examples of health problems caused by mercury exposure are neurological disorders, skin problems [4], infertility, and hormonal disorders [5]. In addition, mercury pollution inhibits plant growth and affects soil fertility [6–8]. The bio magnification of this element in humans occurs through the food chain [9, 10].

The increasing number of traditional gold miners in Indonesia is not exclusively located in Sukabumi. The Bogor District government stated that 6,000 people or 30% of its population work as gold miners [11]. In 2013, the average accumulation of mercury in the hair samples of ASGM workers in Cisarua Village, Bogor was 2.03–9.04 ppm, and 24 (60%) employees experienced mercury poisoning of more than 2 ppm [12]. West Nusa Tenggara and North Sulawesi provinces are among the ASGM hotspots in Indonesia. Mercury contamination was observed in fish and drinking water from the ASGM area in Sekotong, West Lombok [13, 14], and high mercury levels were found in miners' urine and hair [15]. A similar situation was reported in Tatelu, North Sulawesi. Almost half of the fish samples from this location have mercury levels exceeding the WHO threshold for human consumption [16], and high mercury levels were detected in grass, soil, water, fish, and shellfish [17]. Therefore, identifying and evaluating the levels of mercury contamination in ASGM sites are of great importance.

Rice (*Oryza sativa*), Sukabumi papaya (*Carica papaya*), and cassava (*Manihot utilisima* are some of the commodities produced in Sukabumi Regency, and their 2020 production reached 109.966, 119.117, and 817.787 tons, respectively [18]. These plants are often found around the ball mill or “gelundung.” Residents grow these crops and either consume or sell the harvested rice in the market. In this study, paddy, cassava, and papaya were chosen as indicators to determine mercury contamination in plants and soil around ASGM areas in Sukabumi Regency.

Methods

This research was conducted in four villages in Sukabumi Regency in June 2020. Detailed information on sampling locations is shown in Table 1, and the location map is illustrated in Figure 1. In each location, samples were collected around the actively operated ball mill.

Soil sampling. Non-rhizosphere soil samples were collected from the gold processing area (ball mill). Composite soil sampling was carried out diagonally in eight composite points for each sampling area. The tools were first rinsed with aquadest and HNO_3. The soil surface of the sampling location was cleared of plants and litter. Soil sampling was carried out on plots measuring 20 cm × 20 cm with a depth of 20 cm by using a spatula. Root and plant debris were removed from the obtained samples. The soil collected from all points was placed in a basin and mixed until homogeneity was achieved. Approximately 250 g of soil was then transferred to a plastic bag and stored in a cooler with a temperature of ± 6 °C [19].

Rhizosphere soil samples were collected from the sampled plants after the soil surface was cleared of leaves or litter. The soil under the canopy around the roots was collected with a spatula and separated from the plant roots. Approximately 250 g of soil was then transferred to a plastic bag for mercury analysis [19].

Plant sampling. The leaves, stems, fruit, and roots of cassava, rice, and papaya around the ball mill were collected, cleaned, and stored in labeled plastic bags.

Total mercury analysis. Total mercury concentration in soil and plant samples was measured using USEPA 7473. The accuracy of this method was verified by recovery experiment using the spiking technique to confirm Hg loss or contamination during sample preparation and matrix interferences during measurement. In this process, Hg solution was added to the biomarker sample, and the resulting spiked samples were measured, calculated, and compared with the known value of Hg solution. The recovery value for the accuracy of biomarker analysis is at the acceptable range of 80%–100%. In addition, the precision of the mercury analyzer was checked to determine its response to measurement. The relative standard deviation value for the precision of biomarker analysis is also within the acceptable range of less than 20%.

Table 1. Sampling Location

Location	District	Village	Sub-village	Coordinates	Distance to the Ball Mill
1	Waluran	Waluran Mandiri	Sukasari	E: 106° 36′ 40.3″ S: 07° 12′ 09.9″	8.10 m
2	Waluran	Mangun Jaya	Cimanggis	E: 106° 37′ 19.8″ S: 07° 11′ 47.0″	10 m
3	Waluran	Mangun Jaya	Susukan Cagak	E: 106° 37′ 28.3″ S: 07° 11′ 45.5″	15 m
4	Lengkong	Langkap Jaya	Cibuluh	E: 106° 38′ 11.8″ S: 07° 07′ 06.5″	2 m
5	Lengkong	Langkap Jaya	Cibuluh	E: 106° 38′ 19.5″ S: 07° 07′ 09.6″	10 m
6	Lengkong	Langkap Jaya	Pamoyanan	E: 106° 38′ 25.7″ S: 07° 07′ 08.0″	20 m
7	Ciemas (Control)	Ciwaru	Cimarinjung	E: 106° 28′16.32″ S: 7° 10′ 19.90″	No ball mill in the sub-village

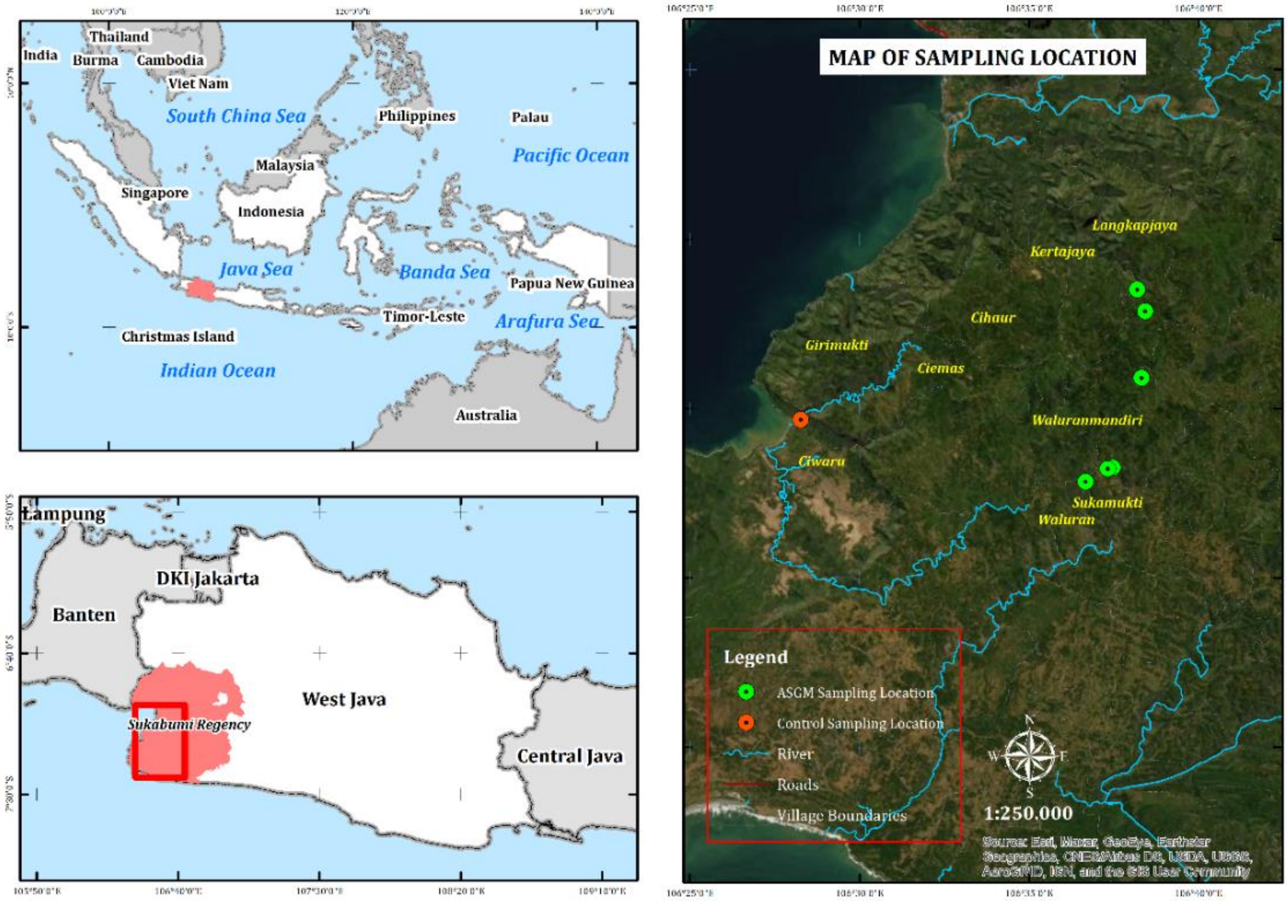

Figure 1. Map of Sampling Location in Sukabumi Regency

The samples were homogenized and weighed to ±0.05–0.2 g. For soil samples; water content was analyzed using USEPA 3540 C. Prior to analysis, the plants were cut into small sizes. Total mercury content was measured using NIC Mercury Analyzer MA-3000. The precision detection limit was 0.01 ng, and the measuring range was 0–10,000 ng. The analysis was carried out with two replications and a spike matrix, and mercury concentration was calculated using Eq.(1) and Eq.(2) for soil and Eq.(3) for the plants:

$$Wc = \frac{A - B}{A} x100\% \qquad (1)$$

where
Wc = water content (%),
A = weight of sample before heating (g), and
B = weight of sample after heating (g).

$$C_x = \frac{C}{B},$$

where
Cx = concentration of Hg (µg),
C = concentration of Hg from the instrument (µg), and
B = dry weight (g);
dry weight = wet weight – (wet weight x Wc). (2)

For mercury concentration in plants:

$$C_x = \frac{C}{B}, \qquad (3)$$

where
Cx = concentration of Hg (µg),
C = concentration of Hg from the instrument (µg), and
B = weight sample (g).

Statistical analysis. IBM SPSS Statistics 25 was used for statistical analysis. Normality was evaluated using Kolmogorov–Smirnov test, and homogeneity was investigated using Levene's test. Data of mercury content in the soil and plants were non-normally distributed and non-homogenous and therefore were analyzed using non-parametric Kruskal–Wallis test. *P* values of <0.05 indicate statistically significant differences.

Results and Discussion

In Langkap Jaya, Mangun Jaya, and Waluran Mandiri village, the sampling points are the actively operated ball mills located next to a house or near a house, paddy field, or garden. Ciwaru village has no gold ore processing; hence, no samples of ball-mil wastewater and sludge were obtained. Analysis results for total mercury in the samples are shown in Table 2. The highest total mercury concentration of 2,589.1 ppm was detected in the ball mill wastewater from Plot 5.

The plants sampled in each location differed due to availability. At the sampling time, the rice plant containing grains was found in only one location. Therefore, only one sample of rice grain was obtained. Cassava and papaya were the most common crops in the six sampling locations. Meanwhile, only rice was found at the control location. Measurement results for total mercury concentration in plant samples are displayed in Table 3. The lowest mercury concentration was found in rice stem samples from Plot 2 (0.03 ppm), and the highest was detected in cassava roots from Plot 1 (43.27 ppm).

Table 2. Concentration of T-Hg in Soil, Ball Mill Wastewater, and Sludge Samples

Location (village, district)	Hg (ppm)	
	Rhizosphere soil	Non-rhizosphere soil
Waluran Mandiri, Waluran	3.1	48.35
Mangun Jaya, Waluran	1.6	2.97
Mangun Jaya, Waluran	88	83.70
Langkap Jaya, Lengkong	16	15.38
Langkap Jaya, Lengkong	0.49	3.24
Langkap Jaya, Lengkong	3.8	2.09
Average	13.75	25.96
Ciwaru, Ciemas (Control)	0.63	1.34

Table 3. Total Mercury Concentration in Plant Samples

No.	Location	Plant	Part	Hg (ppm)
1	Waluran	Cassava	Root	43.27
2			Stem	2.88
3			Leaf	5.67
4	Mangun Jaya	Rice	Root	1.22
5			Leaf	0.46
6			Stem	0.03
7	Mangun Jaya	Cassava	Root	18.87
8			Stem	0.33
9			Leaf	3.54
10	Langkap Jaya	Papaya	Stembark	5.12
11			Bark	7.97
12			Leaf	2.16
13			Fruit	0.10
14	Langkap Jaya	Paddy	Root	0.41
15			Leaf	0.25
16			Grain	0.09
17			Stem	0.14
18		Papaya	Root	0.29
19			Bark	2.51
20			Leaf	2.29
21	Langkap Jaya	Papaya	Root	2.21
22			Stem bark	0.22
23			Leaf	0.90
24	Ciemas (control)	Rice	Root	0.44
25			Leaf	0.074
26			Stem	0.036

Table 4. Average of Mercury Level in Cassava, Paddy, and Papaya

Species	Mercury (Hg) in plant parts (ppm)				
	Root	Stem	Leaf	Fruit	Average
Cassava	31.07	1.61	4.60		12.43
Paddy	0.68	0.03	0.79	0.08	0.40
Papaya	7.12	3.96	1.78	0.09	2.60
Average	12.96	1.87	2.39	0.09	

The Hg contents of forage plants can be categorized based on the following critical limits: high hazard (>3 ppm), low-moderate hazard (0.1–3.0 ppm), and low hazard (<0.1 ppm) [20]. The cassava and papaya samples are classified as high hazards. The total mercury concentration in the samples from the control location was lower than the average for all the samples. However, the total mercury concentration of control rice samples and rhizosphere soil samples was slightly higher than that of non-control rice plants and samples from Plot 5, respectively. According to the Regulation of the Head of BPOM of the Republic of Indonesia Number 23 of 2017 concerning the Maximum Limit of Heavy Metal Contamination in Processed Food, 0.03 ppm is the maximum limit of mercury contamination in fruits, vegetables, and cereals. Therefore, all plant samples have mercury contamination exceeding the maximum limit, making them unsuitable for consumption.

Kruskal–Wallis test results showed no significant difference in mercury concentration among the study sites as the sample source ($p > 0.05$) and between plant species and plant parts ($p > 0.05$). This finding indicates that mercury accumulates evenly in all plant parts. Other studies reported that the mercury concentration in roots is usually higher than that in other parts because of its function as a barrier that prevents mercury uptake from the soil [21–23]. As shown in Table 4, the average Hg content in cassava was higher than that in other plants, and the Hg concentration in the roots was also higher than that in other plant parts. Methylmercury accumulates in rice grains, and inorganic Hg is stored in rice roots [24]. Roots absorb the total mercury in the soil, but mercury is not distributed to other parts [25]. Meanwhile, leaf mercury concentration is related to atmospheric total gaseous mercury concentration [26].

The mercury concentration in Plot 3 is similar to the average value obtained for soil (77,897 ppm) and rice roots (10,813 ppm) in rice fields in Buru Regency, which is irrigated by water and thus is directly connected to the mercury waste disposal source as an irrigation system [27]. In areas lacking ball mills around the rice fields, mercury concentrations were also the same as those in locations close to a ball mill. The absence of a ball mill at the site does not necessarily correlate to a low mercury level in the soil. The soil naturally contains mercury. For example, the average soil mercury concentration is 2.70 ppm in the control location in Bombana area, Southeast Sulawesi, [28]. This finding is attributed to the nature of mercury, that is, it can remain in the atmosphere for 0.5–2 years and spread according to wind direction [29]. Vegetables in ex-mining areas are still contaminated with mercury even after 15 years of closing [30]. Although gold processing with mercury has been discontinued, mercury is still present in these sites. The vaporized mercury spreads even to locations without ASGM activity. In addition, soil is one of the environmental components with the largest mercury reserve [31, 32].

Given that mercury in the ecosystem cannot be degraded, remediation must be carried out. Several studies have attempted to reduce Hg concentration in paddy fields by using phytoremediation, an environmentally friendly technique [33]. Juhaeti *et al*. [34] successfully reduced Hg concentration in rice mud soil and rice from Hg-contaminated rice fields by planting giant salvinia (*Salvinia molesta),* pickerel weed *(Monochoria vaginalis),* carabao grass *(Paspalum conjugatum),* and naked stem dewflower (*Commelina nudiflora)*. Malaysian false pimpernel (*Lindernia crustacea),* trailing crabgrass (*Digitaria radicosa)*, and purple nutsedge (*Cyperus rotundus)* also show potential to reduce mercury content in contaminated soil and increase maize growth [35].

Conclusion

According to the Regulation of the Head of BPOM of the Republic of Indonesia Number 23 of 2017 concerning the Maximum Limit of Heavy Metal Contamination in Processed Food, 0.03 ppm is the maximum limit of mercury contamination in fruits, vegetables, and cereals. Therefore, all plant samples around ASGM sites in Waluran, Lengkong, and Ciemas sub-districts had mercury contamination exceeding the limit and thus are not suitable for consumption. Cassava had higher mercury content (0.33–43.27 ppm) than rice (0.03–1.22 ppm) and papaya (0.06–5.11 ppm). Mercury contamination levels in rhizosphere (0.63–88 ppm) and non-rhizosphere (1.34–83.70 ppm) soils were within a similar range. No significant difference in mercury content was observed for plants and soil obtained near actively operated ball mills versus the control location. Examining atmospheric mercury levels and contamination in water is necessary to further understand the mercury pathway in ASGM areas in Sukabumi Regency.

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