# Health Risk Assessment and Mercury (Hg) Intake from Rice Consumption in Artisanal Small-Scale Gold Mining Area, Indonesia.

（インドネシアの金鉱採掘地域における米による水銀摂取量と健康リスク評価）

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**Graduate School of Environmental and Symbiotic Sciences**
**Prefectural University of Kumamoto**
**September 2020**

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**A dissertation submitted in partial fulfillment of the requirement
for the degree of Doctor of Philosophy
Graduate School of Environmental and Symbiotic Sciences
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# ACKNOWLEDGMENTS

The concern to mercury (Hg) research has been increasing in the past decade, particularly on the effects to human health. Mercury is a worldwide toxic pollutant and many countries has signed The Minamata Convention to bear the commitment to control and eliminate the mercury pollution. Since 1950s, Minamata Disease has been declared as the world disaster which caused by mercury poisoning through consuming the Hg-contaminated fish. In Indonesia, rice as the staple food is the potential source of mercury intake to human body which caused by discharging of mercury wastewater into the environment from Artisanal Small-Scale Gold Mining (ASGM). Therefore, this doctoral work was conducted to evaluate the Hg contamination in rice and to assess their impact to public health.

I would like to thank to my supervisors, ***Prof. Koji Arizono*** and ***Prof. Yasuhiro Ishibashi***, for their guidance and valuable insight to complete my doctoral work in Graduate School of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto (PUK). I am very grateful to receive many valuable experiences and knowledge under their supervision.

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# ABSTRACT

Artisanal and Small Scale Gold Mining (ASGM) contributes as the biggest source of mercury (Hg) emission in The Southeast Asian countries. Fish and other ocean products were believed to be the main source of Hg from food consumption. However, rice paddies are the most impacted agriculture area caused by Hg emission from ASGM sewage water in the rice producer countries, including Indonesia. Consuming of this contaminated rice in long period may pose to health risk effect. Meanwhile, data on Hg evaluation in rice and their impact to public health is still limited. Furthermore, there is still no reliable data on Hg accumulation in rice and health effect in Indonesia. Therefore, this study was aimed to evaluate mercury contamination level in the rice paddy field and health effect to residents by analysis of total mercury (THg) and methylmercury (MeHg) in hair around ASGM area in Lebaksitu village, Indonesia. We collected soil, water, and rice paddy samples in three paddy field areas divided by distance to the Hg hotspot. An elevated mercury concentration was detected in soil (212 – 2465 μg/kg), water (0.008 – 0.927 μg/kg), and rice (27.38 – 219.88 μg/kg) in all paddy field sampling sites. The THg concentration tended to decrease along with the increase in distance. The THg concentration in household rice ranged from 9.1 – 115 μg/kg with an average of 32.2 μg/kg. MeHg concentration in rice constituted 14.7 – 81.8% of the THg. Rice in Lebak-1 had higher THg and MeHg concentrations than those in Lebak-2. The mean THg and MeHg concentration in hair were 3.2 mg/kg and 1.78 mg/kg, respectively. Residents in Lebak-1 had significantly higher THg and MeHg in hair than those collected from Lebak-2. The MeHg ratio to THg in hair varied widely ranged from 15.68 – 92.43%. There was a significant correlation between high intake of MeHg from rice and the accumulation of MeHg in the hair. Our study revealed that the rice paddy field in Lebaksitu Village had been contaminated by mercury distributed primarily from ASGM activities. It was concluded that rice is the potential source of MeHg exposure to humans through daily consumption in rice consumer countries.

**Keywords:** *mercury, rice consumption, gold mining, daily intake, hair, methylmercury*

# CHAPTER I

# INTRODUCTION

## 1.1 Introduction

Mercury (Hg) is a global toxic pollutant that transported through environmental media such as air, water, and soil (Kim & Zoh, 2012). In the environment, mercury is naturally released from volcanos activities, cinnabar ore, coal, and as associated minerals in non-ferrous metals(UNEP, 2013). Anthropogenic activities, however, have increased mercury levels beyond their natural levels and lead to human health risk (Streets et al., 2017). The existence of mercury in the environmental media can be accumulated in the food chain which lead to the biomagnification of the higher level species, and to a further extent can be accumulated in human who consume the contaminated food(Sundseth, Pacyna, Pacyna, Pirrone, & Thorne, n.d.)(Ha et al., 2017).

In the developing countries, mercury is utilized in artisanal small-scale gold mining (ASGM) to extract gold from the ore by amalgamation technique. This technique is considered as the best choice of gold production in ASGM practice, particularly in Africa and Southeast Asia(Gibb & Leary, 2014)(Ramirez-ortiz, Rava, & Ramirez, 2018). The reasons are low-operational cost, uncomplicated technique, short production time, and enable to handle individually(Esdaile & Chalker, 2018)(Steckling, Tobollik, et al., 2017). These mining activities are generally located in the informal business and low-income population with the limitation of knowledge. A large amount of mercury was released into the environment every day(UNEP, 2018).

ASGM is the biggest contributor of mercury emission to the ambient air and water system. It contributes for 37% of global mercury emission, estimated to release about 4100 tons every year(UNEP, 2013). Due to improper waste management system, high amount of mercury is discharged into the river, soil, and agricultural ecosystem(Qiu et al., 2008). In this case, rice paddy field becomes the most impacted agricultural site from ASGM activities.

Indonesia is one of the important ASGM practice in Southeast Asia. It is estimated that more than 1000 informal sites spread throughout Indonesia to feed more than two million people including miners and their communities currently (Bose-O’Reilly et al., 2017)(McGrew, 2016). An extremely high concentrations of mercury in environmental samples were reported around ASGM area in Indonesia.

Tomiyasu et al. reported a high mercury concentration in the paddy field soil around ASGM area in Bogor, ranging from 1.93 – 55.6 mg/kg. Meanwhile, it ranged from 0.17 – 85.2 mg/kg in the river sediment(Tomiyasu, Kono, Kodamatani, & Hidayati, 2013). It was entirely reasonable by considering that the tailings from ASGM may contain 50 – 5000 mg of mercury per kg ore (Esdaile & Chalker, 2018).

ASGM in Indonesia is usually located inside the forest at highland area around rice paddy field. This condition ensures mercury to be easily transported through water flow from the upstream to the downstream area (Ismawati et al., 2015). The contaminated water supply will be deposited to the paddy field soil and become the primary source of mercury contamination in rice. The amalgam process is usually done close to water ways such as river and rice field drainage to utilize lots amount of water for crushing and amalgamation(L. Zhao et al., 2016). The water source which used to supply the rice paddy field has been contaminated with mercury waste during the growing season.

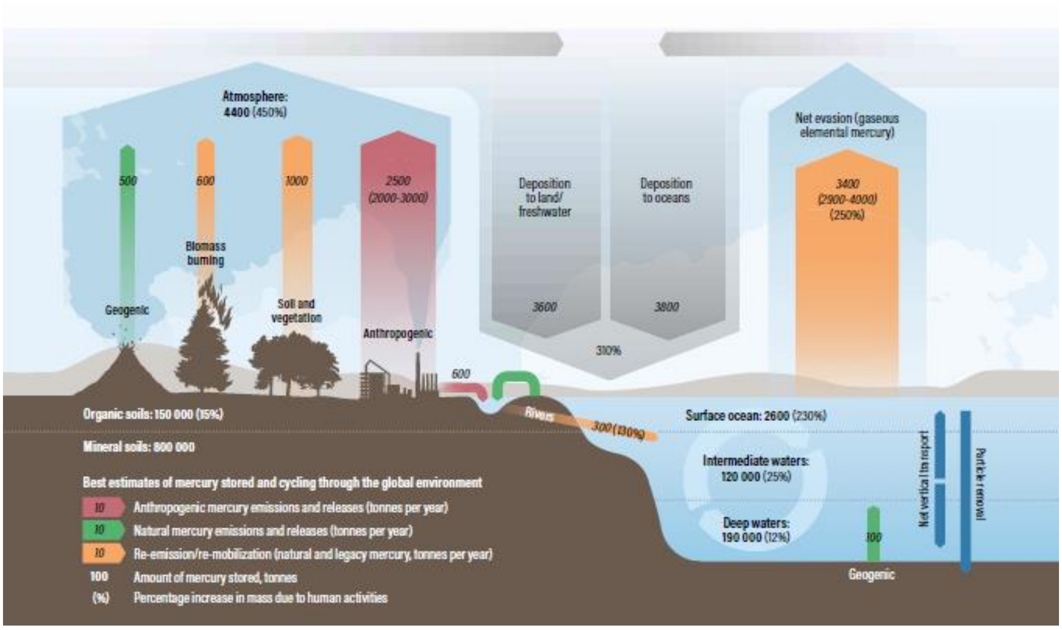

Figure 1. The emission of mercury to environment from natural and anthropogenic sources (UNEP, 2018).

Fish and other aquatic food products are suggested to be the major source of mercury intake to human body, particularly in organic form. The Minamata Disease is an example of mercury accumulation in the human body associated with the high consumption of mercury-contaminated fish which occurred in the 1950s in Japan. However, in South Asian countries including Indonesia, rice is another potential source of mercury from the food stuff. Indonesia is the third largest rice producer countries in the world after China and India with average production by 45 Mt in 2015 - 2018(FAO & OECD, 2018). It is also projected to increase at 1.28% to 53 Mt in 2027(FAO & OECD, 2018). National rice production in Indonesia is mainly used for domestic consumption with the consumption rate per capita around 500 g per day.

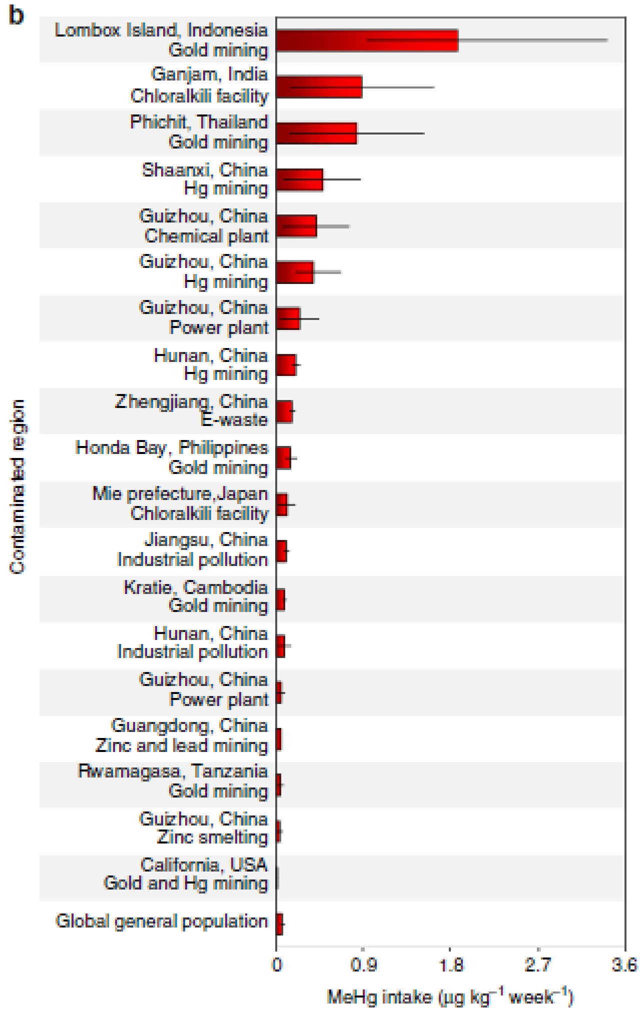


Figure 2. Human methylmercury intake through rice consumption in contaminated area around the world (Liu et al., 2019).

Recent studies suggested that rice plant is the potential source of methylmercury (MeHg) exposure from the foodstuff. It was reported that MeHg in rice contained more than 100 µg/kg in the edible portion, which is 10-100 fold higher than the other locally grown crop plants (Qiu et al., 2008). Typical paddy plantation which submerged in the flooded water creates an appropriate condition for Hg methylation. This condition ensures the methylation process by the presence of sulfur reduction bacteria (SRB) (Zhu et al., 2018). The process of Hg accumulation in the rice plant was initiated by the absorption of Hg compounds from paddy field soil by the roots, but the Hg will be distributed mainly to the rice grain during the ripening period (Meng et al., 2011). A study in Cisitu, West Java, showed that total mercury (THg) in rice reached up to 1100 µg/kg around ASGM area[15)]. Elevated mercury concentrations were found in rice samples grown around the artisanal small-scale gold mining (ASGM) in several areas in Indonesia (Bose-O'Reilly et al., 2017). Long-term consumption of this contaminated rice may pose to high risk of health impacts to local residents. Acute and chronic exposures to mercury both inorganic and organic compounds can lead to significant health impacts including neurological effects (particularly methylmercury), renal dysfunction, and cardiovascular effects (Johansson, Castoldi, Onishchenko, & Manzo, 2007)(World Health Organization, 2016).

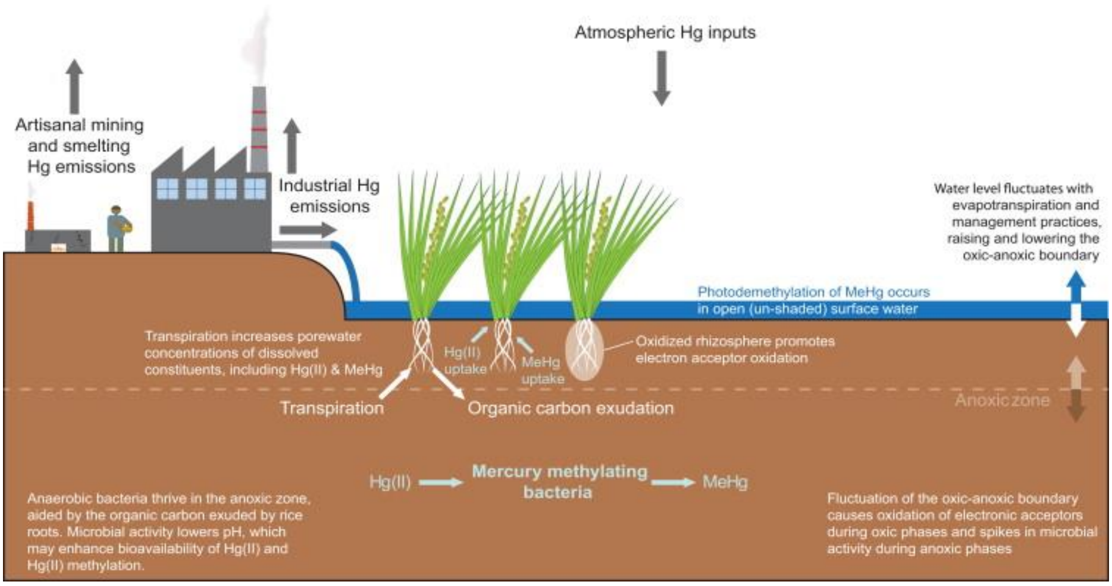

Figure 3. The process of methylmercury accumulation in rice grain (Rothenberg et al.,

2013)

Referring to the Food and Agricultural Organization (FAO) data in 2015, Indonesia is the third largest consumer of rice in the world after China and India. Indonesia consumes about 45.7 million tons of rice every year which most of them are produced domestically. The annual rice consumption per capita in Indonesia is about 163 kg/year; meanwhile, China and India consume 76.4 kg/year and 73.4 kg/year respectively (OECD & FAO, 2015). The existence of mercury pollutants in various parts of Indonesia can be a serious concern. Consumption of Hg-contaminated rice may lead to potential health effect to humans (Hang et al., 2018). However, in Indonesia, the study on evaluation of Hg-contaminated rice and their impact on human health risk is still limited. Up to date, there are no reliable data to show the assessment of mercury exposure from rice consumption and human health risk in Indonesia.

Lebaksitu Village, Banten Province, is a fast growing ASGM in Indonesia. has used mercury since it was explored in 1994. It plays a vital socio-economic role in the community. Increasing of the world gold trading trends also increase demands of mercury which utilized by ASGM miners for about more than 20 years in Lebaksitu Village. This area is drained by small drainage flowed from the Salak Mountains along the paddy fields. The gold miners utilize this water drainage for gold amalgamation which places their production plants close to the drainage. The wastewater is discharged to the drainage and distributed along the paddy field areas. Meanwhile, residents in this area have a high level of rice consumption per capita supplied from their plantation. However, the distribution of mercury contamination in environmental media in Lebaksitu Village is still not available, particularly in the rice plant.

The aim of this study was to evaluate the distribution of mercury contamination in the rice paddy field around ASGM area and the human health risk through assessment of rice consumption intake. We evaluated the accumulation of total Hg and MeHg in the paddy field ecosystem including water supply, paddy field soil, and rice paddies.

The concentration of THg was compared to permissible standard both national and international. We also assessed the range of contamination by dividing the sampling site based on distance to the Hg hotspot area. The daily consumption pattern data were collected using standard questionnaire and analyze the relationship with THg and MeHg accumulation in the hair samples.

# CHAPTER II

## The Evaluation of Mercury Contamination in Rice Paddy Field Around Artisanal Small-Scale Gold Mining Area

## 2.1 Introduction

Mercury has become a global concern due to its toxicity and the ability to bioaccumulate in the food chain. In many developing countries, mercury is utilized in artisanal small-scale gold mining (ASGM) to extract gold from the ore by amalgamation. It is still considered as the best choice of gold production in ASGM practice, particularly in Africa and Southeast Asia (Gibb & Leary, 2014)(Ramirez-ortiz et al., 2018). The reasons are low-operational cost, uncomplicated technique, short production time, and enable to handle individually(Esdaile & Chalker, 2018)(Steckling et al., 2017). These mining activities are generally located in the informal business and low-income population with the limitation of knowledge. Because of improper management, a large amount of mercury was released into the environment. It is estimated that ASGM activities release almost 1400 tons of mercury each year and become the major contributor to the amount 37% of the world mercury contamination annually (UNEP, 2013)(Sundseth et al., 2017).

Indonesia has become one of the important ASGM practice in Southeast Asia. About more than 850 ASGM activities spread throughout Indonesia engaging more than 200 thousand workers currently (BaliFokus, 2013)(Bose-O'Reilly et al., 2016). Several studies reported an extremely high concentration of mercury in environmental samples around ASGM in Indonesia. Tomiyasu et al (2013) reported a high mercury concentration in the paddy field soil around ASGM area in Bogor, ranging from 1.93 – 55.6 mg/kg. Meanwhile, it ranged from 0.17 – 85.2 mg/kg in the river sediment. It was entirely reasonable by considering that the tailings from ASGM may contain 50 – 5000 mg of mercury per kg ore (Esdaile & Chalker, 2018).

ASGM in Indonesia usually takes place at the upland area around rice paddy field. It makes mercury easily transported through water flow from the upland to the lowland area (Ismawati et al., 2015). The contaminated water will be deposited to the paddy field soil and become the primary source of mercury contamination in rice. Recent studies suggested that rice plant is the potential source of methylmercury (MeHg) exposure from the foodstuff. It was reported that MeHg in rice contained levels >100 μg/kg in the edible portion, which is 10-100 fold higher than the other locally grown crop plants (Qiu et al., 2008). Typical paddy plantation which submerged during the growing season has a high potential for Hg methylation. This condition ensures the methylation process by the presence of sulfur reduction bacteria (SRB) (Zhu et al., 2018)(Yang et al., 2018). The process of Hg accumulation in the rice plant was initiated by the absorption of Hg compounds from paddy field soil by the roots, but the Hg will be distributed mainly to the rice grain during the ripening period (Meng et al., 2014). A study in Cisitu, West Java, showed that total mercury (THg) in rice reached up to 1100 μg/kg around ASGM area (Bose-O'Reilly et al., 2016).

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ASGM in Lebaksitu Village, Banten Province, Indonesia, has used mercury since it was explored in 1994. It plays a vital socio-economic role in the community. Increasing of the world gold trading trends also increase demands of mercury which utilized by ASGM miners for about more than 20 years in Lebaksitu Village. This area is drained by small drainage flowed from the Salak Mountains along the paddy fields. The gold miners utilize this water drainage for gold amalgamation which places their production plants close to the drainage. The wastewater is discharged to the drainage and distributed along the paddy field areas. Meanwhile, residents in this area have a high level of rice consumption per capita supplied from their plantation. However, the distribution of mercury contamination in environmental media in Lebaksitu Village is still not available, particularly in the rice plant.

We evaluated the distribution of mercury contamination in the rice paddy field around ASGM area. We evaluated the accumulation of total Hg and MeHg in the paddy field ecosystem including water supply, paddy field soil, and rice paddies. The concentration of THg was compared to permissible standard both national and international. We also assessed the range of contamination by dividing the sampling site based on distance to the Hg hotspot area.

## 2.2 Materials and Methods

### 2.2.1 *Study Area*

This study was conducted at 24 years exploration of artisanal small scale gold mining (ASGM) area in Lebaksitu Village, Banten Province, Indonesia (Figure 4). Lebaksitu Village is one of the active mining in Java where engage for hundreds of miners. The mining site is located in an upland area at 950 meters above sea level with a  population density of 358 people/$km^2$. Currently, gold mining is one of the promising enterprises to residents in Lebaksitu, even though there are still many of them who work as a farmer. Based on our preliminary survey, about 90% of male farmer also work as a gold miner including ore mining, amalgam processing, and amalgam burning. Three paddy fields sites were chosen in this study: Paddy field Site (PS) 1 (0 – 500 meters from mining site; 834 meters above sea level (asl)), PS2 (500 – 1000 meters from mining site; 773 meters asl), and PS3 (1000 – 1500 meters from mining site; 648 meters asl). PS1 is located at Lebaktenjo Block where the Hg amalgamation plant takes place or also known as ball mills plant. Therefore, PS1 is considered to be a high-risk area for mercury exposure.

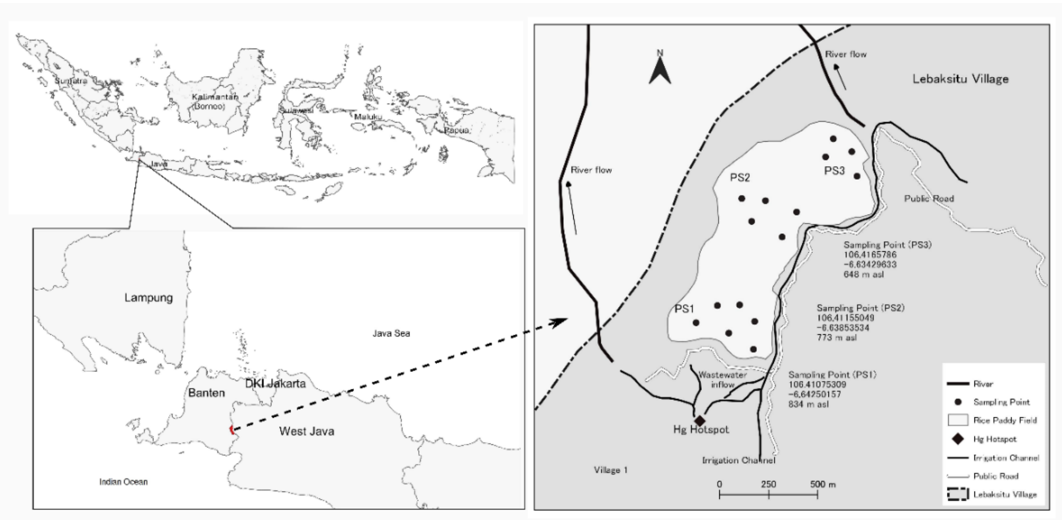



**Figure 4.** Map showing the study area which include 3 paddy field sampling sites (PS). PS 1 (0 – 500 meters from the hotspot); PS 2 (500 – 1000 meters from the hotspot); and PS 3 (> 1000 meters from the hotspot).

### 2.2.3 *Environmental Samples*

Soil, water, and rice plant samples were collected from all paddy field sites. Soil and rice plants were collected during harvesting season (June – July 2018) in order to collect soil and rice plant in the same time, meanwhile, water samples were collected during the growing season (May 2018). Water samples were taken directly into Teflon bottles which capped tightly then place into zipper bags. They were stored in the refrigerator at 4ºC. Paddy field soil was taken about 500 g started from surface to 10 cm depth by plastic scoop then placed into a zipper bag. Soil samples were stored in the refrigerator for -18ºC before freeze-drying. Rice paddies were taken from the root during harvesting then placed into zipper bags and stored in the 4ºC refrigerator. All samples were conducted by clean handling to prevent cross-contamination. Samples were also kept in a cool box during distribution from the site to the laboratory.

### *2.2.4 The procedure of determination of total mercury*

The total mercury (THg) concentration in soil and rice plant were determined using the method of U.S Environmental Protection Agency (U.S EPA) number 7473 by thermal decomposition. Analysis of THg in soil and rice samples were done without any chemical pretreatment. Soil samples were freeze-dried, ground and sieved with 150 µm mesh. Roots, rocks, woods, and other foreign objects were removed from the samples. Samples were then homogenized (Gray et al ., 2015). Rice plant samples were analyzed only for rice grain (white grain). The grains were obtained by separating coat of the seed (husk) using manual hand rice husker using rubber mill to prevent broken grain. The rice grain samples were washed with distilled water then air dried in room temperature. Rice samples were ground and sieved with 150 µm mesh (Zhou et al., 2015). Every sample was weighed 50 mg then added into three sample boat respectively to obtain triplicate data.

The water samples were filtered with 150 µm net to discard any big particle in the water sample before treatment. After that, samples were done by adding 5 ml of 0.5N sulfuric acid ($H_2SO_4$) and 2.5 ml of nitric acid ($HNO_3$) to 100 ml of samples in Teflon bottle. After that, 15 ml of 5% potassium permanganate ($KMnO_4$) was added and wait for 10 minutes to ensure the purple color persists. 8 ml of 5%

potassium persulfate ($K_2S_2O_8$) was then added into the samples and heating into the water bath at 95ºC for 2 hours. The bottle lid was tightened to prevent leak during the heating process. After heating, the samples were cooled down to room temperature then 6 ml of Sodium chloride-hydroxylammonium chloride solution (NaCl-$NH_2OH.HCl$) was added to reduce excess permanganate. After that, 0.3 ml of 10% tin(II)chloride were added and sample was analyzed immediately. A total of 5 ml treated water sample was used for every single measurement. All chemical reagents were done in >98% pure analytical grade.

THg in all samples were determined using Mercury Analyzer MA-3000 (Nippon Instrument Corporation). This machine uses amalgamation technique to collect atomized Hg compound which evaporated from heated samples. The mercury collection tube is heated to liberate the mercury again as mercury gas, and the absorbance is then measured by Cold-vapor atomic absorption at a wavelength of 253.7 nm. The THg in the soil and rice were determined by thermal decomposition measurement. Meanwhile, the THg in the water was determined by the reduction vaporization method. We conducted three replicates measurements for every sample. The coefficient of variation (CV) for each triplicate was under 5%. We confirmed recovery rate was 95 – 112 % for soil and rice samples and 90 – 120% for water samples.

## 2.3 Results and Discussion

### 2.3.1 *The total mercury in paddy field soil and paddy field water samples*

The THg concentration in the paddy field soil samples is listed in Table 1. The paddy fields were irrigated by drainage system contaminated with ASGM wastewater. The sampling sites were divided by the distance to the Hg hotspot. The THg concentrations at all sites ranged from 212 – 2465 µg/kg. The highest THg concentration of paddy field soils was observed at PS1 (2257±151.6 µg/kg), ranging from 2115 – 2465 µg/kg. This concentration was significantly higher than the other sites at PS2 and PS3 located over 500 meters from the Hg hotspot.

A significant negative correlation was found between THg concentration in the paddy field soil and distance to the Hg hotspot ($r$= -0.803) (Table 3). The mean THg concentration in the paddy field soil became lower along with the increase in distance. The THg concentration of paddy field soil decreased sharply at PS2 (625.8±341.5 µg/kg) and fivefold lower at PS3 (471±157.3) compared to PS1. The THg concentrations in the soil samples were above the U.S Environmental Protection Agency (US.EPA) for ecological soil screening (100 µg/kg). These results were also higher than the permissible tolerable value of the Indonesian Ministry of Environment for agricultural soil (500 µg/kg) except for several samples at PS2 and PS3. However, the mean THg concentration at PS3 was lower than the Indonesian standard.

The THg concentrations in paddy field water samples were considerably varied. We found a wide range of THg concentration at every site as shown by standard deviation in Table 1. We confirmed the THg concentration in the paddy field water ranging from 0.009 - 0.927 µg/L (Table 1). The highest mean THg concentration was also observed at PS1 (0.301±0.42 µg/L), ranging from 0.029 – 0.927 µg/L. The mean THg concentration in the paddy field water also decreased sharply at PS2 (0.066±0.100 µg/L) for five times lower than PS1. An extremely low THg concentration in paddy field water was still detected at PS3 located over 1500 meters from the Hg hotspot (0.030±0.031 µg/L). The concentration was three times lower than PS2 and 10 times lower than PS1. Even though the THg concentration in water tended to decrease along with increase in distance, the statistical analysis showed there was no significant correlation between the THg in paddy field water and distance to the Hg hotspot ($r$= -548; $p$>0.05) (Table 3).

Table 1. Total mercury concentration in rice field samples (Soil, Water, and Rice) by distance from the Hg hotspot. Paddy field site (PS) 1 (0 – 500 meters from the hotspot); PS2 (500 – 1000 meters from the hotspot); and PS3 (> 1000 meters from the hotspot).

| Sampling Location | Rice Field Soil (µg/Kg) | | | | Rice Field Water (µg/L) | | | | Rice Grain (µg/Kg) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | n | Min | Max | Mean± SD | n | Min | Max | Mean± SD | n | Min | Max | Mean± SD |
| PS1 | 6 | 2115 | 2465 | 2257± 151.6 | 4 | 0.029 | 0.927 | 0.301± 0.420 | 2 | 203.64 | 219.88 | 211.76 ± 11.4 |
| PS2 | 5 | 212 | 1121 | 625.8± 341.5 | 4 | 0.009 | 0.215 | 0.066± 0.100 | 2 | 81.39 | 100.06 | 90.73± 13.2 |
| PS3 | 4 | 320 | 691 | 471± 157.3 | 4 | 0.010 | 0.070 | 0.030± 0.031 | 3 | 27.38 | 29.37 | 28.57± 1.05 |
| Total Sample | 15 | 212 | 2465 | 1237.2 ± 891.1 | 12 | 0.009 | 0.927 | 0.142 ± 0.255 | 7 | 27.38 | 219.88 | 98.67 ± 82.41 |

The THg concentration in the paddy field soil were consistent with the other reported studies. A study in Buladu, Gorontalo, reported that the THg concentration in the paddy field soil ranged from 484 – 4244 µg/kg around ASGM area[24]. Meanwhile, several studies reported an extremely high concentration around ASGM area in Indonesia (Tomiyasu et al., 2013)(Mallongi, 2014). Tomiyasu et al. reported an elevated concentration in paddy field soil along Hg contaminated river around ASGM area in Bogor, ranged from 1.03 – 73.0 mg/kg. The paddy field soil was located at the downstream area over 12 km from the Hg hotspot. Meanwhile, soil Hg concentration in a natural ecosystem is relatively low, ranging from 20 – 60 µg/kg (Kabata-pendias & Mukherjee, 2007).

The THg concentration in the paddy field water were consistent with several reported data in Hg exposed area (Qiu et al., 2008). The water samples were collected during the rice-growing season about two months before harvesting. The highest value of Hg in the paddy field water was observed at PS1. Meanwhile, the paddy field water at PS3 showed the lowest Hg concentration. The Hg concentration in the paddy field water did not exceed the Indonesian water quality standard for agricultural water (5 µg/L), but it was much higher compared to EPA water quality standard of 0.05 µg/L.

The mean concentration of THg in the water showed similar pattern with THg in the soil. However, there was no significant correlation between THg in the water and THg in the soil ($r$= 0.266; $p$>0.05) (Table 3), implying that THg in paddy field water is not only controlled by wastewater but also by other factors such as water stream and additional sources of water supply. The paddy field also receives water supply directly from the water spring via a piping system. Inflow of piped water into the paddy field may influence the Hg concentration in the paddy field water. We confirmed that the THg was not detectable in the piped water due to a very low concentration (Table 2).

Table 2. Total mercury concentration in the soil and water samples at the hotspot and village 1 area.

| Sampling Location | n | Range | Mean concentration |
|---|---|---|---|
| Hotspot soil | 1 | - | 32100 µg/Kg |
| Village 1 soil | 2 | 0.717 - 1120 | 918 µg/Kg |
| Wastewater Inflow | 2 | 0.890 – 0.909 | 0.899 µg/L |
| Piping water | 2 | < 0.001 | < 0.001 µg/L |

Total Hg gas was reported to have a significantly higher concentration (403±399 ng/m$^3$) at ASGM area compared to regional background area in China (Meng et al., 2014). The Hg gas can be easily oxidized to inorganic Hg and precipitated to the ground and water surface (Zhao et al., 2016). However, we did not determine the atmospheric Hg in this study. Several studies have suggested that Hg concentration in paddy field soil from wet deposition can be neglected due to very low contribution (Kabata-pendias & Mukherjee, 2007). Even though, future research should be designed to evaluate the atmospheric contribution to Hg accumulation in the upland paddy field by determining water velocity factor and weather condition.

### 2.3.2 *The total mercury in rice plant*

In this study, our samples including rice plant were determined by a distance of 500 – 1500 meters from the Hg hotspot. The total Hg in the rice grain varied from 27.38 – 219.88 µg/kg (Table 1). The highest concentration was detected at PS1 (211.76 ± 11.4 µg/kg), ranging from 203.64 – 219.88 µg/kg. The THg concentration decreased for three times lower at PS2 (90.73±13.2 µg/kg), ranging from 81.39 – 100.06 µg/kg. PS3 which was located over 1500 meters from the Hg hotspot contained the lowest concentration among all sites (98.67±82.41 µg/kg). The THg concentrations in rice were similar to the concentration pattern in the soil and water samples based on distance to the Hg hotspot. Statistical analysis showed a significant correlation between THg concentration in rice and the distance to the Hg hotspot ($r$= -0.945; $p$<0.01) (Table 3).

The mean THg concentration in rice was relatively high compared to the other studies in Indonesia. Rice grains collected in Parbulu, Maluku, irrigated with Hg-contaminated water were reported to have lower mean Hg concentration (31.16 μg/kg) compared to our investigation (Hindersah et al., 2018). Ismawati et al (2015) reported a comparable value of Hg concentration in rice grain around Hg hotspot ranged from 101 μg/kg – 200 μg/kg. Comparing the Hg concentration around Hg hotspot (PS1), our result showed a higher level of Hg with range by 100 μg/kg - 219.88 μg/kg.

Our statistical analysis showed a significant correlation between THg in rice grain and THg in the corresponding soil (r = 0.893, $p<0.01$). This result was consistent with the study conducted by (Meng et al., 2010) that correlation analysis showed a positive relationship between THg in soil and the corresponding rice grain. During the growing season, mercury is first absorbed by the plant roots then transferred to stalk and leaf in the premature phase (Meng et al., 2014). However, in the ripening period, mercury including other minerals such as potassium (K) and phosphorus (P) will start to accumulate in the seed including husk (Ogawa et al., 2018). The THg in seed tends to accumulate much higher in husk than grain. An elevated THg concentration was found in husk (2969 μg/kg) for 70 times higher than grain (42.77 μg/kg) (unpublished data). In the ripening period, the rice plant begins to increase the biomass of rice grain and feed it with minerals which stored from other tissues or the paddy field soil (Zhao et al., 2016). This result suggested that soil is the primary source of THg in rice tissues.

Rice plant is typically grown under a flooded condition for about three months since the plantation period (Yoshida, 1981). This typical wetland ecosystem provides a contamination pathway of mercury from the source of ASGM to the rice plant (Yin et al., 2018). Interestingly, several studies have reported that rice plant has higher bioaccumulation of mercury including inorganic Hg and MeHg than the other plants (Ren et al., 2014). Many factors influence the accumulation of Hg in rice such as soil pH, mineral contents in the soil, and atmospheric deposition. Rice plant can absorb Hg from atmospheric deposition during growing season through rice plant leaf (Zhou et al., 2015). However, weather condition such as wind speed and rainfall will influence the deposition of Hg from the ambient air to the surface of the rice plant. Rothenberg et al., suggested that THg in rice originated partly from the soil, and distribution of Hg in the soil to rice grain depended on irrigation practices and soil Hg levels (Rothenberg et al., 2013).

Bioaccumulation of Hg in rice can be a potential threat to human health in Indonesia since it is consumed as the staple food. Indonesia is the third largest rice consumer in the world after China and India. Individual rice consumption is about 250 g/day for adult group (OECD & FAO, 2015). The U.S EPA has established the permissible daily intake of methylmercury by 0.1 μg/kg/day. However, there is no regulation standard about Hg permissible limit of the rice crop in Indonesia.

The previous study reported that individual rice consumption rate in Lebaksitu ranged from 100 - 700 g/day for adult level, meanwhile it was under 180 g/day for fish consumption (Haq et al., 2018). Based on our rough calculation, by determining THg concentration in rice, residents in Lebaksitu consumed THg from rice about 0.049 – 2.584 μg/kg/day, whereas it was reported that MeHg in rice ranged from 1.4 – 90 % of total mercury (Qiu et al., 2008). Compared to Hg intake from fish, it was reported that THg concentration in fish was 220 – 360 μg/kg. It estimated that residents in Lebaksitu consumed below 1.129 μg/kg/day from fish. Considering these calculations, rice was estimated to be the primary source of Hg through the food chain in Lebaksitu. MeHg intake from rice could exceed the permissible value. However, Future research should be considered to determine health risk impact by rice consumption among residents at ASGM area.

### 2.3.3 *The distribution of total mercury in the paddy field*

The distribution of THg concentration by the distance to the Hg hotspot showed similar pattern among THg in the soil, water, and rice: PS1>PS2>PS3 (Table 1). Furthermore, the negative significant correlation was found between distance to the hotspot and the corresponding soil and rice. These results indicated that the source of mercury contamination was primarily transported from the hotspot. The contaminated wastewater and tailings were transported via irrigation channel along the paddy field. The Hg concentration in the soil and wastewater inflow around Hg hotspot were also confirmed (Table 2). The total Hg concentration in the hotspot soil and wastewater inflow channel were 32100 μg/kg and 0.899 μg/L, respectively. These results corroborated our findings that the accumulation of mercury in the paddy field soil is likely to have been caused by the Hg wastewater which transported from Hg hotspot since there was no mercury sources from other stream. In the water system, Hg transport has a strong correlation with suspended particles or particulate matter in the water column (Riscassi, Hokanson, & Scanlon, 2011). The Hg compound will deposit to the paddy field soil or sediments, even though the atmospheric factors also contribute to the distribution of Hg in soil (Kim & Zoh, 2012).

The THg in the paddy field tended to decrease along with the increase in distance from the ASGM working plant, but it can be distributed for over than 100 km from the source to the downstream area (Kim & Zoh, 2012). The relevant result was also reported in the study conducted in the gold mining area, Texas, U.S. Hg concentration in soil decreased at a more distal location from the mining site. The mercury in soil was still detected around 8 km even in a relatively low concentration (Gray et al., 2015). However, THg concentration can increase at the downstream for several cases. A study of river transport of mercury in Peru showed a significant increase of THg at 300 km from the source of ASGM. Analysis of suspended particulate mercury which also followed by increased of total suspended solid suggested the existence of additional sources to the river. The additional sources such as industry, mixing of several streams, sewage discharged, and gold amalgam burning can contribute to THg concentration (Diringer et al., 2015).

ASGM activities have impacted the paddy field ecosystem in Lebaksitu particularly at PS1. The higher concentration at the upstream area was caused by the deposition of mercury mining waste discharged from ASGM plant. The decreasing pattern of Hg concentration to the more distal location indicated that ASGM was the primary source of Hg in Lebaksitu area. However, the distribution of THg at the more distal location out of Lebaksitu village such as downstream of the river should be determined for further study.

In our knowledge, this is the first study which evaluating the Hg accumulation in paddy field ecosystem around ASGM in Indonesia. Some studies have determined mercury concentration in rice plant in Sulawesi, Bogor, and Maluku, but there is still a lack of data to explain the distribution of Hg in the Indonesian paddy field. The concentration of Hg in the paddy field samples were not evaluated using individual samples, therefore, the intake of mercury in the rice tissue from soil and water could not be determined. Thus, further research is needed to evaluate the relationship of mercury intake in the rice plant from contaminated soil and water in the paddy field ecosystem. However, our results on the distribution pattern of Hg concentration in the soil, water, and rice suggested that ASGM in Lebaksitu played a significant contribution to the accumulation of mercury in the paddy field.

### 2.4 Conclusion

Our study revealed that the paddy field in Lebaksitu contained a high level of mercury from ASGM activity. In this study, the highest concentration was found at the closest paddy field to ASGM activities for soil, water, and rice samples. The concentration of THg tended to decrease along with the increase in distance from the Hg hotspot with five times lower at the distance over 1 km. The similar pattern of THg distribution among soil, water, and rice suggested that Hg wastewater was discharged to the paddy field through the irrigation water and indicated that source of Hg in Lebak was transported primarily from of the ASGM. Mercury concentration in the paddy field soil exceeded U.S Environmental Protection Agency (US.EPA) for ecological soil screening and Indonesian standard for agricultural soil. The accumulation of Hg in paddy field soil was the primary source of Hg accumulation in rice plant. The Hg accumulation in the rice plant was estimated to be a potential risk to human health who consume the rice as the staple food.

# CHAPTER III

## The Dietary Intake of Mercury from Rice and Human Health Risk in Artisanal Small-Scale Gold Mining Area

## 3.1 Introduction

Mercury (Hg) has been known as the global toxic pollutant that transported through environmental media such as air, water, and soil (Kim & Zoh, 2012). In the environment, mercury is naturally released from volcanos activities, cinnabar ore, coal, and as associated minerals in non-ferrous metals (UNEP, 2013). Anthropogenic activities, however, have increased mercury levels far above their natural levels (Streets et al., 2017). The existence of mercury in the environmental media can be accumulated in the food chain, particularly in organic form (methylmercury), which lead to the biomagnification of the higher-level species, and to a further extent can be accumulated in human body who consume the contaminated food (Sundseth et al., n.d.)(Ha et al., 2017). Acute and chronic exposures to mercury compounds can lead to significant health impacts including nervous systems damage, renal dysfunction, and cardiovascular effects (Johansson et al, 2007)(WHO, 2016).

Many studies have suggested that fish and other aquatic food products are the major sources of mercury intake to human through food consumption. The Minamata Disease which occurred in the 1950s in Japan is an example of mercury poisoning which associated with the high consumption of mercury-contaminated fish. Elevated mercury concentration in hair has a significant correlation with high rate of fish consumption (Akagi & Naganuma, 2000). However, in Southeast Asian countries including Indonesia, rice is another potential source of mercury from the foodstuff. Elevated mercury concentrations were found in rice samples grown around the artisanal small-scale gold mining (ASGM) in several areas in Indonesia (Novirsa et al., 2019)(Bose-O'Reilly et al., 2017). Long-term consumption of this contaminated rice may pose to high risk of health impacts to residents. Rice paddy is usually grown under the flooded water condition in 90 days, creating an appropriate environment for methylation process. Discharged inorganic mercury from ASGM can be easily methylated by the presence of sulfate-reducing bacteria (SRB)(Meng et al., 2011). Methylmercury (MeHg) is then accumulated in the soil and absorbed by the paddy roots to store it in the rice grain until the ripening period (Qiu et al., 2008).

The toxicokinetic mechanism of mercury toxicity from rice consumption is still not well studied compared to fish consumption. However, some reports from animal studies showing that consumption of mercury polluted rice affects the antioxidant enzymatic activities by decreasing superoxide dismutase (SOD) and glutathione peroxidase (GSH-Px) in serum and liver (Li et al., 2018). Bose-O'Reilly et al (2017) conducted a study to assess the health effects among inhabitants around the gold mining area with elevated Hg concentration in rice. It was found that more than 70% of the participants had sleep disturbances, subjective tremor, finger-to-nose tremor, ataxia of gait.

ASGM is the biggest contributor of mercury emission to the ambient air and water system. It contributes to 37% of global mercury emission, estimated to release about 4100 tons every year (UNEP, 2013). In Indonesia, the number of ASGM practices has increased since the 2000s and estimated for more than 1000 informal sites across the country to feed more than two million people including miners and their communities (Bose-O'Reilly et al., 2017)(McGrew, 2016). This traditional gold mining system utilizes mercury to extract gold from the ore by amalgamation method. Because of the improper waste management system, high amount of mercury is discharged into the river, soil, and agricultural ecosystem (Qiu et al., 2008). In this case, the rice paddy field becomes the most impacted agricultural site of ASGM activities. ASGM usually takes place close to waterways such as river and rice paddy field drainage to utilize lots amount of water for crushing and amalgamation (L. Zhao et al., 2016). The water source used to supply the rice paddy field has been contaminated with mercury waste during the growing season (Novirsa et al., 2019).

Indonesia is the third-largest rice producer countries in the world after China and India with average production by 45 Mt in 2015 – 2018 (FAO & OECD, 2018). It is also projected to increase at 1.28% to 53 Mt in 2027 (FAO & OECD, 2018). The rice consumption in Indonesia is considerably high among Asian countries together with Bangladesh, Vietnam, and the Philippines (Liu et al., 2019)(Milovanovic & Smutka, 2017). National rice production in Indonesia is mainly used for domestic consumption, meanwhile, rice from other countries are also imported to fulfill the national rice demand. In Indonesia, rice is cultivated under various geographical conditions including the difference of climate and water supply. Some rice paddy fields are cultivated around the Hg contaminated area where the sewage water connected to paddy field irrigation. It may become a potential health risk to residents who consume the contaminated rice as the staple food. Therefore, in this study, we aimed to evaluate the THg and MeHg accumulation in rice and its impact to human health through analysis of Hg accumulation in human hair samples. The probable daily intake is also calculated to estimate the Hg intake from rice consumption.

## 3.2 Materials and Methods

### *3.2.1 Study Area and Population*

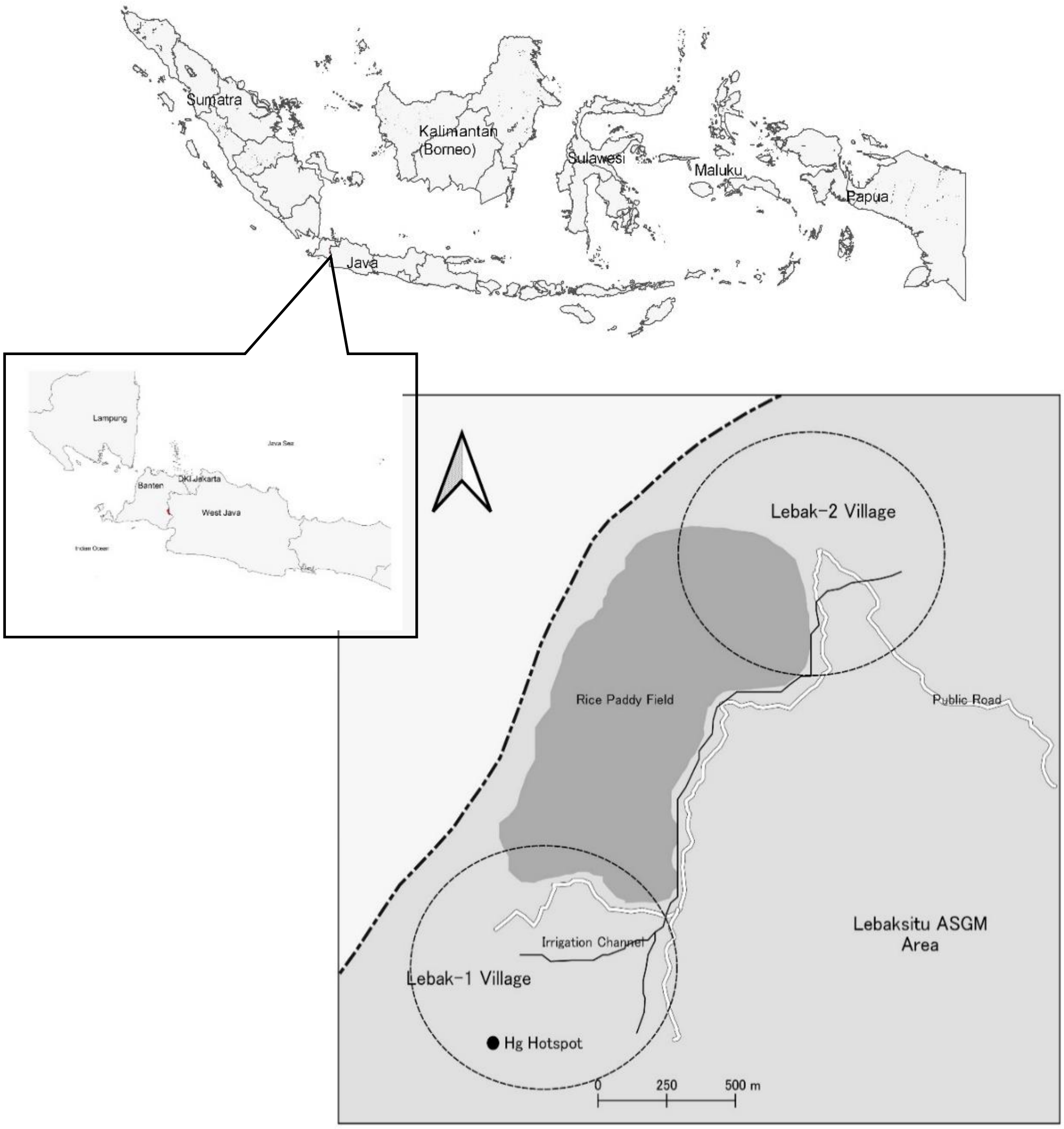
Our study was conducted in Lebaksitu ASGM area, Lebak Regency, located in the western part of Java Island, Indonesia (Figure 5). ASGM activity in Lebaksitu has been operated for more than 20 years where hundreds of people engage their life in this work. Indonesian statistical bureau (BPS) estimated more than 500 peoples working as gold miners in Lebaksitu area with a total population of around 3700 people. Based on the latest report, there are 159 units of gold processing machines spread throughout the village by 2019 (BPS, 2019). Two villages were selected in our study, Hg hotspot village (Lebak-1) and the downstream village (Lebak-2) located above 2 km from the Hg hotspot area (determined as the low-risk area). These villages span at an altitude of 950 meters above sea level surrounded by hills and forests. The preliminary data on environmental mercury in these villages have been reported in our previous study (Novirsa et al., 2019). A total of 41 adult residents who have lived for more than one year in Lebaksitu were included in our study. All participants were selected randomly.

### 3.2.2 *Rice and hair samples collection*

We collected 10 hulled rice samples randomly from resident's house, 5 rice samples from Lebak-1 and 5 rice samples from Lebak-2. All rice samples from Lebaksitu are grown locally and used for private consumption or sale within the village. Rice samples from local market in the Jakarta capital area were also collected as a comparison of non-contaminated rice. The place of origin was confirmed by an interview with the merchant. Rice samples were stored in zipper locked plastic bags and kept in 4ºC refrigerator before analysis.

Hair samples were cut from the scalp using scissors from each participant. The length of the hair used for analysis was 5 cm or less from the scalp. This length represents 3 – 5 months of exposure periods before sampling time. It is considered that generally hair grows about 1 cm per month. The hair was tied with paper tape at the scalp side then stored in zipper locked plastic bags.

Figure 5. Map of study area



### 3.3.3 *Determination of total mercury and methylmercury*

Analysis of total mercury (THg) in rice grain was done by thermal decomposition method using direct mercury analyzer MA-3000 atomic absorption spectrometry. Rice samples were washed with distilled water then air-dried at room temperature. After that, the grains were ground and sieved with 150 µm mesh and stored in 15 ml PP test tube. Every sample was weighed at 50 mg into the sample boat for analysis. For hair samples, hair was first washed with distilled water and acetone to remove any external contaminants on the hair surface. After dry, the hair was cut into fine pieces using sharp and stainless steel scissors. 50 mg hair sample was weighed for each sample boat to measure the THg concentration.

For MeHg analysis in rice and hair, around 100 mg of rice and hair samples were digested in 15 ml PP tube with 5N NaOH and 0.1% L-Cysteine solution in a block incubator at 75 °C for 1 – 2 hours to extract all of the mercury compounds (Yoshimoto et al, 2016). After cooled to room temperature, the digested sample solution was treated with distilled water, methyl isobutyl ketone (MIBK), and washed with hexane to separate any containing fats. Vigorous shaking and centrifugation were performed to separate the aliquot and solvent (Step 1). The extracted aliquot (2 ml) was then transferred to a new 15 ml PP tube for the next step. Then, the aliquot was treated with 5M HBr, 2M $CuCl_2$, and toluene. After shaking and centrifugation, the toluene layer contained MeHg was reverse-extracted using 0.2% L-Cysteine – 2% NaOAc solution (Step 2). This solution is used for MeHg analysis by thermal decomposition using mercury analyzer MA-3000 (Nippon Instrument Corporation) at a wavelength of 253.7 nm. All chemicals reagents were purchased from Wako Chemical, Japan. Samples were measured in triplicate at 200 µl in each sample boat to control consistency and homogeneity. The total mercury compound per sample boat (ng) was obtained from the measurement result and the MeHg concentration was calculated by entering the result value into the formula.

### 3.3.4 *Quality control*

Determination of THg and MeHg concentration were validated using triplicate measurements, control of blanks, spikes, and measurement of certified reference materials (CRM). To confirm THg and MeHg, we used Cod Fish Tissue (NIMJ CRM 7402-a No.250) with the certified values were 0.61±0.2 mg/kg for THg and 0.58±0.2 mg/kg for MeHg. The obtained values were 0.61±0.1 mg/kg for THg and 0.59±0.1 mg/kg for MeHg. The coefficient of variations (CV) for triplicates was under 5%. We confirmed the recovery rates were 97 - 99 % for hair samples and 95 – 98% for rice samples.

### 3.3.5 *Epidemiological data*

Epidemiological data were collected using a standard questionnaire for environmental and health risk assessment in ASGM adopted from WHO (WHO, 2016). The participants were interviewed by trained public health officer for around 15 minutes to collect personal identification, demographic data, dietary intake, lifestyle, and health status. We delivered and explained informed consent to the participants before they were interviewed. Participants who reject the informed consent were excluded from the study. We keep the security of personal information for not being leaked to public and limited only for statistical analysis purposes.

### 3.3.6 *Probable Daily Intake (PDI) of Hg from rice*

We calculated the probable daily intake (PDI) of Hg from rice consumption among adult residents in Lebaksitu ASGM area to estimate the daily intake of THg and MeHg to humans. PDI was calculated using the following equation (WHO, 1990):

$$PDI = (C \times R \times A)/BW \qquad (1)$$

PDI is calculated by multiplication of mercury concentration in rice (C= μg/kg), consumption rate (R= kg/day), and absorption rate of Hg by the human body (A) which 7% for inorganic mercury (I-Hg) and 95% for MeHg. The rice consumption rate was obtained from food frequency questionnaire by interviewing their consumption pattern for the past 1 month. Bodyweight (BW) was an average value obtained from epidemiological data. I-Hg is the THg concentration minus MeHg concentration. The THg PDI is the sum of I-Hg PDI and MeHg PDI.

3.3.7 *Ethical Approval*

The prefectural university of Kumamoto ethic committee has approved the research condition and method of this study.

3.3.8 *Statistical Analysis*

Statistical analysis was performed using statistical package software. Demographic distribution, consumption pattern THg and MeHg Concentrations were described by descriptive analysis to show frequency, mean, or standard deviation. The relationship between variables was performed using t-test analysis, cross-tab, correlation, and regression analysis. Data were checked for normal distribution before analysis, however, non-parametric analysis will be performed in case of non-normal distribution.

## 3.4 Results and Discussion

3.4.1 *Hg Concentration in Rice*

The concentration of THg and MeHg in rice are summarized in Table 3. The rice samples were collected randomly from household stock used for daily consumption both in Lebak-1 and Lebak-2. The residents usually consume the rice from their paddy field which grown locally in the village. The THg concentrations in rice were higher than World Health Organization (WHO) standard for food products other than fish (100 µg/kg) and Indonesian National Standard (30 µg/kg) in 30% rice samples, ranged from 9.1 – 115 µg/kg. The mean THg and MeHg concentration in rice from Lebak-1 were significantly higher than rice collected from Lebak-2. It indicated that the rice from Lebak-1 was not in safe level compared to rice from Lebak-2, ranged 13.8 – 115 µg/kg and 9.1 – 23.2 µg/kg, respectively. Lebak-1 is located within 500 meters of Hg hotspot area, therefore the paddy fields were seriously exposed by Hg than in other villages. These results also showed that rice grown in Lebak-1 has been contaminated significantly by Hg from ASGM emission than those grown in Lebak-2. Residents living in Lebak-1 may pose to high risk of health effects due to consuming high-level mercury in rice.

In this study, the THg in rice was much lower than some gold mining areas in China; Guizhou (187µg/kg, range: 24.8 – 548 µg/kg)(Meng et al., 2010); Wanshan (93.6 ± 114.2 µg/kg); and Danzhai (85.8 ± 43.6 µg/kg)(Meng et al., 2014). However, it was slightly higher than other studies in Cambodia (12.7 µg/kg, range: 9.9 – 16.7 µg/kg)(Cheng et al., 2013). For MeHg concentration in rice, our results showed higher mean concentration than that in Cambodia (1.54 µg/kg) and some mercury mining in China, such as Guangdong (1.2 µg/kg) and Guizhou (5.7 µg/kg), but it was far lower than in Wanshan (30.4 µg/kg) and Danzhai (20.8 µg/kg ) as summarized in the previous study (Zhao et al., 2019). Some studies reported Hg concentrations in rice around ASGM area in other parts of Indonesia, but Hg in fish was discussed as the main concern instead of rice. Bose-O'Reilly et al (2016) reported high level of Hg contamination in rice grown at ASGM area in Cisitu, ranging from 89 – 1180 µg/kg. A high range of Hg concentrations was also reported by Mallongi et al (2014) in Gorontalo ASGM area, the THg concentration in rice ranged from 113 – 1084 µg/kg. It should be noted that rice is the staple food in Indonesia with high consumption level per capita in Asia (Liu et al., 2019). Therefore, study on Hg contamination in rice and health risk assessment in Indonesia should be increased in the future.

Table 3. THg and MeHg concentrations in rice collected in Lebaksitu ASGM area

| Sampling Area | n | THg (µg/kg) | | MeHg (µg/kg) | | MeHg/THg % | |
|---|---|---|---|---|---|---|---|
| | | Average | Range | Average | Range | Average | Range |
| ***Lebaksitu*** | | | | | | | |
| Lebak1 | 5 | 48.5 | 13.8 - 115 | 14.0 | 4.9 – 20.7 | 40.8 | 14.7 – 81.8 |
| Lebak2 | 5 | 15.9 | 9.1 – 23.2 | 9.8 | 6.5 – 11.7 | 64.6 | 50.5 – 79.5 |
| Both Villages | 10 | 32.2 | 9.1 - 115 | 11.9 | 4.9 – 20.7 | 52.7 | 14.7 – 81.8 |
| ***Jakarta Capital Rice Market*** | | | | | | | |
| Place of origin : | | | | | | | |
| 1. Sumatera | 2 | 1.5 | 1.5 – 1.6 | < 0.001 | < 0.001 | NA | NA |
| 2. West Java | 4 | 1.6 | 1.4 – 1.7 | < 0.001 | < 0.001 | NA | NA |
| 3. East Java | 5 | 1.7 | 1.3 – 2.4 | < 0.001 | < 0.001 | NA | NA |
| Total | 11 | 1.6 | 1.3 – 2.4 | < 0.001 | < 0.001 | NA | NA |

In this study, the ratio of MeHg to THg concentrations in rice ranged from 14.7 – 81.8%. These results were similar to other studies reported the accumulation of MeHg in rice grown in gold or mercury mining area, ranging from 1.4 – 91% (Qiu et al., 2008)(Wang et al, 2018). Previous studies suggested that wide variation of MeHg ratios in rice was influenced by the condition of soil-water system in the paddy field ecosystem (Zhu et al, 2018)(Meng et al., 2011). Other than that, high level of I-Hg in the atmosphere can contribute to the proportion of Hg compounds in the rice grain. Paddy leafs can absorb I-Hg from atmospheric deposition, while MeHg is mainly uptake by the roots from paddy field soil (Wang et al., 2018). Interestingly, the mean MeHg ratio of THg in rice for Lebak-1 was lower than those from Lebak-2. The rice grown in Lebak-1 probably absorb both I-Hg from atmospheric deposition and MeHg from the soil in high amount because of the location in Hg hotspot area, meanwhile, rice in Lebak-2 absorbs almost MeHg from the soil. Rice is the potential source of MeHg with ratios to THg were 10 – 100 times higher than other crops (Qiu et al., 2008). It was suggested that I-Hg could not be methylated in the rice tissue, but the paddy soil which submerged in the flooded water during growing season provides a good condition for methylation proses in the soil by sulfur reduction bacteria (Meng et al., 2011).

The THg concentration in commercial rice collected from the Jakarta rice market in this study showed significantly lower than rice from Lebaksitu ASGM area. The mean THg concentration (1.6 µg/kg, range: 1.3 – 2.4 µg/kg) was far below the Indonesian National Standard. Moreover, this results also lower than other commercial rice reported in Sri Lanka (1.73 µg/kg, range: 0.21 – 6.13 µg/kg)(Xu et al., 2020), China (4.47 µg/kg, range: 1.06 – 22.7 µg/kg)(H. Zhao et al., 2019), Pakistan (4.51 µg/kg, range: 0.44 – 157 µg/kg)(Aslam et al., 2020), and Cambodia (8.14 µg/kg, range: 6.16 – 11.7 µg/kg)(Cheng et al., 2013). It means that commercial rice in Indonesia is still in a safe level based on Indonesian National Standard for THg in cereals. This also suggested that the existence of ASGM in Indonesia has contributed to high Hg contamination in rice.

### 3.4.2 *Consumption pattern and mercury intake*

The results of rice consumption pattern and PDI were summarized in Table 4. Calculation variables were obtained by interview using a standard food frequency questionnaire (FFQ) by confirming the last one month of consumption history. Their consumption history including amount of rice per mealtime, meal frequency, and the origin cultivation place of the rice. Then, THg and MeHg PDI were calculated based on the questionnaire data including consumption rate and body weight. For Hg concentration in rice, we used the mean THg and MeHg value from each village.

Based on FFQ questionnaire results, the rice consumption rate in Lebaksitu ranged from 260 – 900 g/day with an average of 520 g/day. Rice was consumed more than 500 g/day by 49% of residents in Lebaksitu. The residents consumed the rice 2 times/day by 58.5% and 3 times/day remaining with the consumption amount per time ranged from 150 – 300 g/ mealtime. Our results were slightly higher than in other communities in Indonesia, but still in the same range. The rice consumption rate reported in Ciguha, West Java, had an average 445 g/day, ranged from 200 – 900 g/day (Kusnoputranto et al, 2017). This also much higher than the study conducted by Bose-O'Reilly et al (2017) in Cisitu, the average rice consumption rate was 280 g/day. Meanwhile, the national rice consumption per capita in Indonesia was 150 kg/year or around 420 g/day, particularly in rural communities (Bentley & Soebandrio, 2017).

Table 4. Rice consumption pattern and daily intake of THg and MeHg in rice.

| Variables | Both Villages (n = 41) | Lebak1 (n = 21) | Lebak2 (n= 20) |
|---|---|---|---|
| *Rice Consumption Pattern* | | | |
| Rice Source: | | | |
| - Local (%) | 97 | 100 | 95 |
| - other (%) | 3 | 0 | 5 |
| Consumption rate (g/day) | 520 (260 - 900) | 629 (360 - 900) | 407 (260 - 600) |
| 1 time/day (%) | 0 | 0 | 0 |
| 2 times/day (%) | 58.5 | 28.6 | 90 |
| 3 times/day (%) | 41.5 | 71.4 | 10 |
| > 3 times/day (%) | 0 | 0 | 0 |
| *Body Weight (kg)* | | | |
| Mean (range) | 52.7 (30 - 90) | 53.6 (30 - 90) | 51.9 (40 - 75) |
| *THg PDI (µg/kg/day)* | | | |
| Mean (range) | 0.116 (0.040 - 0.240) | 0.164 (0.090 - 0.240) | 0.066 (0.040 - 0.100) |
| *I-Hg PDI (µg/kg/day)* | | | |
| Mean (range) | 0.014 (0.002 - 0.036) | 0.025 (0.014 - 0.036) | 0.003 (0.002 - 0.004) |
| *MeHg PDI (µg/kg/day)* | | | |
| Mean (range) | 0.102 (0.040 - 0.199) | 0.139 (0.079 - 0.199) | 0.063 (0.040 - 0.093) |

The rice consumption rate in Lebak-1 was significantly higher than in Lebak-2 at 629 g/day and 407 g/day, respectively. It showed that Lebak-2 had a similar consumption rate with general communities in Indonesia. Refer to Lebaksitu Administration Office, Lebak-1 was categorized as remote area and their economic status was lower than in Lebak-2. We assumed that residents in Lebak-2 consume more variety of food so that they consume lower amount of rice than in Lebak-1. As mentioned before, residents in Lebaksitu consume rice which harvested from their agricultural land and sold to fulfill the needs in the village. A total of 97% of the participants in this study answered that the rice was obtained by their land or bought from other house in the village. It can be concluded that residents in Lebaksitu were mainly exposed to Hg-contaminated rice from their agricultural land.

The overall average THg PDI for adult residents in Lebaksitu ASGM area was 0.116 μg/kg/day, ranged from 0.04 – 0.240 μg/kg/day. This average value was below the limit of provisional tolerable weekly intake (PTWI) of THg set by FAO/WHO (2010) for foods other than fish of 4 μg/kg/week or equal to 0.57 μg/kg/day. The THg PDI in Lebak-1 was significantly higher than in Lebak-2, the average THg PDI were 0.164 μg/kg/day (range: 0.09 – 0.24 μg/kg/day) and 0.066 μg/kg/day (range: 0.04 – 0.100 μg/kg/day), respectively. The THg PDI in Lebak-1 was 2 times higher than PDI in Lebak-2. The average PDI of MeHg was 0.102 μg/kg/day, ranged from 0.04 – 0.199 μg/kg/day. This value was lower than the limit of PTWI for MeHg at 1.6 μg/kg/week or equal to 0.23 μg/kg/day, but it reached over the reference dose (RfD) for MeHg recommended by the US.EPA of 0.1 μg/kg/day. There were 84% of residents in Lebak-1 exceeded the US.EPA RfD for MeHg, however, there were no residents in Lebak-2 who exceeded the RfD value. It can be concluded that rice in this study was not in a safe level to meet the consumption rate in Lebaksitu ASGM area, particularly for those who live in Lebak-1. High MeHg intake was probably caused by high consumption rate of rice as the staple food and higher MeHg concentration in the rice. Long-term consumption may affect adverse effects to human health.

Several studies evaluated the Hg intake in mercury polluted areas. Our results showed similar pattern with the study conducted by Feng et al (2008), suggested that rice consumption was the main source of MeHg exposure to residents living in mercury mining areas. The MeHg intake ranged from 0.01 – 0.21 µg/kg/day which constituted about 95% of the total MeHg exposure to residents in Wanshan Hg mining area, China. Other than that, the average daily rice consumption in China was reported to be lower than Indonesian daily consumption (Zhao et al., 2019)(Liu et al., 2019).

We estimated the THg intake from rice in Indonesian general communities was below 0.001 µg/kg/day, which derived from the THg concentration in the Jakarta rice market. To date, there were no reliable studies evaluated the Hg intake in Indonesian general population on rice consumption. This estimation value was far lower than general population reported in Sri Lanka (0.015 µg/kg/day)(Xu et al., 2020), China (0.0056 µg/kg/day (MeHg), range: 0.0012 – 0.0134 µg/kg/day)(Zhao et al., 2019), and Kratie, Cambodia (12.6 µg/kg/day)(Cheng et al., 2013). Even though Indonesian rice consumption rate was considerably high in Asia, the THg concentrations in rice were very low for the general population. However, Indonesia was reported to be the highest MeHg intake from rice globally based on contaminated regions (Liu et al., 2019). It was estimated that MeHg intake from rice in Lombok ASGM area, Indonesia, could reach 1.9 µg/kg/week (equal to 0.27 µg/kg/day), ranged from 0.94 – 3.4 µg/kg/week, followed by Ganjam, India (chloralkali) and Phicit, Thailand (gold mining) which below than 1 µg/kg/day. Krisnayanti et al (2012) reported the mean MeHg concentration in rice in Sekotong ASGM area, Lombok, was 57.7 µg/kg, ranged from 10.6 – 115 µg/kg which was five times higher than our present study. It can be concluded that rice is the potential source of Hg intake to residents living in ASGM area where the rice is cultivated under a high Hg contaminated environment.

In the general communities, fish and other aquatic food products are the main sources of MeHg accumulation in the human body. It contributed to around 80 – 90% of the total MeHg diet (Akagi & Naganuma., 2000). Generally, mercury compounds in fish are in MeHg form which constituted around 90% of THg (Akagi et al., 1995). Based on our rough calculation, MeHg intake from fish contributed only 21% of the total MeHg diet considering that residents in Lebaksitu consumed fish 2 – 3 times per week with an average of 10 g/day. MeHg from rice contributed around 70% of the total MeHg diet, while the other 10% was obtained from vegetables and other food products.

### 3.4.3 *THg and MeHg concentration in hair*

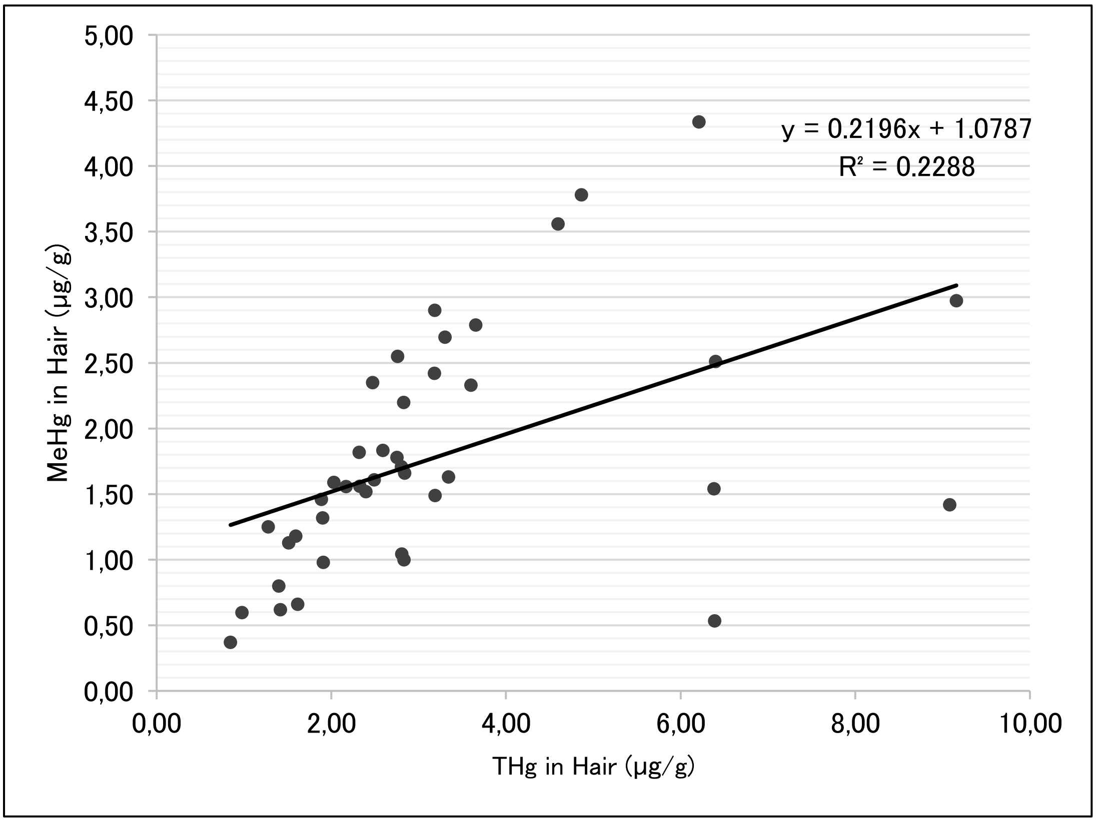
The concentration of THg and MeHg in hair were summarized in Table 5. Hair samples were collected from the adult participants who agreed to join the study after confirming the informed consent. The mean THg in hair for both villages was 3.2 µg/g, ranged from 0.847 – 9.15 µg/g. The mean THg in hair collected from Lebak-1 was significantly higher than collected in Lebak-2, 3.51 µg/g and 2.87 µg/g, respectively. For MeHg in hair, the mean concentration was 1.78 µg/g for both villages, ranged from 0.37 – 4.33 µg/g. Similar to THg in hair, the mean MeHg in hair samples of Lebak-1 residents was significantly higher than hair samples collected from Lebak-2. It indicated that residents in Lebak-1 were exposed to Hg greater than residents living in Lebak-2. The accumulation of Hg in the hair showed that ASGM has contributed to the health risk of the residents living in Lebaksitu.

Table 5. THg and MeHg concentrations in hair collected from Lebaksitu ASGM area

| Sampling Area | n | THg (µg/g) | | | | MeHg (µg/g) | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Mean | Min | Max | SD | Mean | Min | Max | SD |
| Lebak1 | 21 | 3.51 | 1.27 | 9.15 | 1.88 | 2.12 | 0.62 | 4.33 | 9.76 |
| Lebak2 | 20 | 2.87 | 0.84 | 9.08 | 2.05 | 1.42 | 0.37 | 2.90 | 6.80 |
| Both villages | 41 | 3.20 | 0.847 | 9.15 | 1.97 | 1.78 | 0.37 | 4.33 | 0.90 |

The mean THg and MeHg in this study were comparable with other studies evaluating rice consumption in Hg contaminated areas in China (Feng et al., 2008) and Colombia (Salazar-Camacho et al., 2017). The accumulation of MeHg in hair is mainly from food consumption such as fish and rice, while I-Hg is generally obtained from high level of Hg contamination in the air (Y. Li, 2013)(Sheehan et al., 2014). The mean MeHg in hair related to fish consumption in general communities could be higher than the communities living in the contaminated area with low fish consumption (Aslam et al., 2020)(Salazar-Camacho et al., 2017). It should be noted that many factors may contribute to the MeHg accumulation status in hair such as age, individual susceptibility, gender, smoking habit, and lifestyle (Marcinek-jacel et al., 2017).

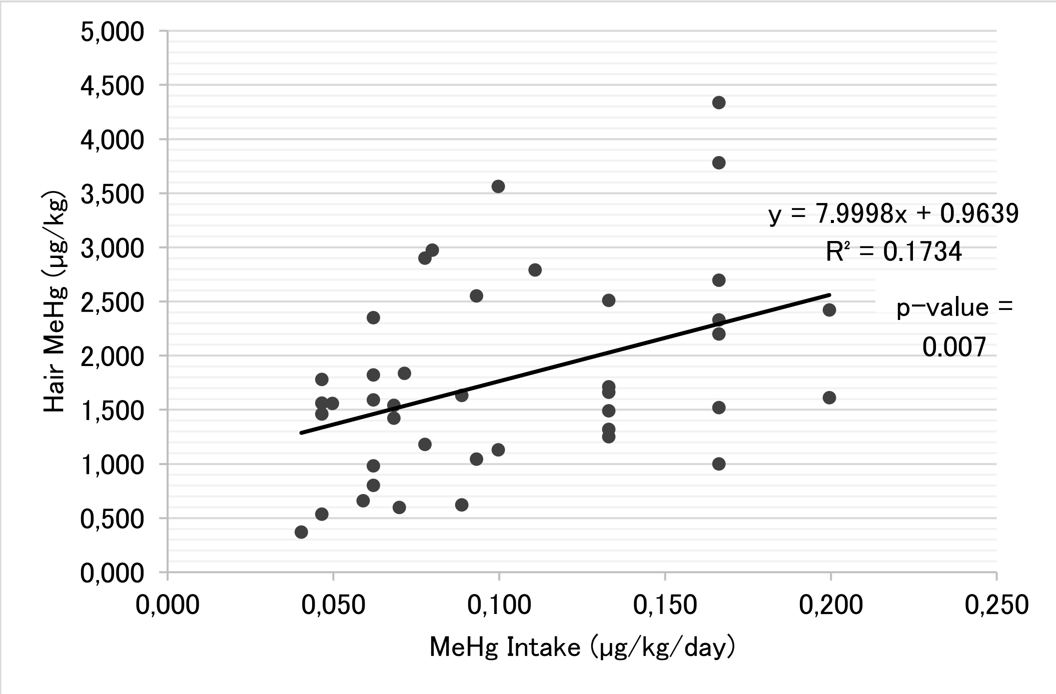
Figure 6. The correlation between THg and MeHg concentration in hair




The proportion of MeHg of THg in hair varied widely with an average of 60.42%, ranged from 15.68 – 92.43%. Interestingly, there was no significant difference between hair MeHg ratio to THg in Lebak-1 and in Lebak-2. Hair MeHg ratio to THg in Lebak-1 and Lebak-2 were 60.40% and 60.45%, respectively. Meanwhile, in general communities, MeHg ratio constituted about 70 – 90% of the total mercury which mainly obtained from seafood or rice consumption (Srogi, 2007)(Hoang et al., 2017). Wide range of MeHg ratio to THg in hair was possibly caused by the existence of high I-Hg concentration in the air. Inhaled I-Hg or gaseous mercury ($Hg^0$) is absorbed by alveolus into the blood system and passes the blood-brain barrier for approximately 80% (Ha et al., 2017)(WHO, 1976). Akagi et al (1995) reported the average proportion of MeHg to THg ranged from 35.6 – 43.0% in the gold miners and extremely lower in the gold shop workers at around 13%. The mean hair THg in gold shop workers was higher than the gold miners. Similar results were also reported by Feng et al (2008) in Wanshan Hg mining area, China, that MeHg concentration in hair constituted around 42 – 62% in all villages. It was significantly lower than the control village with no Hg exposure (82.7%). It can be concluded that residents in Lebaksitu ASGM area were also possibly exposed to high level of I-Hg from the air. However, future studies should be performed to evaluate the I-Hg existence in the air.

The correlation of THg concentrations and MeHg concentrations in hair samples were shown in Figure 6. There was a positive correlation between THg and MeHg in the hair, but with low correlation strength (p-value= 0.002, R= 0.228). It indicated that there was different pattern of I-Hg and MeHg exposure between participants. As mentioned above, some residents might be exposed to high concentration of I-Hg from the air such as gold miners or gold shop workers. Unfortunately, we did not identify the participant's occupation or their house environment condition in this study due to sensitive private reasons among the residents in Lebaksitu. Feng et al (1998) reported a correlation between hair THg and MeHg concentrations in Indonesia, China, and Japan. Hair THg and MeHg correlation in Medan, Indonesia (P>0.05; R= 0.216) was similar with our results. The lowest correlation was found in Harbin, China (P>0.05; R= 0.064). Hair MeHg concentrations in Indonesia and China were not close to the hair THg which resulted from higher exposure of I-Hg compared to the Japanese community. Hair MeHg in Tokushima, Japan, correlated closely to the hair THg (P<0.01; R= 0.932). It was true that the Japanese community was primarily exposed to Hg from seafood consumption.

Figure 7. The correlation between MeHg Intake (µg/kg/day) from rice and MeHg concentration (µg/g) in hair




We found a significant positive correlation of MeHg intake from rice and MeHg concentrations in hair as shown in Figure 7. These results confirmed our estimation that residents in Lebaksitu were mainly exposed to MeHg from rice consumption. High rice consumption rate as the staple food resulted in the high accumulation of MeHg in hair even though the MeHg concentrations in rice were relatively low. Rice could be a significant source of MeHg exposure from food consumption in Southeast Asia, particularly in Indonesia (Liu et al., 2019).

## 3.5 Conclusions

In this study, we found that rice is the potential source of MeHg accumulation in the human body via daily consumption, particularly in the country where rice is consumed as the staple food. High consumption rate of Hg contaminated rice may pose to high accumulation of MeHg in the human body even though the MeHg concentration in rice is relatively low. Lebak-1 which located in the Hg hotspot area received higher risk of Hg exposure than in Lebak-2 which considered as low level of Hg exposure. The mean concentration of THg and MeHg in rice and hair samples were higher in Lebak-1 compared to those in Lebak-2. It was suggested that discharged mercury from ASGM contributed to the accumulation of Hg in the rice grain cultivated around the ASGM area.

# CHAPTER IV

# CONCLUSIONS

## 4.1 Conclusions

Our study found that the rice paddy field has been contaminated with a high level of mercury from ASGM activity in Lebaksitu ASGM area. The highest Hg concentration for soil, water, and rice samples were found at the nearest area to ASGM hotspot. The concentration of THg in the soil, water, and rice grain tended to decrease along with the increase in distance from the Hg hotspot. The similar pattern of THg distribution among soil, water, and rice suggested that Hg wastewater was discharged to the paddy field through the irrigation water and indicated that source of Hg in Lebak was transported primarily from of the ASGM. Mercury concentration in the paddy field soil exceeded the U.S Environmental Protection Agency (US.EPA) for ecological soil screening and Indonesian standard for agricultural soil. The accumulation of Hg in paddy field soil was the primary source of Hg accumulation in rice plant.

We found that rice is the potential source of MeHg accumulation in the human body via daily consumption where rice is consumed as the staple food in Lebaksitu ASGM area. Lebak-1 Village which located in the Hg hotspot area received higher risk of Hg exposure than in Lebak-2 Village which considered as low level of Hg exposure. High consumption rate of Hg contaminated rice may pose to high accumulation of MeHg in the human body even though the MeHg concentration in rice is relatively low. The mean concentration of THg and MeHg in rice and hair samples were higher in Lebak-1 compared to those in Lebak-2. It was suggested that discharged mercury from ASGM contributed to the accumulation of Hg in the rice grain cultivated around the ASGM area. The Hg accumulation in the rice plant was estimated to be a potential risk to human health who consume the rice as the staple food.

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