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Original Article

The dietary intake of mercury from rice and human health risk in artisanal small-scale gold mining area, Indonesia

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ABSTRACT — Artisanal and Small Scale Gold Mining (ASGM) contributes as the biggest source of mercury (Hg) emission in Southeast Asian countries including Indonesia. Rice paddies are the most impacted agriculture area caused by Hg emission from ASGM sewage water. It may pose to health risk effect to humans due to the consumption of the rice as the staple food. This study was aimed to evaluate the Hg accumulation in rice and health effect to residents in Lebaksitu ASGM area by analysis of total mercury (THg) and methylmercury (MeHg) in hair. Two villages were selected in this study, Hg hotspot village (Lebak-1) and low Hg exposure village (Lebak-2). The THg concentration in 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. It was concluded that rice is the potential source of MeHg exposure to humans through daily consumption in rice consumer countries.

Key words: Mercury, Rice consumption, Gold mining, Daily intake, Hair, Methylmercury

INTRODUCTION

Mercury (Hg) has been known as the global toxic pollutant that transported through environmental media such as air, water, and soil (Kim and 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*., 2017; 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

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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 and 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 (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 and OECD, 2018). It is also projected to increase at 1.28% to 53 Mt in 2027 (FAO and 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 and 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.

MATERIALS AND METHODS

Study area and population

Our study was conducted in Lebaksitu ASGM area, Lebak Regency, located in the western part of Java Island, Indonesia (Fig. 1). 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 m 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.

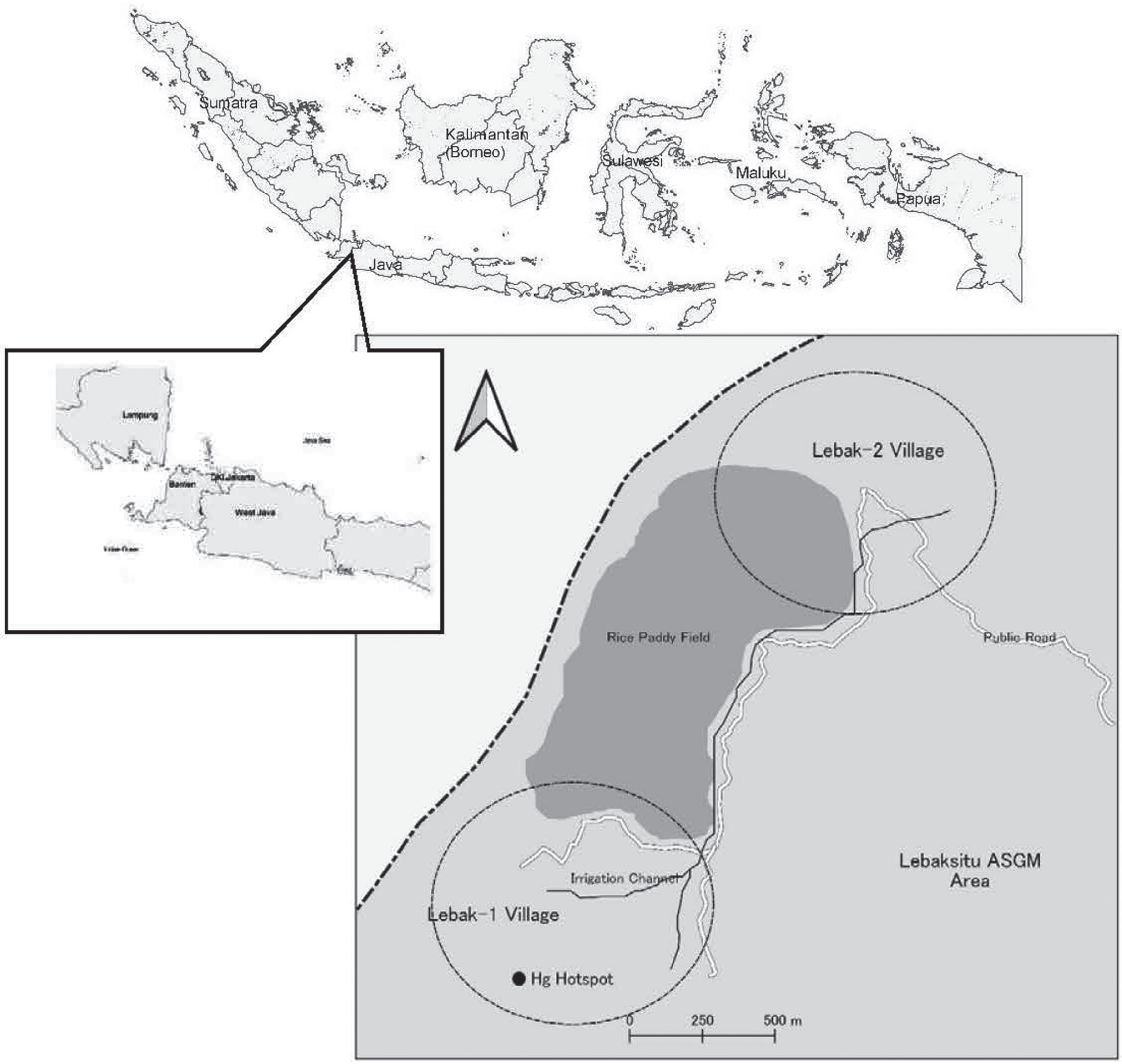

Fig. 1. Map of study area.

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 4°C 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.

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 hr 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, Tokyo, Japan) at a wavelength of 253.7 nm. All chemicals reagents were purchased from Wako Chemical, Osaka, 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.

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.

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 min 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.

Probable daily intake (PDI) of Hg from rice

We calculated the probable daily intake (PDI) of Hg from rice consumption among adult residents in Lebak-situ 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.

Ethical approval

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

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.

RESULTS AND DISCUSSION

Hg concentration in rice

The concentration of THg and MeHg in rice are summarized in Table 1. The rice samples were collected randomly from household stock used for daily consumption both in Lebak-1 and Lebak-2. The residents usually con-

Table 1. THg and MeHg concentrations collected in Lebaksitu ASGM area.

SamplingArea	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

sume 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 m 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 (187 μg/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.

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*., 2015; 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) (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.

Consumption pattern and mercury intake

The results of rice consumption pattern and PDI were summarized in Table 2. 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 and Soebandrio, 2017).

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 approached the limit of provisional tolerable weekly intake (PTWI) of

Table 2. 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)

THg set by FAO/WHO (2010) for foods other than fish of 1 μg/kg/week or equal to 0.142 μg/kg/day. There were 37% of the residents reached over the limit of PTWI in Lebaksitu. 71% of residents in Lebak-1 exceeded the PTWI, while there were no residents exceeded the limit in Lebak-2. 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 and 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.

THg and MeHg concentration in hair

The concentration of THg and MeHg in hair were summarized in Table 3. 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.

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 (Li, 2013; Sheehan *et al*., 2014). The mean MeHg in hair related to fish consumption in general communities could be

Table 3. 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

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).

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 Fig. 2. 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.

We found a significant positive correlation of MeHg intake from rice and MeHg concentrations in hair as shown in Fig. 3. 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).

In 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.

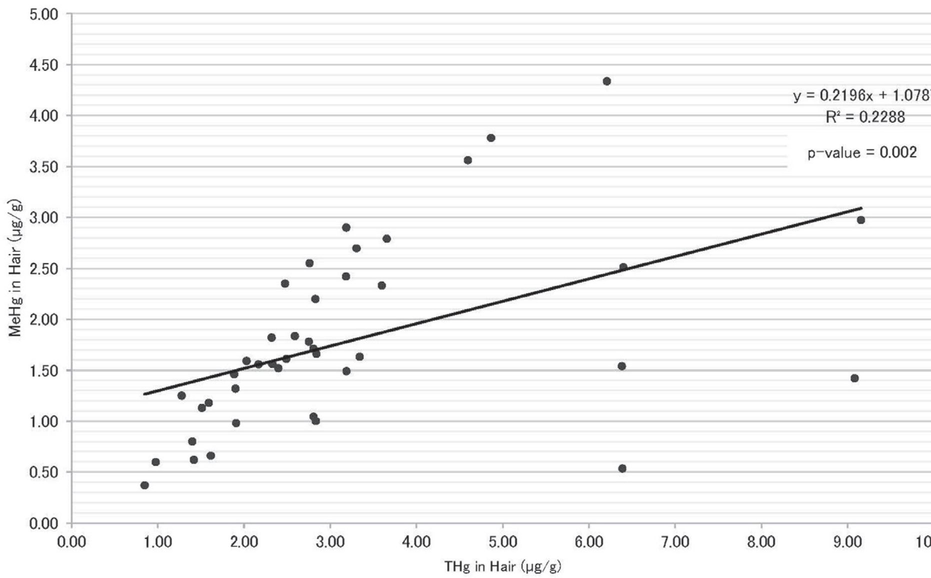

Fig. 2. The correlation between THg and MeHg concentration in hair.

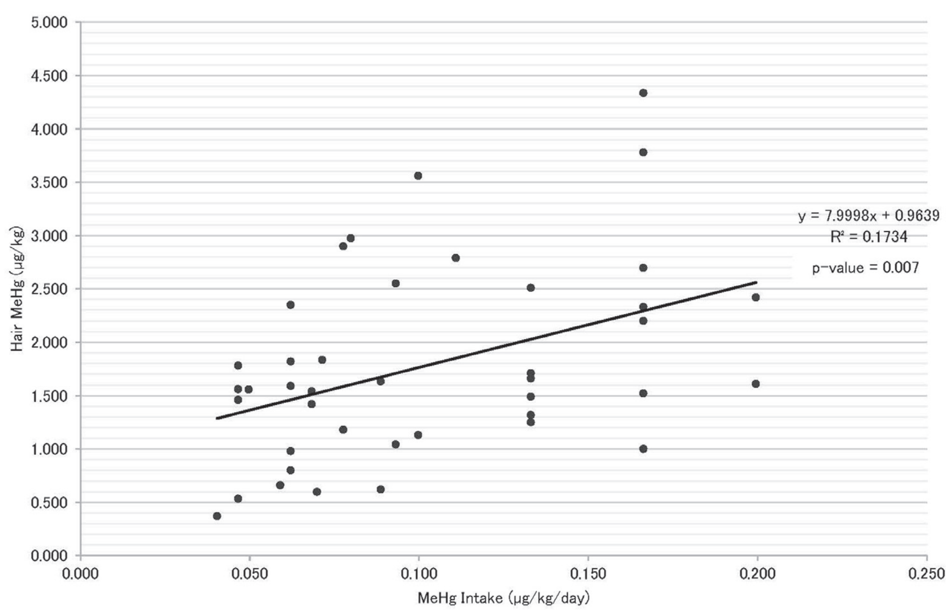

Fig. 3. The correlation between MeHg Intake (μg/kg/day) from rice and MeHg concentration (μg/g) in hair.

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Conflict of interest---- The authors declare that there is no conflict of interest.

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