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Assesing The Risks of Mercury Contamination In Terrestrial Systems At Artisanal Buladu Gold Mine In Gorontalo Province, Indonesia

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ABSTRACT

Background: Mercury (Hg) emitted from open burning of amalgam in gold recovery at artisanal small scale gold mine in Buladu Village attributed in Hg contamination in terrestrial systems in surrounding area. **Objective:** to investigate Total mercury (THg) concentrations in dry deposit, surface soil and foodstuffs were investigated as well as the potential environmental and health risks in the area of concern. **Results:** The concentrations of THg in Deme, Buladu and Wubudu Villages in dry deposit were 136, 501 and 326 $\mu g^{-1}m^2$/day, in surface soil were 2184, 4465 and 4597 μgkg^{-1}dw, in meat and seed of cocoa (*Theobroma cacao*) were 975, 1212, 1335 $\mu g\ kg^{-1}$ww and 1189, 2014, 2973 μgkg^{-1}ww, in cocoa (*Cocos nucifera L.*) meat and milk were 1125, 1348, 1581$\mu g\ kg^{-1}$ww, and 235, 405, 477 $\mu g\ kg^{-1}$ww and in corn were 170, 240 and 327 μgkg^{-1}ww in summer season, respectively. Likewise, in rainy season, THg concentrations in surface soil were 1189, 2014 and 2973 μgkg^{-1}dw, in cocoa meat and seed were 454, 867, 728 μgkg^{-1}ww and 453, 528, 459 μgkg^{-1}ww, in coconut meat and milk were 357, 477, 405 kg^{-1}ww and 201, 356, 310 μgkg^{-1}ww and in corn were 124, 310 and 226 μgkg^{-1}ww, respectively. The value of hazard quotient (HQ) for dry deposit, surface soil and shallow well water were at risks with the values of 10, 46 and 1.7, respectively, whereas target hazard quotient (THQ) values of edible part of cocoa, coconut and corn were very low and not at risk with the values of 0.2, 0.2, 0.1, respectively. **Conclusion:** Environmental status of terrestrial systems in Buladu gold mine and vicinity area were at risk while the local foodstuffs were still safe for consumption.

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INTRODUCTION

Gold mine in Buladu was initiated by the Dutch government in the early 18th century during the Dutch East Indies era (1800-1865). In 1990, mine activities were carried out by local people using tromols, the equipment for amalgamation. This mining area have been legally bound by the agreement between local people and government in 2008 and then implemented since 2010. In the amalgam process, Hg is used to recover gold from mineral ore altogether with large gravels, water and some certain kind of leaves. This artisanal gold mine has operated for about 10 - 12 hours per day using a 2-3 shifting system (4 hours per shifting) for one set of tromols (20-35 tromols) [1, 2].

In terrestrial communities, Hg goes directly to the atmosphere and deposite onto the ground which then might be absorbed by the living plant in surrounding the amalgam centre. Another major pathway of Hg absorption into the plant is via stomata of a plant leaf from dry deposit fall out. Some Hg emitted in term of gasses will resuspense from the soil and water and then enter the atmosphere, where it potentially might be transported and redistributed over all the Earth's surface [3]. The purpose of this study was to investigate the THg concentrations in dry deposit, surface soil, foodstuffs and shallow well water. Hg bioaccumulation in foodstuffs including cocoa, coconut, corn, and shallow well water as well as risk assessment at the Buladu gold mine and vicinity areas were also investigated.

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MATERIALS And METHODS

Study area:

Artisanal Buladu gold mine is located in Sumalata District, North Gorontalo Province, Indonesia. It is an active and important gold mine which has been managed in traditional way by local communities. Starting from the 1990s, this region became a traditional gold mining area, using simple mechanization. The new mining holes were created and the former mining pits of Dutch government were also operated by using hoe, crowbar, chisels and hammers [2]. Three stations of communities of Deme, Buladu and Wubudu Villages were selected for sample collection.

THg mass balance:

An assessment of Hg mass balance during amalgamation process was conducted by weighing the total amount of Hg used and Hg recovered after the amalgam was squeezed.

Dry deposition:

In consideration of possible ground contaminant resuspension, dry deposit dish sampling set points were located at the level of 2.5 m above ground. A set of glass containers (27 x 17 x 10 cm^3) was used to collect the deposit fall out of THg atmospheric particulates. The dish containers were filled with deionized water about five cm dept and refilled in every afternoon during three days of collection period. After three days, samples were collected in the HDPE (high density polyethylene) bottle, the containers were rinsed using the deionized water and then kept in the freezer prior to be analyzed.

Surface soil:

Surface soil samples were collected in both summer (September, 2011) and rainy seasons (February, 2012). At surface layer with 10 cm dept in each 25 m^2 sampling plot and analyzed on a basis of dry weight (dw). In soil sample preparation, wood pieces, pebbles, roots, branch, and dust were discarded and soil were sieved through a 2 mm mesh sieve [4], [5].

Foodstuffs; Cocoa, coconut and corn:

Foodstuffs comprising of cocoa (*Theobroma cacao),* coconut (*Cocos nucifera)* and corn (*Zea mays)* were collected in both summer and rainy seasons from nine sub-sampling plots at those three stations (TC1-TC3) in three different villages. Both meat and seed of cocoa were analyzed for THg as well as coconut meat and milk, and corn seed.

Shallow well water:

Shallow well water samples were collected both in summer and rainy seasons at the same locations as did for soils and food stuffs. The stations were located near the elementary schools in Deme Village and Buladu Village and also the junior high school in Wubudu Village.

THg analysis:

All samples were digested for THg by the method used at the Wetland Biochemistry Institute, Lousiana State University [6]. THg concentration was determined by automated cold vapor, CV-AAS (Cold Vapor Atomic Absorption Spectrophotometer; SHIMADZU, Spectr. AA 6200) after $NaBH_4$ (Sodium Borohydride) reduction with detection limit was 0.001 µg L^{-1}.

Laboratory quality control;

All samples were analyzed at the certified Laboratory of Chemistry Analysis (Laboratorium Kesehatan) in Makassar City, Indonesia. In order to obtain an accuracy in procedures of analyses, calibrations were done using seven replicate samples of standard reference material (SRM 1646a estuarine sediment) from the U.S. Department of Commerce, National Institute of Standard and Technology (NIST) Gaithersburg, MD 20899 and three replicates of blank. The method detection limit (MDL) with seven reagent blanks was calculated and used as a tool for verification. All samples were analysed in three replicates at the 95 % of confidence level.

Potential environmental risks:

The potential environmental risk was determined using a quantitative screening hazard quotient (HQ). The ratio of the exposure estimated to the effect concentration considered representing a safe environmental concentration or screening benchmark is shown in the following formulation.

HQ = EEC/Screening Benchmark (1)

EEC = Estimated (maximum) environmental contaminant concentration at the site (how much contaminant in soil and dry deposit. (e.g. mg contaminant / kg soil). *Screening Benchmark* = Generally a no-adverse effects level concentration (NOAEL); if the contaminant concentration is below this level, the contaminant is not likely to cause adverse effect. However, If the HQ value is > 1 then it indicates the state of risks to the environment [7],[8], HQ < 0.1, no hazard exists; HQ 0.1-1.0, hazard is low; HQ 1.1-10 hazard is moderate and HQ > 10, hazard is high.

Potential health risks:
The provisional tolerable weekly intake (PTWI):

The PTWI was used to evaluate the health concerns of foodstuffs and consumption in study area. The PTWI guideline recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) shows appropriate safe exposure levels, which were used to estimate the amount of contaminants ingested over a lifetime without appreciable risks [9]. In this case, PTWI guidelines for THg recommended at a level of 5 µg/kg bw [10] was used. The estimated weekly intakes (EWI) were calculated using the equation below:

$$EWI = (C_{Hg} \times Cons_R)/BW \quad (2)$$

where; EWI is estimated weekly intakes; C_{Hg} is Hg concentration in contaminated biota; (coconut, cocoa and corn); $Cons_R$ is weekly consumption of food stuffs in local community; gram per week and BW is human body weight (base on 70 kg)

Target Hazard Quotient (THQ):

Consumption of Hg containing food has been identified as a health risk. The U.S. Environmental Protection Agency (US EPA) and the National Academy of Sciences recommend keeping this corresponds to a reference dose (RfD) of THg do not greater than 5.0×10^{-4} µgg^{-1}/day. Non-cancer risk assessment is typically conducted to estimate the potential health risks of pollutants using the THQ, it is a ratio of the estimated dose of a contaminant to the dose level below which there will not be any appreciable risk. If the value of THQ is less than unity, it is assumed to be safe for risk of non-carcinogenic effects. The method is available in US EPA Region III Risk Based Concentration Table (US EPA, 2000).

$$THQ = \frac{EF \times ED \times FIR \times C}{RFD \times BW \times AT} \times 10^{-3} \quad (3)$$

where; THQ is target hazard quotient ; EF is exposure frequency (365 days/year) ; ED is the exposure duration (70 years); FIR is food ingestion rate ; C is metal concentration ; RFD is oral reference dose (Hg = 5.0×10^{-4} µg g^{-1}/day) ; BW is average body weight (70 kg) and AT is averaging exposure time for non-carcinogens (365 days/year x ED)

RESULTS AND DISCUSSION

Quality control for laboratory:

The calculated method detection limit (MDL) in this experiment was of 0.57 µg Hg/L. The certified SRM 1646a value is 0.04 µg g^{-1} and measured value in this research was 0.0389 ± 0.0078 (µg/g dw) with recovery percentage of 97.25.

THg Mass Balance

During amalgamation processing in 20 tromols, the yield of unpurified gold was around 36 g, resulting in 1.8 g gold produced in 1 tromol. After squeezing and burning, the Hg lost was about 2.5 g/tromol, or 2.5 g of Hg was consumed for 1.8 g gold produced or rounded up to 1.3 g Hg per 1 g of produced gold.

THg in dry deposition:

The study in Thailand at Phanompha artisanal gold mine, Phichit Province, suggested that the high elevated atmospheric Hg deposition could be absorbed by leaf via stomata of foodstuffs growing nearby the point source of open burning. Among the THg concentrations of soil, dry deposite and foodstuffs, the correlation was found significantly only in THg in dry deposite and foodstuffs and consistent with the study in Phichit Province, Thailand [11].

THg in surface soil:

The magnitude of THg at three stations in Deme Village were ranged from 2851 to 6492µg /kg dw, in Buladu Village ranged from 1577 to 2216 µg /kg dw and in Wubudu Village from 14.784 to 22.291 µg /kg dw,

respectively. The highest value was obtained from Wubudu areas where some point sources of mercury releases existed. Those THg values were about 10 times higher than those found in the other two villages. There was no definite variation in term of distance from the main point source of mining area with regard to the THg concentration at surface soil in these areas. A relevant study in Bibiani-Anwiaso-Bekwai District, a gold mining community of the Western Region of Ghana, revealed that THg of surface soils samples were ranged from 0.067 to 0.876 mg/kg. The THg of soil at the depths of 20, 40 and 60 cm were varied from 0.1 to 1.1, 0.04 to 4.04 and 0.19 to 5.0 mg/kg, respectively [12]. It can be concluded that all soil samples from those sites exceeded the guidelines accepted by the Canadian Soil Quality Guideline for agricultural soils (6.6 mg/kg) and the UK soil guideline value for inorganic Hg for allotments (8 mg/kg) [13].

There was no significant different among the soil characteristics in community except for CEC (Table 1). In term of particle size (sand, silt and clay) in those three villages the difference of percentages was small with 28.69 %- 31.92 % were sand, 54.82 %-57.54 % were silt, and 13.07 %- 13.87 % were clay, respectively in summer. Category of the soil texture of those three stations was silt loam and clay loam based on the soil triangle determination (Ref.).

In addition, the analysis of soi particle sizes in rainy season were found in the range of 26.40 % - 39.90 % for sand, 23.78 % - 53.27 % for silt, and 18.63 %- 37.77 % for clay, respectively. The relationship between THg concentrations and OM content was linear and the higher OM content, the higher THg concentration.

Table 1: Soil characterizations in community areas (September, 2011 and February, 2012)

Stations	pH (pH)		CEC (cmol/kg)		OM (%)		Particle Size Analysis. Sand (%)		Silt (%)		Clay (%)		Soil Texture	
	SS	RS	SS	RS	SS	RS	SS	RS	SS	RS	SS	RS	SS	RS
TC 1	7.3 8.4	7.0 7.1	9.7 15.8	10.3 11.5	0.6 0.5	0.5 0.6	28.7 31.9	26.4 28.7	54.8 57.5	33.5 38.6	13.1 13.9	33.7 37.8	Silt loam	Clay loam
TC 2	6.9 7.6	6.6 7.2	12.1 15.3	6.7 8.0	0.8 1.9	0.8 1.8	29.3 30.8	38.8 39.9	55.8 57.0	23.8 24.8	13.4 13.9	36.3 36.8	Silt loam	Clay loam
TC 3	6.7 6.8	6.1 6.2	17.1 21.0	16.1 17.1	2.4 4.3	2.3 3.7	29.2 29.5	27.7 28.1	57.1 57.5	53.1 53.3	13.3 13.4	18.6 19.2	Silt loam	Silt loam

SS = Summer season RS = Rainy season
TC1= surface soil in Deme TC2= surface soil in Buladu TC3= surface soil in Wubudu

THg in foodstuffs:

THg concentrations in cocoa meat and seed were in the range of 872 - 1043 µg kg^{-1}ww and 546 – 753 µg kg^{-1}ww, coconut meat and milk were in the range of 225 - 786 µg kg^{-1}ww and 18.68 - 23.51 µg L^{-1}ww and in corn were in the range of 11.47 – 18.57 µg kg^{-1}ww in summer, respectively. In TC3, THg in cocoa meat was the highest one and there was no significantly correlation of the THg concentration in surface soil and the dry deposit. In addition, THg concentrations in coconut and corn varied within the sample sites. It was obvious that THg at TC2 in Buladu Village that closed to pollution sources (ore amalgam processing) was comparatively higher than those in the other sites (Figure. 2). The highest THg concentrations in cocoa meat that was found in TC2 and TC3 which located closer to the private amalgamation process. Hg absorbed and affected plants via two main pathways; through the root system, and via dry arial deposition through plant leaf. In addition Hg entering the soil can go to a wide variety of species changes as a result of the prevailing edaphic conditions controlling bioavailability (Ref.). Also, Hg have a great environmental mobility in rainy season as a result of intense precipitations events when a large number of secondary soluble minerals exist [14]. This factor may lead to a variety Hg content in a certain station.

THg in shallow well water:

THg in shallow well water from those three villages were found in the range of 1.7–2.0 µg L^{-1} in summer season, while those in rainy season were higher and ranged from 2.4 to 3.4 µg L^{-1} and exceeded the safe guideline for fresh drinking water (Table 2).

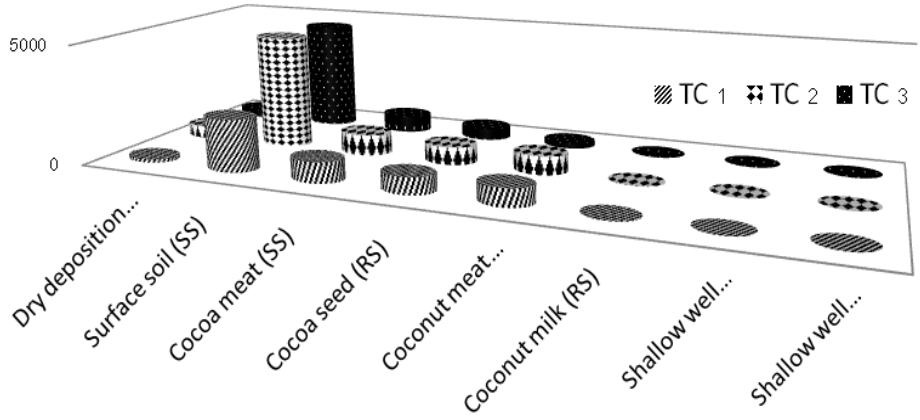
Table 2: THg of dry deposition and shallow well water (September, 2011 and February 2012)

Station	Locations	Dry deposit (µg L^{-1} m^3) SS	Shallow well water (µg L^{-1}) SS	Shallow well water (µg L^{-1}) RS
TC 1	Deme Elementary school	136	1.9	2.7
TC 2	Buladu Elementary school	501	1.7	2.4
TC 3	Wubudu Junior High School	326	2.0	3.4
	Guideline	50 (µg L^{-1} m^3)	2.0 (µg L^{-1})	

SS = Summer Season RS = Rainy Season

THg in environmental compartments of terrestrial systems:

THg concentrations of dry deposition, surface soil, foodstuffs and shallow well water in three villages in summer is presented below;



TC1= Deme Village; TC2 = Buladu Village ; TC3 = Wubudu Village
SS = summer season RS = rainy season

Fig. 1: THg of dry deposition, surface soil, cocoa, coconut and shallow well water

Potential Environmental Risks:

In general, exposure via drinking water and food ingestion have been considered to be major sources of exposure to potentially toxic elements such as Hg.

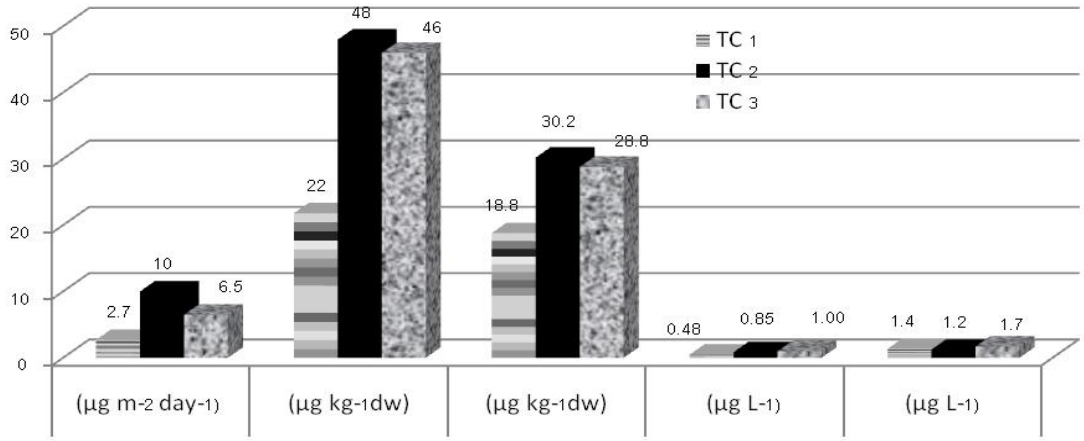


Fig. 2: Potential environmental risks evaluation Hazard Quotient (HQ) for dry deposition in summer season and surface soil in summer and rainy seasons.

Development of risk assessment methodologies for contaminated sites and gold mine working has highlighted the importance of the inadvertent and deliberate ingestion of soils and dusts [15], [16]. The HQ values obtained from this study has given a significant contribution of the methodology to provide information of relationship between surface soil and dry deposition due to Hg contamination in seasonal period.

Potential Health Risks :

Estimated Weekly Intake (EWI):

In the case of Hg, the PTWI guideline for THg recommended at a level of 5 µg/kg bw was used [10], [17]. The EWI of Hg depends on the Hg concentrations in food and the daily food consumption. In addition, the body weight of the human can influence the tolerance level of pollutants.

Table 4: EWI of Hg consumption of cocoa, coconut and corn (September, 2011 - February, 2012)

Station	Station description	EWI of Hg (µg kg^{-1}dw)									
		Cocoa *(Theobroma cacao L.)*				Coconut *(Cocos nucifera L.)*				Corn *(Zea mays L.)*	
		Meat		Seed		Meat		Milk		Grain	
		SS	RS	SS	RS	SS	RS	SS	RS	SS	RS
TC1	Deme School	2.93	1.36	4.76	2.27	6.74	1.79	0.71	0.61	0.85	0.62
TC2	Buladu School	3.64	2.60	8.72	2.64	7.91	3.19	1.43	1.07	1.64	1.55
TC3	Wubudu School	4.01	2.18	6.71	2.30	5.63	2.38	1.22	0.93	1.20	1.13
Standard		PTWI 5.0 µg/kg bw for cocoa, coconut and corn									

SS = Summer season ; RS = Rainy season

The calculation was assumed that the local populations consume cocoa, coconut and corn from local area and the EWI that were calculated from the equation was based on Hg levels from those foodstuffs samples. In Indonesia, cocoa, coconut and corn were the agricultural foods for daily consumption besides rice as the main food. The Indonesian adult has an average daily intake of 40 g cocoa meat per day, 50 g cocoa seed per day, 50 g coconut meat per day, 30 g coconut milk per day and 100 g corn per day. The comparison EWI of Hg through consumption of cocoa, coconut and corn in dry and rainy season and the PTWI are shown in Table 4.

Base on the measurement of the EWI of cocoa meat, corn meat, coconut meat and coconut milk consumption per week for body weight of 70 kg, the results showed that the highest EWI value for Hg were 3.13, 3.76, 3.74, 0.07 and 0.09 µg/kg bw in summer season, respectively. Likewise, in rainy season they were 0.73, 1.56, 1.39, 0.05, and 0.09 µg/kg bw, respectively. Of those values gained in the three different sites near schools, none of the EWI exceeded the PTWI (>5 µg/kg bw) and safe for consumption. Foodstuffs consumers in the study areas might not be prone to be risk because the contamination was quite low.

Calculated EWI of THg through foodstuffs consumption in summer season were closed to the values of P TWI in rainy season .In case people living in the surrounding area who consumed the cocoa, coconut and corn cultivated in the study area for only 7 consecutive days, they were not at risk. However, There might be posed to the risks for ones who consume coconut meat from TC 2 in both summer season (EWI = 3.13) and rainy season (3.56 and 3.76 µg/kg bw). In conclusions, the EWI of Hg for those food stuffs were lower than PTWI and revealed that the agricultural foodstuffs cultivated in this area were safe to consume and were not at risks.

Target Hazard Quotient (THQ):

The THQ for Hg caused by consumption of cocoa, coconut and corn are listed in Table **5**. The calculated THQ values through the consumptions of those foodstuffs were less than one and presented the health risk associated with Hg exposure was insignificant.

Table 5: THQ through consumption of foodstuffs (September, 2011 and February, 2012)

Station	Station description	THQ									
		Cocoa *(Theobroma cacao L.)*				Coconut *(Cocos nucifera L.)*				Corn *(Zea mays L.)*	
		Meat		Seed		Meat		Milk		Meat	
		SS	RS	SS	RS	SS	RS	SS	RS	SS	RS
TC 1	Deme school	0.11	0.05	1.36	0.65	0.19	0.05	0.02	0.02	0.05	0.04
TC 2	Buladu school	0.14	0.10	2.49	0.75	0.23	0.09	0.04	0.02	0.09	0.09
TC 3	Wubudu school	0.15	0.08	1.92	0.66	0.16	0.07	0.03	0.03	0.06	0.06
Standard		HQ > 1 = at risks									

SS = summer season RS = rainy season

From this study, all villages of Wubudu, Buladu and Deme are located close to the point source where ore amalgam processing is actively delivered almost every day. As a result, atmospheric Hg dust deposition have a high potential to fall onto these areas which then be absorbed by the plants [18]. Hg existence in vegetation mostly originates from several mechanisms, including the uptake from the atmosphere, atmospheric deposition to foliage and uptake from plant roots [19]. Heavy metals are components that always exist and accumulate in soils, and they are absorbed by plants only in ionic forms. In some disturbed sites such as ex-mining land; it has been reported that some areas have a large amount of heavy metals [20]. The formation of heavy metal ions can

be due to the lowering of soil pH, and/or, an excessive introduction of free heavy metal ions from contaminated sources such as sewage, polluted water and fertilizer.

Although some studies and statements says that, inorganic mercury was not considered a major source of effects in the soil compartment because it is bound to the soil particles and is not very bioavailable to plants or organisms. In this study, the uptake of gaseous elemental mercury through leaves is much more efficient than the uptake of soil mercury (Hg (II)) in roots, and the main exposure of plants may therefore be through the air.

Conclusions:

Hg contamination in Buladu gold mine and the vicinity areas were highly elevated. In the light of the spatial distribution characteristics of THg in soil, there were two Hg sources: tailing excess and disposed to the ground and the Hg emission from the open burning of amalgam. Most of the findings in this research have high elevated THg in terrestrial foodstuff in the community and adjacent of the mining area. All of those foodstuffs are consumed by people and some are sold out to the other provinces. Another finding is that, the elevated THg concentration in the distance vicinity areas in foodstuffs and surface soil merely generated by the Hg emission as a result of atmospheric fall out that occur continuously. Fortunately, the results obtained from health risks assessment revealed that people who consumed those foodstuffs cultivated in study area were not at risk. However, there might be potential health risks with respect of the continual long period of Hg contaminated foodstuffs consumption.

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